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Original Research

Ex-ante life cycle evaluation of spent lithium-ion battery recovery: Modeling of complex environmental and economic impacts

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ABSTRACT

The recycling of lithium-ion batteries (LIBs) is essential for promoting the closed-loop sustainable development of the LIB industry. However, progress in LIB recycling technologies is slow. There are significant gaps between academic research and industrial application, which hinder the industrialization of new technologies and the improvement of existing ones. Here we show a universal model for spent LIB-lithium recycling (*SliRec*) to evaluate the applicability and upgrading potential across various recycling technologies. Instead of modeling the entire recycling process, we focus on partial processes to enable a comparative analysis of environmental and economic impacts. We find a strong correlation between lithium concentration (LC) and the advancement of recycling technologies, where higher LC is associated with a reduced carbon footprint and increased economic benefits. The implementation of high-level recycling technology can result in an 85.91% reduction in carbon footprint and a 5.97-fold increase in economic returns. Additionally, we explore the effects of technological interventions through scenario analysis, demonstrating that while low-level recycling technology faces more substantial challenges in upgrading, it holds greater potential for reducing carbon emissions ($-2.38 \text{ kg CO}_2\text{-eq mol}^{-1}$) and enhancing economic benefits ($\text{CNY } 11.04 \text{ mol}^{-1}$). Our findings emphasize the significance of process modeling in evaluating the quality of spent LIB recycling technologies, and can provide comparative information for the application of emerging technologies or the upgrade of existing ones.

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1. Introduction

The pursuit of low-carbon development is driving an optimization of the energy structure, pushing society toward a more sustainable future. The rising proportion of commercial renewable energy in the energy mix has substantially promoted the development of lithium-ion batteries (LIBs) [1–3] through strategies such as the electrification of vehicles [4,5], the expansion of wind and solar energy capacities [6–8], and the popularization of energy storage technologies [9,10]. However, the continued exploitation of rare resources from the earth, such as lithium, cobalt, and nickel, limits the sustainable development of LIBs, making these resources

another potentially unsustainable commodity, much like fossil fuels [11,12]. Consequently, the recovery of spent LIBs has become a critical component in promoting the closed-loop sustainable development of these batteries [13].

Despite significant efforts and billions of dollars invested in developing efficient resource-recycling technologies, many attempts have failed to achieve or sustain the desired outcomes. As a result, many emerging technologies, such as selective leaching [14,15], novel solvents [16], selective separation [17,18], electric redox [19,20], and other methods [21–23], have struggled to reach industrialization. Therefore, accelerating the application of advanced LIB recycling technology has become increasingly urgent.

Currently, the challenges in applying advanced LIB recycling technologies lie in two areas: (1) the lack of an ex-ante assessment for emerging technologies to provide a judgment for industrial application; and (2) the absence of a theoretical basis for

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established technologies to guide their upgrading direction. Conventional life cycle assessment (LCA) methodologies often treat unit processes in both established and emerging technologies as “black boxes,” focusing on fixed inputs and outputs of energy and materials [24]. This paradigm neglects the intricate influences of process design, operational conditions, and other process variables on energy and material flows, particularly in chemical engineering [25–27]. Such simplifications introduce significant uncertainties in LCA outcomes. While these uncertainties can be partially evaluated through sensitivity analyses and Monte Carlo simulations [28,29], the lack of detailed quantitative models of process mechanisms restricts the ability to predict and optimize environmental footprints by adjusting process variables.

Research suggests integrating detailed process modeling within the product system framework to address the challenges associated with insufficient life cycle inventory (LCI) data for emerging technologies [30]. For instance, the ex-ante LCA approach used by Tecchio et al. [31] emphasizes predicting environmental impacts using theoretical models and data from pilot-scale experiments without full-scale industrial data. However, this approach does not account for the variability in energy consumption and material flows.

In addition, the difficulty of establishing an evaluation method for spent LIB recycling is mainly due to differences in the types of LIBs and recycling technologies. For example, the technology of using strong acids and reducing agents is suitable for the recovery of $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ (NCM) and LiFePO_4 (LFP) [32]. However, due to the scarcity of cobalt and nickel, recycling NCM can yield greater environmental and economic benefits than recycling LFP [33,34]. As a result, differences in recovered products can lead to opposite conclusions when evaluating the same technology [34,35]. Moreover, differences in recycling technologies, such as chemicals, equipment, and routes, result in significant evaluation errors [36,37]. Therefore, to establish a universal and reliable evaluation method, it is necessary to classify recycling technologies and explore their commonalities. In terms of LIB type, the commonality among LIBs, including LiCoO_2 (LCO), NCM, and LFP, is the lithium component. Regarding technology type, the commonality among recycling processes is lithium recovery [38]. In other words, all recycling technologies for spent LIB include the lithium recycling process (LRP), which could be regarded as their commonality.

Based on the aforementioned methodology review, this study advocates the adoption of mechanism-based process modeling. By simulating the influence of process variables on the inventory data of LIB recycling processes, more reliable data can be obtained. Utilizing tools such as Aspen Plus for simulation provides a robust basis for understanding the chemical processes involved [39–41]. This modeling approach enables a quantifiable analysis of how process variables influence the environmental and economic benefits of LIB recycling, enhancing the accuracy and applicability of LCA in guiding sustainable technology development.

Here, we establish an evaluation model for spent LIB recycling technologies based on the abovementioned commonalities. The correlation between LRP and the overall recycling process is revealed by analyzing different LIB types and recycling technologies, focusing on environmental impact and economic benefits. Based on this, we propose a standard model, the spent LIB-lithium recycling (*SliRec*) model, which includes factors such as chemical, product, and energy consumption, to perform an ex-ante assessment of the application potential of emerging technologies. Furthermore, the effects of intervention strategies are investigated. Specifically, models of the environmental and economic costs of the interventions are established. We set up many scenarios to investigate changes in the net environmental impact and economic benefit of recycling technologies after implementing the

intervention. The results reveal the upgrading challenges and potential of recycling technology, providing a theoretical basis for improving established technologies.

2. Methods

2.1. Modeling analysis of spent LIB recycling technologies

We developed an integrated *SliRec* to rigorously evaluate the industrial feasibility of emerging recycling technologies. This model meticulously examined the influences of three categories, chemicals, products, and energy, on essential applicability indexes such as carbon footprint and economic return. Furthermore, we used the model to investigate the complexities of technological upgrades and revealed the potential of various recycling technologies through strategic interventions. A detailed description of the modeling process was given in the Supplementary Materials.

2.1.1. Baseline model

We first established a baseline scenario for precipitating lithium from the liquor (Supplementary Material Section 1), charting the relationship between the recycling rate of lithium (RRL) and lithium concentration (LC). This correlation was utilized to translate applicability indexes of emerging technologies (chemical input, product output, and energy input) into corresponding LC values (Supplementary Material Section 2). To do this, we first calculated the mass flows of different indexes (X_a – X_d , Y_e , Y_f) following the base setting. Then, their environmental impacts and economic benefits were calculated by combining the parameters of carbon footprint (k_a – k_f) and price (k'_a – k'_f). More details could be found in Supplementary Materials.

2.1.2. Quantification analysis

Given the lack of related studies on the applicability of different recycling technologies, we applied LRP as a unifying framework across different recycling technologies for the first time. We first quantified the applicability indexes using a balance equation (net balance = output – input) grounded in the baseline models. The details are shown in Section 3.1 in Supplementary Material. Next, we estimated the carbon footprint and economic benefits of LRP by adjusting LC (x) or its RRL (x_β), which served as an indicator of the development level of the technology (see Section 3.2 in Supplementary Material).

2.1.3. Scenario development

We first formulated a baseline scenario to represent the upgrade potential of different recycling technologies (high and low levels) by setting various RRL increments (n) (Section 3.3 in Supplementary Material). Here, the concentration process (CP) functioned as an intervention. Since LCs represented various levels of recycling technology in our model, applying CP could reflect the technical adjustments. In addition, CP represented the minimum level of intervention. If the planned intervention led to a better outcome than CP, it could be inferred that the technical upgrade was significant. Conversely, if the planned intervention did not result in a better outcome, it could be concluded that the corresponding technical upgrade was unnecessary.

We then explored eight scenarios: the first four assess technical levels by fixing $x = 0.25, 1, 4,$ and 10 mol L^{-1} , while the other four evaluated upgrade potential by fixing $n = 1, 10, 30,$ and 80% . Next, we estimated the carbon footprint and economic benefit of LRP with different technical levels and proposed strategies for improving recycling technologies. It was important to note that this study aimed to explore the potential rather than predict the future.

Due to data limitations, the model had certain constraints and might be improved in several aspects, as discussed in Section 4 of Supplementary Material.

2.2. Evaluation of different recycling technologies for spent LIBs

This LCA methodology aligned with the guidelines set by ISO 14040 (ISO 14040 2006; ISO 14044 2006), including goal and scope definition, LCI analysis, life cycle environmental impact assessment, and interpretation of results.

2.2.1. Goal and scope definition

This study aimed to comprehensively assess the carbon footprint of various recycling technologies for spent LIBs. The functional unit was defined as the carbon footprint per kilogram of recycled cathode material. The system boundaries began with the cathode material obtained from spent LIBs, which was then processed through different recycling technologies [42]. These processes included the leaching of raw metals, separation of metals, recovery of metallic lithium, and waste treatment stages (outlined in Fig. 1). This assessment also accounted for indirect emissions from the recycling processes, including releasing gases and other by-products during chemical treatments and reactions involved in LIB recycling, such as leaching and separation, with indirect emissions primarily arising from electricity consumption. This study compared three reported recycling technologies with detailed technical parameters in Table S1 (Supplementary Material). Since the focus was on the metal recovery process, common processes such as transportation, dismantling, and preprocessing were not included.

2.2.2. LCI analysis

The inventory analysis phase involved collecting relevant foreground and background data associated with the entire product system of spent LIB recycling. Foreground data included quantities of raw material consumption, energy usage, recycled products, and waste generation. The material flow input-output of unit processes was modeled based on experiments with three reported spent LIB recycling technologies (Supplementary Material Fig. S1). Energy consumption for each unit process was simulated using Aspen Plus (Supplementary Material Fig. S2). Environmental impacts within the upstream and downstream supply chain were also accounted for. Background datasets in this study were sourced from the Ecoinvent database, which provided upstream production data for the required raw materials. The detailed LCI analysis method was presented in Table S2 (Supplementary Material), with specific information on the referenced background datasets listed in Table S3 (Supplementary Material).

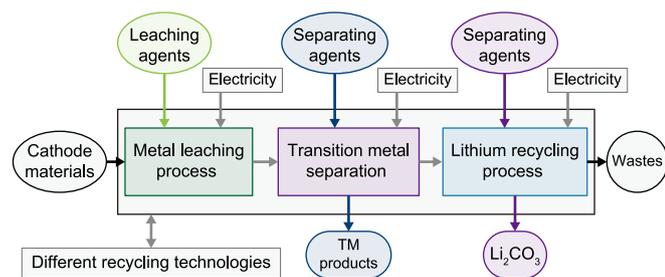


Fig. 1. System boundary diagram for different recycling technologies of spent lithium-ion battery (LIB). The functional unit for this analysis is defined as “1 kg of cathode materials”. TM: transition metals.

2.2.3. Evaluation of carbon footprint and economic benefit

Based on the established *SliRec* model and LCI analysis, the carbon footprint was quantified using the IPCC GWP100a methodology. Energy inputs, raw material consumption, and waste treatment were positive environmental contributions. By contrast, recycled lithium (Li) and cobalt (Co), substituting virgin material production, were accounted for as negative contributions. The net balance of the carbon footprint indicated whether recycling LIB materials resulted in carbon emission reduction benefits (negative value) compared to primary production from natural resources. Combined with price parameters, a life cycle cost (LCC) analysis was conducted to assess the economic benefits. Detailed information was available in the Supplementary Material. A sensitivity analysis was conducted by applying a 1% increase to each foreground or background inventory data item, as shown in Table S9 (Supplementary Material).

3. Results and discussion

3.1. Variable analysis: is LC related to the development level of spent LIB recycling technology?

The goal of recycling spent LIB is to recover transition metals (TMs) and Li from the cathode, accomplished through different recycling technologies (Fig. 2a). LRP is a common factor across different recycling technologies for spent LIB. We further aimed to investigate the correlation between LRP and the overall recycling process from environmental and economic perspectives. Data, including the carbon footprint and economic benefit, were calculated and applied for correlation analysis (see Supplementary Material). The total recycling process of spent LCO was referred to as total recovery of type 1 (TR-1). Given the high economic value and carbon reduction potential of Co-containing products, we excluded Co-containing products from the total recycling process to represent the recovery of other less expensive LIB chemistries, such as LFP and LiMn_2O_4 (LMO), referred to as the total recovery of type 2 (TR-2).

Fig. 2b presents the relationship between TR-1, TR-2, LRP, and LC carbon footprints. The results indicated that different LCs were obtained by different recycling technologies, such as technology 1 (hydrothermal treatment [43]), technology 2 (reductive leaching [44]), and technology 3 (selective leaching [15]). The obtained LCs showed a clear correlation with the carbon footprint of TR-1 and TR-2; higher LCs led to lower carbon footprints. Thus, the LC of the obtained leachate could represent the development level of recycling technologies. Notably, the carbon footprint of LRP was positively associated with that of TR-1 and TR-2. Hence, LRP could serve as an alternative to the total recycling process in evaluating the environmental impact of recycling technologies. Similarly, the results demonstrated that LRP could be used to analyze the economic benefit of different recycling technologies for spent LIBs (Fig. 2c), replacing TR-1 and TR-2. Specifically, a relatively low profit of CNY 201.71 per kg LCO was obtained under 3.49 mol per L LC (TR-1) due to differences in the type of Co-containing products (Supplementary Material Table S10). This situation was avoided in TR-2, which excluded Co-containing products. In summary, LRP could be used to evaluate the applicability of recycling technologies for spent LIBs instead of TR-1 and TR-2. Specifically, LC was taken as the independent variable representing the development level of recycling technologies, with corresponding dependent variables such as carbon footprint (C) and economic benefit (B) included in the model analysis.

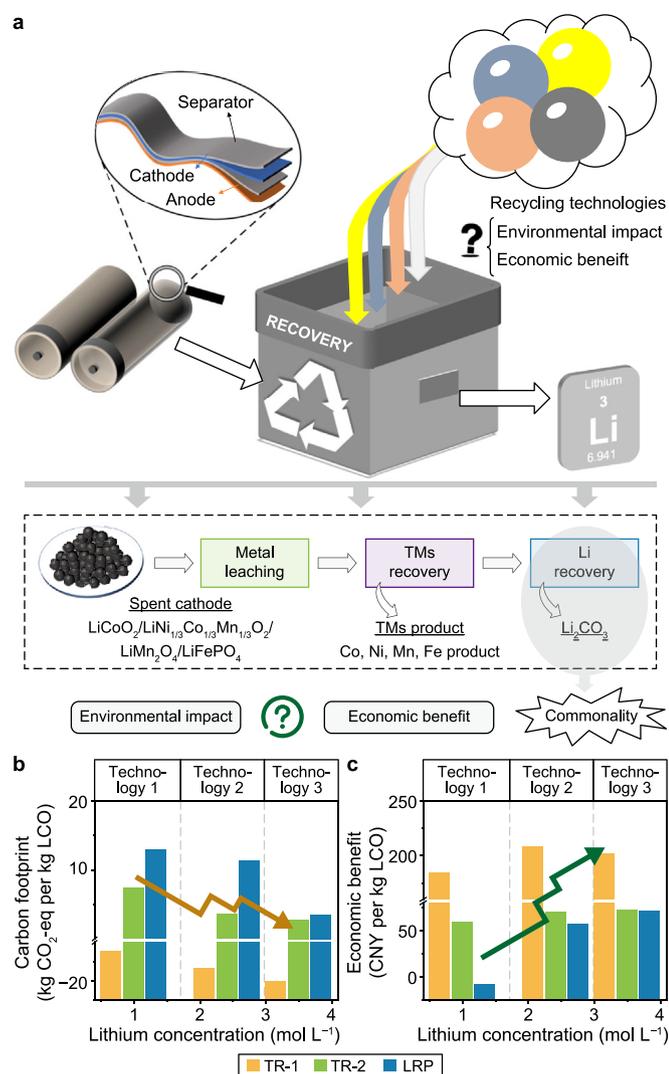


Fig. 2. a. Diagram of breakdowns of the recycling process of spent lithium-ion battery: metal leaching, transition metals (TMs) recovery, and lithium (Li) recovery. b. The relationship between the carbon footprints of total recovery types 1 and 2 (TR-1 and TR-2) and the lithium recycling process (LRP) with varying lithium concentrations (LC). c. The relationship between the economic benefits of TR-1, TR-2, and LRP and the LC. LCO: LiCoO₂.

3.2. Standardized modeling: how do we model LRP to quantitatively analyze spent LIB recovery?

Based on the previous discussions, the LRL was modeled to assess emerging technologies, referred to as the *SliRec* model. The modeling process was simplified and presented in Fig. 3. The relationship between RRL and LC was analyzed and equated. Then, the corresponding material flow, including chemicals (X_a , X_b), products (X_c , X_d , Y_f), and energy (Y_e), was quantified. Finally, the applicability indexes (carbon footprint and economic benefit) were modeled by introducing parameters of carbon footprint (k_a – k_f) and price (k'_a – k'_f).

The scenario for modeling LRP was first established (see Supplementary Material Fig. S3) to quantify the material flow. A detailed description of the basic setup is provided in Section 1 of the Supplementary Material, and the relationship equation between RRL (X_β) and LC (x) was established (Supplementary Material Fig. S4a). The results indicated that LC positively affects the RRL. The

LC should be around 1.29 mol L⁻¹ to achieve 85% RRL, the suggested industrial standard for the comprehensive utilization of waste power batteries in China. Furthermore, baseline models for chemical input, product output, and energy input were established for the *SliRec* model (full details can be found in Section 2 in Supplementary Material). Specifically, the handling capacity of Li-containing leachate was fixed at 1 mol (i.e., $xV = 1$ mol, V represented the volume of Li-containing liquor) to analyze mass flows, and the initial volume was fixed at 1 L (i.e., $V = 1$ L) to analyze energy input and water output. As a result, models for Na₂CO₃ input (X_a), water input (X_b), Li₂CO₃ output (X_c), wastewater output (X_d), energy input (Y_e), and evaporable water output (Y_f) were developed (Supplementary Material Figs. S4b–g).

Based on these baseline models, the *SliRec* models were derived according to the balance equation by introducing parameters of carbon footprint and price (see Supplementary Material Fig. S4h). First, the direct recovery of lithium was modeled to evaluate the applicability of emerging technologies, with LC representing the level of technologies. The corresponding modeling process is detailed in Section 3.1 of the Supplementary Material. The models for carbon footprint (C) and economic benefit (B) are presented in equations (1) and (2).

$$C = V(0.0612x + 0.8901) \quad (1)$$

$$B = V(8.7818x - 2.5720) \quad (2)$$

The sensitivity analysis revealed that the carbon footprint changed by less than 1% when applying a 1% increase in the variate (Supplementary Material Fig. S7a). It was found that wastewater treatment contributed the most to this change, accounting for 78.35%. Relatively, Fig. S7b (Supplementary Material) showed a significant variation in economic benefit under low Li concentration (from 5.07% to 0.48%), while it remained stable under high Li concentration (below 0.20%). The economic benefit was primarily influenced by Li₂CO₃ output, which accounted for 59.79%.

Furthermore, model verification was carried out to validate the constructed models. To avoid deviations caused by Co-containing products, we verified the models using the recycling technologies for less expensive LIB chemistry, where Li-containing products dominated the total recycling process. The actual calculated results of three recycling technologies were used for the verification (Fig. 4). Fig. 4a demonstrates that the predicted carbon footprints closely matched the actual results as the technological level increased. The difference ratio decreased from 26.5% to 3.0%. This is because the increased chemical input did not significantly increase Li₂CO₃ output in low-level technologies. Thus, the input of the actual operation was not enough to generate a slight increase in output. This also explains the difference ratio in predicted economic benefits in Fig. 4b. The predicted economic benefits were similar to the actual results, with a difference ratio of less than 12.0%. It can be concluded that the constructed models are well-suited for predicting recycling technologies for spent LIBs. It should be noted that the predicted value exceeded the actual value. This discrepancy is due to the theoretical Li₂CO₃ output used in the predictive model, which is higher than the actual output. Additionally, an increase in Li₂CO₃ output would lead to an increase in Na₂CO₃ input and wastewater output. As a result, in the prediction of the carbon footprint model, the overall carbon footprint increased because the carbon footprints of Na₂CO₃ input and wastewater output were predominant throughout the process. Conversely, the economic benefit of Li₂CO₃ output predominated in the prediction of economic benefit, ultimately leading to a higher overall economic benefit result.

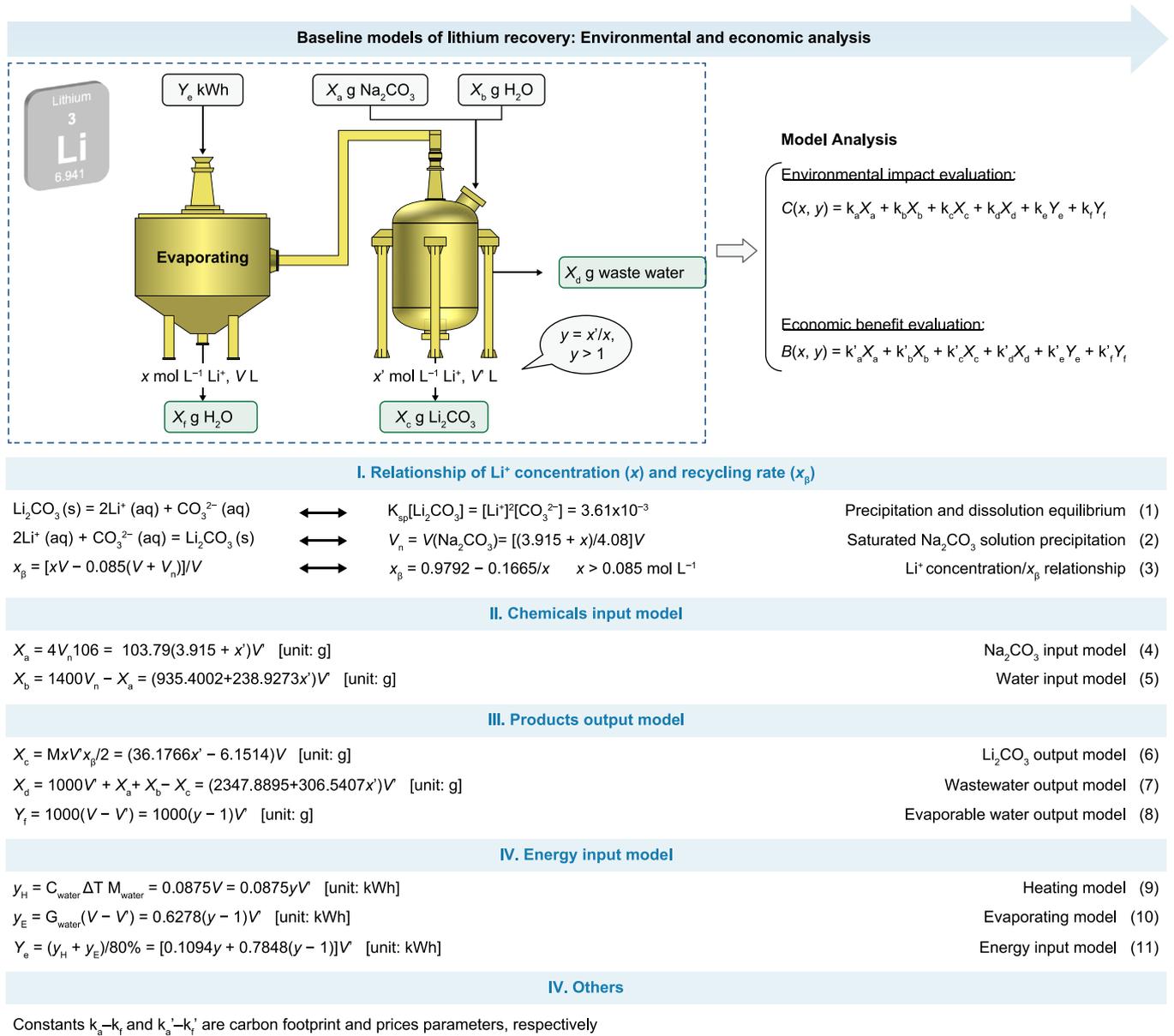


Fig. 3. The mathematical modeling framework for quantifying the inventory data of the lithium recycling process (LRP) for environmental and economic analysis.

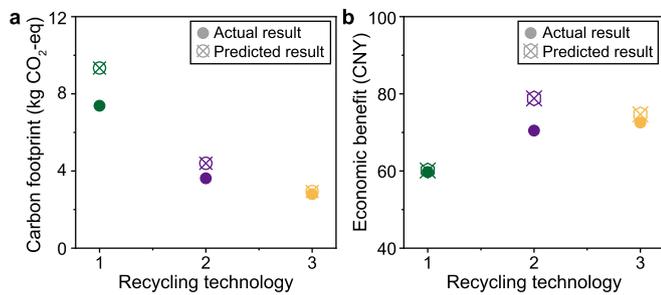


Fig. 4. Verification of the constructed models by comparing the predicted values of assessment indicators with the actual calculated results. **a.** Carbon footprint. **b.** Economic benefits.

3.3. Quantification analysis: is implementing interventions for spent LIB recycling technology worth it?

Based on the discussion above, the standardized *SliRec* model was proposed for analyzing the spent LIB recovery (Fig. 5a). For quantitative comparison, the handling capacity was assumed to be 1 mol (i.e., $xV = 1$ mol). First, it was assumed that no intervention was taken (i.e., $V = V'$). The modeling results without intervention, including carbon footprint (C_0) and economic benefit (B_0), were calculated and presented in Fig. 5b and c. Fig. 5b indicated that LC was negatively correlated with the carbon footprint of LRP. The result showed that the carbon footprint decreased from 3.62 kg CO₂-eq per mol Li to 0.51 kg CO₂-eq per mol Li as LC increased from 0.25 to 2.00 mol L⁻¹, achieving an 85.91% carbon emission reduction. Correspondingly, the economic benefit ranged from CNY -1.51 per mol Li ($x = 0.25$ mol L⁻¹) to CNY 7.50 per mol Li ($x = 2.00$ mol L⁻¹) (Fig. 5c). Notably, the economic benefit was

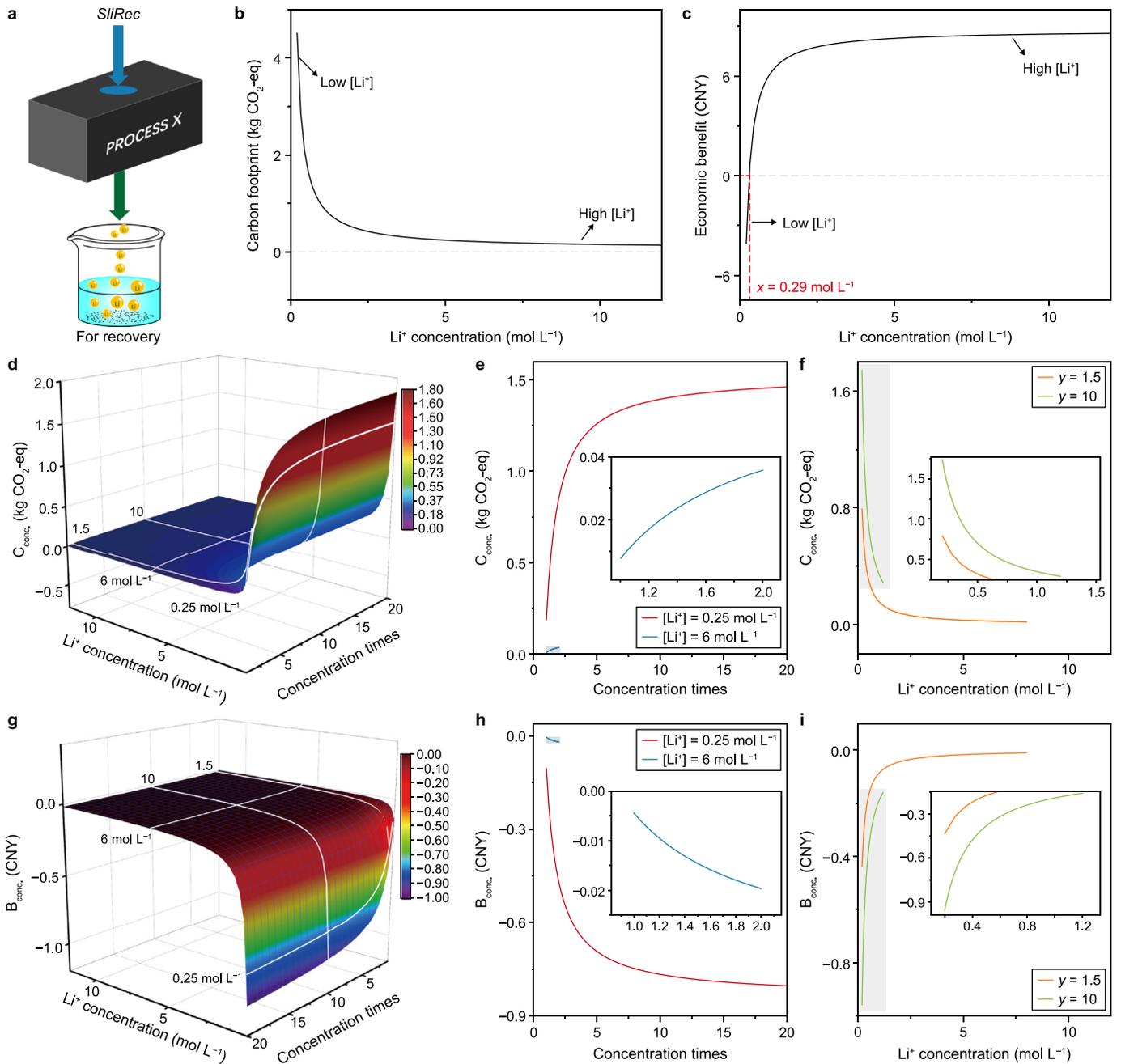


Fig. 5. a, Diagram of the spent lithium-ion battery-lithium recycling (*SliRec*) model. b–c, *SliRec* model including carbon footprint (b) and economic benefit (c). The dotted red line represents the economic equilibrium point. d, Carbon footprints of concentration process (CP) ($C_{conc.}$, kg CO₂-eq). e–f, Carbon footprints of CP with limited conditions: e, lithium concentrations ($[Li^+]$) of 0.25 and 6 mol L⁻¹ represent the low and high technical level, respectively; f, concentration times (y) of 1.5 and 10 represented the weak and strong intervention, respectively. The inserted figures are the enlarged views of the shaded areas. g, Economic benefit of CP ($B_{conc.}$, CNY). h–i, Economic benefit of CP with limited conditions: h, $[Li^+] = 0.25$ and 6 mol L⁻¹; i, $y = 1.5$ and 10. The inserted figures are the enlarged views of the shaded areas.

positive only when LC was over 0.29 mol L⁻¹. It could be concluded that a higher LC leads to a lower carbon footprint and a higher economic benefit. Therefore, high LC is crucial for recovering spent LIBs, especially for low-cost LIB chemistries such as LFP and LMO.

Furthermore, the effects of the intervention were studied, with CP representing the intervention. Specifically, changes in LC (presenting the development level of recycling technologies) due to intervention were realized and controlled by CP. Thus, CP was used to represent interventions taken for recycling technologies. Herein, concentration times (1–20) represented the intervention intensity.

The corresponding carbon footprint ($C_{conc.}$) and economic benefit ($B_{conc.}$) of applied interventions were quantified in Section 3.2 of the Supplementary Material. The model functions for the carbon footprint and economic benefit of CP were calculated and shown in equations (3) and (4).

$$C_{conc.} = \frac{0.3819}{x} - \frac{0.3351}{xy} \tag{3}$$

$$B_{\text{conc.}} = \frac{0.1839}{xy} - \frac{0.2101}{x} \quad (4)$$

where x and y represent the Li concentration (mol L^{-1}) and concentration time, respectively.

The results indicated that the initial LC significantly affected the carbon footprint of interventions (i.e., CP) (Fig. 5d). For comparison, LCs of 0.25 and 6 mol L^{-1} were used to represent different technical levels for analysis (Fig. 5e). Considering that low LC leads to a high carbon footprint and low economic benefit (Fig. 2b and c), an LC of 0.25 mol L^{-1} represents low-level recycling technology (LL-RT), while an LC of 6 mol L^{-1} represents high-level recycling technology (HL-RT). The results showed that for LL-RT, the carbon footprint of intervention sharply increased at the initial stage (from 0.31 kg $\text{CO}_2\text{-eq}$ at $y = 1.1$ to 1.08 kg $\text{CO}_2\text{-eq}$ at $y = 3$) and then plateaued at about 1.44 kg $\text{CO}_2\text{-eq}$ ($y = 15$). In comparison, the intervention carbon footprint of HL-RT remained below 0.05 kg $\text{CO}_2\text{-eq}$. Different y values were set to investigate the intervention space for recycling technologies (Fig. 5f). The results showed that weak intervention ($y = 1.5$) was applied to most recycling technologies, while strong intervention ($y = 10$) was applied to LL-RT. Strong intervention for LL-RT could lead to a carbon footprint exceeding 1.6 kg $\text{CO}_2\text{-eq}$, while the carbon footprint of weak intervention remains below 0.4 kg $\text{CO}_2\text{-eq}$ ($x > 0.4 \text{ mol L}^{-1}$).

The economic benefit of intervention was analyzed and shown in Fig. 5g. The results showed that low LC led to a high economic cost of the intervention. The results indicated that the economic benefit of HL-RT was in a small range of CNY 0 to CNY -0.02 due to its limited intervention space (Fig. 5h). For LL-RT, the intervention cost increased from CNY 0.21 ($y = 1.1$) to CNY 0.81 ($y = 10$) as intervention intensity increased from 1.1 to 10. Fig. 5i presents the economic benefits of interventions with different intensities. The results showed that the intervention cost for LL-RT was higher than for HL-RT, with higher intervention intensity leading to higher costs. To sum up, LL-RT had a greater intervention space than HL-RT; however, its intervention could result in a higher carbon footprint and economic costs. Considering that intervention can increase LC, reduce reagent dosage, and increase product revenue, it is necessary to further analyze the net economic benefit and carbon footprint of spent LIB recovery after the intervention.

3.4. Scenario development: what is the direction of upgrading established technologies?

It was assumed that RRL increased from x_β to $(x_\beta + n)$ after the intervention, the handling capacity of Li-containing liquor set at 1 mol L^{-1} (i.e., $xV = 1 \text{ mol}$). Thus, concentration time (y) could be calculated and presented as equation (5). Based on this, the corresponding carbon footprint ($C_{y,n}$) and economic benefit ($B_{y,n}$) of recycling technologies after the interventions were calculated as shown in equations (6) and (7). Consequently, the net values of carbon footprint (ΔC_n) and economic benefit (ΔB_n) were determined as shown in equations (8) and (9), with the results displayed in Fig. 6a and b, respectively. Detailed information is provided in Section 3.3 of the Supplementary Material.

$$y = \frac{0.9792 - x_\beta}{0.9792 - n - x_\beta} = \frac{0.1665}{0.1665 - nx} \quad (5)$$

$$C_{y,n} = C_{\text{conc.}} + C = \frac{V}{100} [(6.12 - 333.3333n)x + 93.69] \quad (6)$$

$$B_{y,n} = B_{\text{conc.}} + B = V[(8.7818 + 14.3429n)x - 2.5982] \quad (7)$$

$$\Delta C_n = C_{y,n} - C_0 = \frac{1}{100} \left(-333.3333n + \frac{4.68}{x} \right) \quad (8)$$

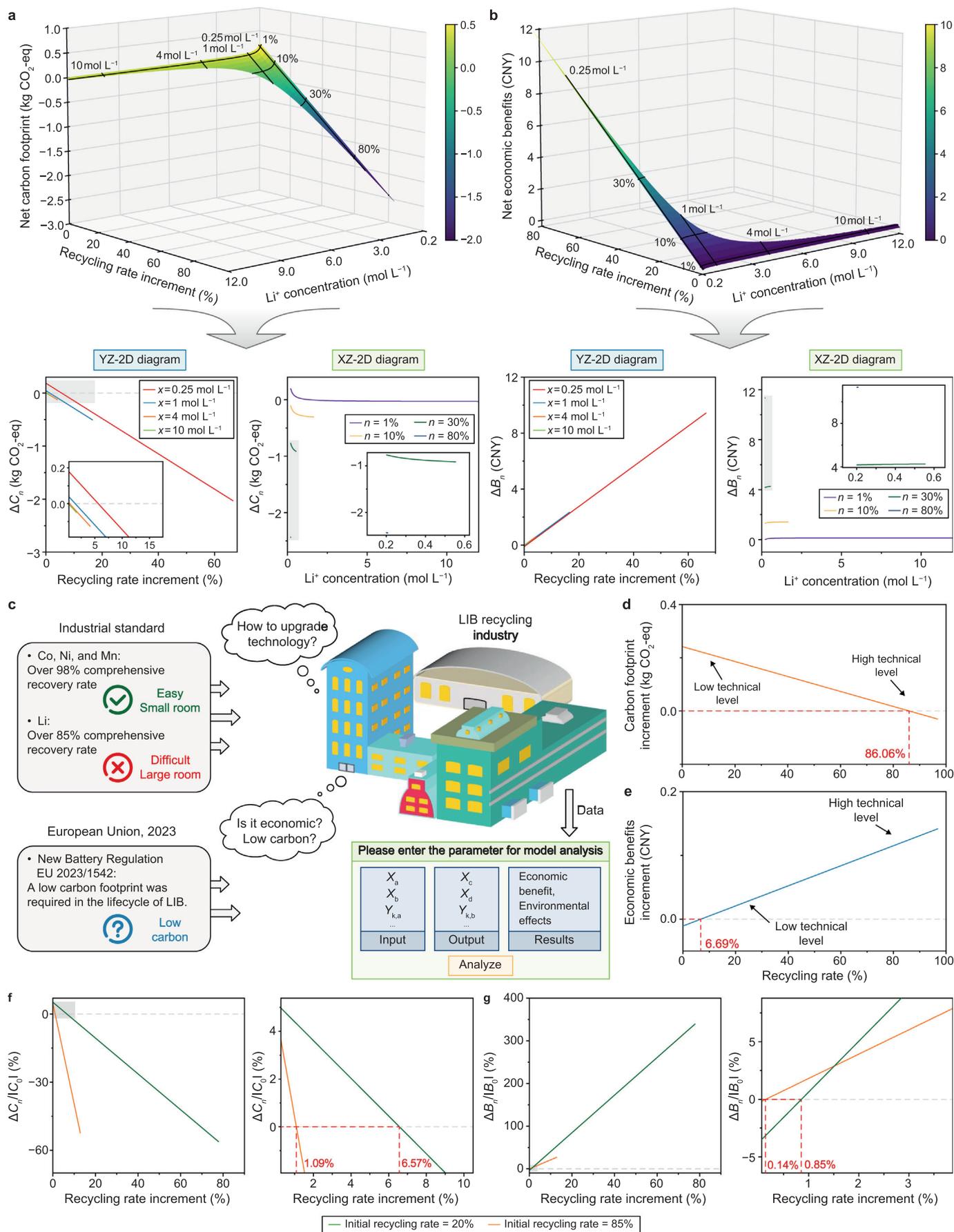
$$\Delta B_n = B_{y,n} - B_0 = 14.3429n - \frac{0.0262}{x} \quad (9)$$

In the equations above, n represents the increment of RRL, $n \leq 0.1665/x$ (%); x represents LC (mol L^{-1}); x_β represents the RRL of Li liquor with $x \text{ mol L}^{-1}$ (%); and y represents the concentration time.

We explored four scenarios regarding technical levels by fixing $x = 0.25, 1, 4,$ and 10 mol L^{-1} (see YZ plane), and four scenarios regarding technical upgrade by fixing $n = 1, 10, 30,$ and 80% (see XZ plane) (Fig. 6a and b). The results showed that changes in the carbon footprint of HL-RTs ($x > 2 \text{ mol L}^{-1}$) were not significant (YZ plane in Fig. 6a). For a given technology, a larger RRL increment resulted in a smaller carbon footprint (XZ plane in Fig. 6a). Additionally, the economic benefits of most interventions were negative, especially for HL-RTs. For a given RRL increment, HL-RT required a higher economic cost than LL-RT (YZ plane in Fig. 6b). The XZ plane of Fig. 6b indicates that strong interventions in LL-RT could enhance economic benefit. For example, for LL-RT (0.25 mol L^{-1}), its economic benefit increased from CNY 0.04 to CNY 11.37 mol^{-1} as the RRL increment increased from 1% to 80%. Interestingly, the economic benefit increment for HL-RT was smaller than that for LL-RT. Additionally, under the same intervention, there was no significant difference in economic benefit increments between LL-RT and HL-RT.

High standards for recycling spent LIB have been introduced in China and Europe (Fig. 6c). China has implemented the Interim Measures for the Management of Industrial Standard Announcement for the Comprehensive Utilization of Waste Power Batteries (2019), which requires comprehensive recovery rates of 98% for Co, Ni, and Mn and 85% for Li [45]. The recovery rate of transition metals has clearly reached technical saturation, close to 98%. By contrast, the recovery rate for lithium is set at only 85%, indicating significant room for improvement. Recently, Europe has introduced a strict regulation (EU-2023/1542) on the environmental friendliness of recycling technologies, requiring an electric passport for batteries [46]. The electric passport must contain detailed life-cycle information that proves low carbon emissions. Therefore, developing economically viable and low-carbon technologies for recycling spent LIBs is significant. Based on our model, the difficulty and potential for upgrading recycling technologies were further discussed.

For a given level of technology, we assumed a 1% increase in the RRL to explore the corresponding changes in carbon footprint and economic benefit. The results showed that the carbon footprint increment ($\Delta C_{y,1\%}$) of LL-RT (20%) was 0.19 kg $\text{CO}_2\text{-eq mol}^{-1}$ with a 1% increase in the RRL, while for HL-RT (85%), it was 3.01×10^{-2} kg $\text{CO}_2\text{-eq mol}^{-1}$ (Fig. 6d). The upgrade of LL-RT faced significant pressure due to its high carbon footprint. Additionally, the increment in economic benefit ($\Delta B_{y,1\%}$) is presented in Fig. 6e. The results showed that a 1% increase in the RRL was always accompanied by increased economic benefits, with a greater economic increment observed for HL-RT. For example, a 1% increase in the RRL of HL-RT (85% RRL) resulted in an economic increment of CNY 0.12 mol^{-1} . In the same case, LL-RT (20% RRL) only achieved an economic increment of CNY $2.08 \times 10^{-2} \text{ mol}^{-1}$. An economic loss was even observed when the technical level was as low as 6.69%. It can be concluded that upgrading LL-RT is more challenging than upgrading HL-RT, and HL-RT can achieve greater economic benefits with



lower carbon emissions. In most cases, both economic benefit and carbon footprint increased with weak intervention (i.e., a 1% increase in RRL). However, a negative carbon footprint increment and positive economic benefit increment can be achieved by increasing intervention intensity (i.e., RRL increment) (XZ-plane in Fig. 6b and XZ-plane in Fig. 6c). Thus, it is possible to achieve both environmental friendliness and economic benefits through proper interventions.

The carbon footprint growth and economic benefit of spent LIB recovery after interventions was investigated (Fig. 6f and g). The results indicate that carbon footprint growth rates gradually decreased with increased RRL increment (Fig. 6f). The carbon footprint increments of LL-RT (i.e., 20% RRL) and HL-RT (i.e., 85% RRL) turned negative when n reached 6.57% and 1.09%, respectively. The minimum carbon footprint growth rates of LL-RT and HL-RT were -56.27% and -52.45% , respectively. Fig. 6g indicates that the maximum economic benefit growth rates of LL-RT and HL-RT were 339.66% and 27.01%, respectively. Furthermore, as presented in Table S11 (Supplementary Material), the carbon footprint reduction potential of LL-RT (i.e., $-2.38 \text{ kg CO}_2\text{-eq mol}^{-1}$) was much higher than that of HL-RT (i.e., $-0.39 \text{ kg CO}_2\text{-eq mol}^{-1}$). The economic benefit increments of LL-RT and HL-RT were CNY 11.04 and CNY 1.83 mol^{-1} , respectively. Therefore, both in environmental and economic aspects, small enterprises with LL-RT have greater potential for technology upgrades than large enterprises with HL-RT.

In summary, large enterprises with HL-RT have advantages over small enterprises with LL-RT in terms of economic and environmental protection. Additionally, it is relatively easy for large enterprises to realize the growth of environmental and economic benefits through technical upgrades. By contrast, small businesses struggle to initiate technical upgrades and need to make more effort to achieve the same results as large businesses. However, overall, small enterprises can achieve greater environmental and economic benefits from technical upgrading than larger enterprises. Therefore, large enterprises must take the lead in technology upgrades and establish higher technical standards in the industry to promote the technical upgrading of small enterprises and ultimately advance the industry.

4. Limitations and outlook

As ex-ante LCA relies on predictive modeling rather than historical data, the availability and accuracy of data for emerging technologies can vary. Assumptions made without empirical data can introduce variability in the results. Additionally, differences in operational conditions, such as temperature, pressure, and feedstock composition, can affect the performance and environmental impact of recycling processes. Conducting sensitivity analysis in the future could help evaluate the impact of key variables on our results. By simulating and understanding the range and likelihood of different outcomes, we can provide more detailed insights into potential impacts.

Furthermore, decarbonizing the electricity supply can significantly reduce the carbon footprint of LIB recycling processes [47,48]. Decarbonizing the electricity sector creates a positive feedback loop, where greener electricity supports further reductions in greenhouse gas emissions. Similarly, grid decarbonization is crucial for enhancing the offsets from energy recovery and recycled materials [49]. To further explore the impact of electricity

decarbonization, we can conduct a scenario analysis to evaluate the carbon footprint and economic benefits of LIB recovery under different power grid decarbonization scenarios. For instance, comparing a scenario with a high penetration of renewable energy in the grid by 2030 to one with slower adoption rates can provide insights into how different decarbonization pathways affect the overall sustainability of LIB recycling. This additional analysis will help stakeholders understand the range of potential outcomes and make more informed decisions regarding the timing and scale of grid decarbonization efforts.

In addition to evaluating the direct economic benefits of LIB recycling, it is essential to consider the impact of carbon pricing mechanisms. Carbon taxes and certified emission reductions can greatly influence the economic viability of recycling technologies. A carbon tax creates a financial incentive for industries to lower their carbon footprint. For LIB recycling, reduced emissions lead to lower costs, making these technologies more economically viable. Our analysis indicates that including carbon taxes in the economic assessment can enhance the competitiveness of recycling technologies, especially those that achieve significant carbon footprint reductions. By incorporating carbon pricing mechanisms, our study highlights the dual benefits of advanced LIB recycling technologies in terms of environmental impact and economic return. This approach provides stakeholders with a comprehensive view of the synergies and trade-offs, promoting the development and industrial use of green and economically viable recycling solutions.

5. Conclusions

Developing efficient resource recycling technologies is crucial for the sustainable development of the LIB industry. Our study advances our understanding of the quantified applicability of spent LIB recycling technologies through modeling analysis and provides guidance for technological upgrades within the industry. According to our established standardized model (*LiRec*), the applicability indexes of different recycling technologies were concretized as carbon footprint and economic benefit for quantitative analysis, offering a comparative assessment for industrialization. The LC of the leachate was positively correlated with the development level of recycling technologies, where a higher LC resulted in a lower carbon footprint and a higher economic benefit. Through scenario development, we examined the upgrading challenges and potential of established technologies, such as LL-RT and HL-RT. It was found that a 6.57% RRL increment in LL-RT (i.e., 20% RRL) could achieve a reduction in carbon footprint, whereas HL-RT (i.e., 85% RRL) required only a 1.09% increment, indicating that LL-RT faces greater challenges in technological upgrades. Nonetheless, LL-RT exhibits significant potential for technological enhancement, with the potential for carbon footprint reduction and economic benefit increments being $2.38 \text{ kg CO}_2\text{-eq mol}^{-1}$ and CNY 11.04 mol^{-1} , respectively.

Our findings address two key dilemmas: (1) how to quantify the applicability indexes of emerging technologies and (2) where the direction of upgrading established technologies lies. This study is the first to use the partial process (LRP) rather than the overall recycling process to evaluate and compare various recycling technologies. Since many emerging technologies have not yet been applied in the industry, the data utilized are at the laboratory level and differ from those of large-scale production. Given these data

Fig. 6. a–b, The net values of carbon footprint (ΔC_n , a) and economic benefit (ΔB_n , b) after interventions. The inserted figures are the enlarged views of the shaded areas. c, Diagram of industrial standards for a technical upgrade of recycling spent lithium-ion battery (LIB). d–e, Carbon footprint (d) and economic benefit (e) of increasing lithium recovery by 1% under different recycling rate of lithium (RRL). The dotted red line highlighted the balance point of the economy or carbon footprint, respectively. f–g, The potential growth rate of carbon footprint (f) and economic benefit (g) under different RRL increments. C_0 and B_0 represent the carbon footprint and economic benefit of recycling spent LIB without intervention, respectively. The dotted red lines in the enlarged views highlight the balance point of the economy or carbon footprint.

constraints, the generalization of findings has its limitations. Future studies can further explore the correlation between LRP and the overall recycling process. The *SliRec* model employed in this study provides new insights into the quantitative evaluation of spent LIB recycling technologies.

CRedit authorship contribution statement

Jiefeng Xiao: Writing - Review & Editing, Writing - Original Draft, Supervision, Resources, Funding Acquisition, Formal Analysis, Data Curation, Conceptualization. **Jiaqi Lu:** Writing - Review & Editing, Writing - Original Draft, Software, Resources, Formal Analysis, Data Curation. **Bo Niu:** Writing - Review & Editing, Supervision, Data Curation. **Xiaohua Liu:** Software, Methodology, Investigation. **Junming Hong:** Writing - Review & Editing, Supervision, Investigation. **Zhenming Xu:** Writing - Review & Editing, Supervision, Conceptualization.

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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Appendix A. Supplementary data

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