



Life cycle assessment of hydrometallurgical recycling for cathode active materials

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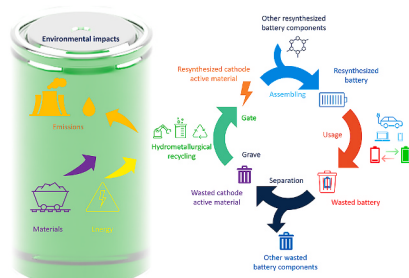
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HIGHLIGHTS

- Compared hydrometallurgical recycling processes for cathode active materials.
- Conducted life cycle assessment based on the ReCiPe method.
- Found the most effective recycling processes for NMC, LFP, and LCO.
- Compared NMC, LFP, and LCO considering specific capacity and lifespan.

GRAPHICAL ABSTRACT



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ABSTRACT

As battery usage keeps growing due to a boom in the electric vehicles market, battery recycling has become a crucial issue. Cathode active material is the most valuable component of the battery and attracts researchers' attention. Much research has been focused on finding the optimal condition of a certain hydrometallurgical recycling process for cathode active material. However, there is no comprehensive comparison between different hydrometallurgical recycling processes. In this research, life cycle assessment is adopted to evaluate the environmental concerns, which is a major reason for using batteries and recycling batteries, of different hydrometallurgical recycling processes for three widely used cathode active materials: NMC, LFP, and LCO. The environmental impact of each process is assessed based on the ReCiPe method using standardized processes and

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unified reactors. As a result, the most effective hydrometallurgical recycling processes for NMC, LFP, and LCO have been found. Moreover, the most effective recycling processes lower the environmental impact by over 50% compared to the average of those evaluated. Also, the ultimate comparison between different cathode active materials shows that LFP recycling has a lower environmental impact than NMC and LCO, considering specific capacity and lifespan.

1. Introduction

As the growing environmental concerns and technological breakthroughs, electric vehicles (EVs) have become increasingly popular over the last few years. Moreover, more than 280 million EVs are projected to be on the roads by 2030 [1,2]. Due to the growing demand for EVs, lithium-ion batteries (LIBs) usage is increasing dramatically [3]. This results in the problem of handling wasted LIBs. Thus, LIBs recycling has become a critical topic because of the increasing cost of mineral materials, environmental issues, and regulatory requirements [4]. Through a pretreatment process of spent LIBs before recycling, different components can be separated [5]. Among the components of the LIBs, the cathode is the most valuable part, so it attracts most researchers' attention regarding recycling [6–8]. Cathode contains other composites besides cathode active material. However, their weight varies among different batteries, making it difficult to compare different recycling methods fairly. For this research, we focused on the recycling of cathode active material. For the recycling of cathode active material, three recycling processes are widely used: hydrometallurgical, pyrometallurgical, and direct [9]. Amid the recycling processes, pyrometallurgical recycling's high energy requirement and hazardous gas emissions make it not the favored recycling method in the aspect of environmental protection [10]. Direct recycling has not been widely commercially used [11]. As a result, hydrometallurgical recycling draws attention from researchers due to its high recovery of metals with high purity, low energy consumption, and very low gas emissions [12]. Researchers tried to find the optimal conditions for hydrometallurgical recycling, especially for the leaching process, including leaching agent concentration, leaching time, leaching temperature, and liquid to solid (L/S) ratio, to achieve the highest recycling rate using only one type of leaching agent. However, there is no comprehensive comparison between different leaching agents, much less the comparison of different hydrometallurgical recycling processes for cathode active material. Though some review papers compare the recycle rate of different leaching processes using different leaching agents [4,6,13], there is no conclusion in view of environmental impact for the most effective leaching process of the commonly used cathode active materials, due to the lack of comparison of materials/energy inputs and waste outputs. As for the whole hydrometallurgical recycling for cathode active material, there is no study that provides the industry with the most effective solution to achieve high recycling rates, while lowering the process's environmental impact.

To comprehensively compare hydrometallurgical recycling processes for cathode active material, we converted different recycling

processes from literature to standard grave-to-gate processes (from waste cathode active material to new cathode active material) as shown in Fig. 1a. The full hydrometallurgical recycling processes are obtained from different literature, while considering different leaching agent for leaching. To ensure a standardized comparison, the recycled cathode active material must be the same weight as the wasted cathode active material. So, the replenishment amounts for the valuable metal compounds depend on their recycling rates. For life cycle assessment (LCA), the ReCiPe method, which is a method for the life cycle impact assessment, and the ecoinvent database are adopted. Using the ReCiPe method, a final score representing the process's environmental impact can be calculated [14]. As shown in Fig. 1b, based on the market share of cathode active materials [15], we chose $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$ (NMC), LiFePO_4 (LFP) and LiCoO_2 (LCO) as the target cathode active materials to find the most effective hydrometallurgical recycling processes.

The final score of environmental impact for each standardized process is calculated. As a result, we found the most effective hydrometallurgical recycling processes for NMC, LFP, and LCO. Since different cathode active material has different hydrometallurgical recycling processes, specific capacity, and battery lifespan. An ultimate comparison of different cathode active materials shows that LFP has a lower environmental impact than NMC and LCO, for the close-loop LCA of hydrometallurgical recycling.

2. Methods

2.1. Literature review: leaching

Leaching is the first and essential step of hydrometallurgical recycling. For different cathode active materials, different leaching agents are adopted. Hydrogen peroxide is often added as an oxidant [16]. It can improve the leaching process by reducing leaching agent usage and increasing the recycling rate. Because literature has already optimized the leaching agent concentration, leaching time, leaching temperature, and L/S ratio of a certain type of leaching agent. We directly used the optimized leaching conditions of each leaching agent from the literature. For the same leaching agent, several literatures have listed recycling processes. However, they usually have different optimal leaching agent concentration, leaching time, leaching temperature, L/S ratio, and they claim different recycling rates. Therefore, we chose the literature with the highest recycling rate. If more than one literature claimed different results of an identical process, we chose the most recent one that claimed improvement and higher accuracy.

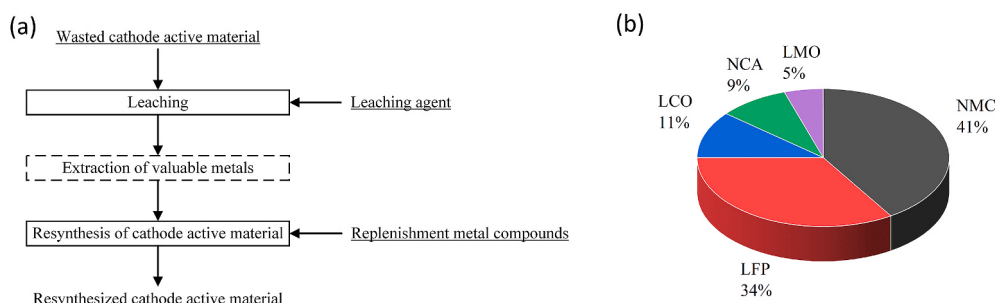


Fig. 1. (a) Flowchart of grave-to-gate process for cathode active material recycling. (b) Market share of different cathode active materials.

Table 1
NMC leaching.

Leaching agent	Leaching time (mins)	Temperature (°C)	L/S ratio (mL/g)	Recycling rate (%)				Ref
				Lithium	Cobalt	Nickel	Manganese	
4 M Ammonia + 1.5 M Ammonium sulphate	300	70	100	95.3	80.7	89.8	4.3	[18]
1 M Citric acid + Hydrogen peroxide	40	60	12.5	98.0	98.0	98.0	98.0	[19]
0.4 M Citric acid + 0.2 M Phosphoric acid	30	90	50	100	91.6	93.4	92.0	[20]
1.5 M Lactic acid + Hydrogen peroxide	20	70	50	97.7	98.9	98.2	98.4	[21]
0.5 M Nitric acid + 0.5 M Ascorbic acid	10	85	50	100	100	100	100	[22]
2 M Sulfuric acid + Hydrogen peroxide	120	60	10	98.9	98.4	98.8	98.6	[23]
3 M Trichloroacetic acid + Hydrogen peroxide	30	75	20	99.7	91.8	93.0	89.8	[24]

2.1.1. NMC

NMC can operate in a large voltage range and has a relatively low cost [17]. Here we focused on NMC111 ($\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$) because it has been more thoroughly researched than others. The leaching process of NMC can obtain four valuable metals: lithium, cobalt, nickel, and manganese. Table 1 presents a comprehensive list of the different leaching methods for NMC. The recycling rate of lithium and cobalt, the most valuable metals in NMC, are similar among all the leaching agents. However, the L/S ratio of different leaching agents varies a lot. These will dramatically influence the environmental impact caused by the usage of leaching agents. Also, the differences in leaching time and temperature result in a difference in energy consumption, which will finally contribute to environmental impact. Besides, air pollutants during leaching will contribute to the environmental impact.

2.1.2. LFP

LFP is a relatively safe cathode active material because phosphates are not prone to thermal runaway and will not burn [31]. Also, LFP is cobalt free [32,33], greatly reducing the manufacturing cost [34]. Besides, recycling LFP is easier than recycling NMC since it only has two valuable metals: lithium and iron. As shown in Table 2, most leaching processes for LFP only require room temperature, which means less energy is needed for the leaching process. This finally reduces the environmental impact of the leaching of LFP.

2.1.3. LCO

LCO has a relatively high specific capacity and is easy to synthesize in bulk quantities, making it a popular cathode material [35]. However, due to the relatively short cycle life and low thermal stability of LCO, LCO recycling is of great significance. Because LCO has a longer history than NMC and LFP, there is more research on leaching for LCO, as shown in Table 3.

2.2. Literature review: grave-to-gate recycling

For the full hydrometallurgical recycling process of cathode active material, standardization of the process is necessary to make them comparable. Here we considered the hydrometallurgical recycling process as the process in which input wasted cathode active material and output resynthesized cathode active material. In order to standardize the recycling process for comparison. The wasted cathode active material is 100% recycled to the resynthesized cathode active material. However,

there are losses of materials during the recycling process. So, metal compounds are replenished to ensure the weight of resynthesized cathode active material equals the weight of input waste cathode active material. Besides, we assumed that any leaching agent could be combined with any hydrometallurgical recycling process freely after adjusting the ions concentration by evaporation. Furthermore, the leaching efficiency will influence the weight of replenishment metal compounds. Since different cathode active materials have different recycling processes, we analyzed each cathode active material individually. Each cathode active material has several options for recycling: options 1-4 for NMC, 5-6 for LFP, and 7-10 for LCO.

2.2.1. NMC

NMC can be treated with solvent extraction after leaching to recover valuable metals. As shown in Fig. 2a, Cyanex 272 is used as an extractant to separate cobalt and manganese from the leach solution. Followed by steps using D2EHPA, cobalt can be separated. Then, dimethylglyoxime selectively precipitates nickel from the solution with lithium and nickel ions. After that, the high-purity Li_2CO_3 , $\text{Ni}(\text{OH})_2$, $\text{Co}(\text{OH})_2$, and $\text{Mn}(\text{OH})_2$ can be obtained through chemical precipitation [45]. Finally, using a solid-state method, NMC can be resynthesized [46]. This process is the recycling process option 1.

However, separating metals costs a lot of extractants, leaching agents, and energy. Researchers tried a new process: option 2, as shown in Fig. 2b. Manganese, cobalt, and nickel are co-extracted and separated from lithium by D2EHPA in kerosene. Then, Li_2CO_3 is recovered through precipitation. From the organic load phase, stripping liquor can be obtained. Finally, NMC is directly regenerated from stripping liquor without separating metal individually by the coprecipitation method [47].

Fig. 2c shows the recycling process option 3, which is a more straightforward process. It involves a gel formation step for leach solution after adding supplemental metal compounds and aqueous ammonia. After drying and calcination, NMC powder can be made [19].

Similarly, the recycling process option 4 uses the coprecipitation method to synthesize precursors after replenishing nickel, cobalt, and manganese. Then NMC is resynthesized through solid-state sintering [48]. The process is illustrated in Fig. 2d.

2.2.2. LFP

As shown in Fig. 3a, the recycling process option 5 for LFP separates FePO_4 directly from the leach solution. Then, Li_2CO_3 is obtained through

Table 2
LFP leaching.

Leaching agent	Leaching time (mins)	Temperature (°C)	L/S ratio (mL/g)	Recycling rate (%)		Ref
				Lithium	Iron	
1.25 M Acetic acid	30	50	8.3	94.5	99.3	[25]
Citric acid (lemon juice) + Hydrogen peroxide	90	25	15	94.8	96.0	[26]
0.1 M Oxalic acid	30	25	100	99.3	94.0	[27]
0.6 M Phosphoric acid	20	25	20	94.3	97.7	[28]
3 M Sodium persulfate	20	25	3.3	99.9	99.9	[29]
0.3 M Sulfuric acid + Hydrogen peroxide	120	60	11.8	95.8	99.9	[30]

Table 3
LCO leaching.

Leaching agent	Leaching time (mins)	Temperature (°C)	L/S ratio (mL/g)	Recycling rate (%)		Ref
				Lithium	Cobalt	
1.25 M Ascorbic acid	20	70	40	98.5	94.8	[36]
1 M Citric acid + Hydrogen peroxide	70	70	25	99.0	99.0	[37]
2 M Hydrochloric acid	90	80	50	92.2	99.0	[38]
3 M Hydrochloric acid + Hydrogen peroxide	40	80	20	81.0	79.0	[39]
1 M Nitric acid + Hydrogen peroxide	60	75	50	85.0	85.0	[40]
1 M Oxalic acid	150	95	66.6	98.0	97.0	[41]
1.5 M Phosphoric acid + Glucose	120	80	500	99.8	98.0	[42]
0.7 M Phosphoric acid + Hydrogen peroxide	60	40	20	99.0	99.0	[43]
2 M Sulfuric acid	90	80	50	91.0	55.8	[38]
2 M Sulfuric acid + Hydrogen peroxide	30	75	10	95.0	94.0	[44]

precipitation. The recovered Li_2CO_3 mixed with the previously recycled FePO_4 (with the ratio of Li_2CO_3 : $\text{FePO}_4 = 1.05$) can prepare LFP [25].

Recycling process option 6 further simplifies the resynthesis for LFP. After leaching, supplements are added to adjust the molar ratio of Li: Fe: P to 3: 1: 1. Then, through a hydrothermal process, LFP is resynthesized [49]. The process is illustrated in Fig. 3b.

2.2.3. LCO

For LCO, recycling process option 7 uses PC-88A to extract cobalt from the leach solution. After strip and crystallization, $\text{CoSO}_4 \cdot 6\text{H}_2\text{O}$ can be obtained. For the raffinate, Li_2CO_3 is obtained through precipitation [50]. Using a solid-state method, LCO can be resynthesized [46]. The process is illustrated in Fig. 4a.

As shown in Fig. 4b, the recycling process option 8 uses Cyanex 272 to extract cobalt from the leach solution. Like option 7, $\text{CoSO}_4 \cdot 6\text{H}_2\text{O}$ can be obtained after strip and crystallization. The raffinate is extracted by Cyanex 272 again before precipitation to obtain Li_2CO_3 [44]. Finally, LCO can be resynthesized using the solid-state method [46].

Researchers further simplified the recycling process. Recycling process option 9 adjusts the molar ratio of Li: Co to 1.1: 1 after leaching. At the same time, citric acid is added to prepare a gelatinous precursor. After the calcination of the precursor, purely crystalline LCO is successfully obtained [40]. The process is illustrated in Fig. 4c.

Also, LCO can be resynthesized through stoichiometric, well crystallized, and structurally ordered compounds from the recovered cobalt and lithium compounds. As shown in Fig. 4d, for the recycling process option 10, cobalt can be recovered by precipitation with oxalic acid. After calcination, Co_3O_4 can be obtained. Also, Li_2CO_3 can be obtained after evaporation and calcination. Then, the recovered Co_3O_4 and Li_2CO_3 compounds can be used as precursors for the resynthesis of LCO [51].

2.3. Life cycle assessment

LCA is a tool to assess the environmental impacts of a wide range of products and activities [52,53]. It can help decision-makers identify improvement strategies without burden shifting [54]. One of the major reasons for using and recycling batteries is environmental concerns [55]. So, we adopted LCA to compare different recycling options and find the most effective hydrometallurgical recycling options for cathode active materials. This LCA study has been conducted based on ISO 14040 and ISO 14044 standards, which are globally established standards [56].

2.3.1. Goal and scope

In this study, we compared different hydrometallurgical recycling processes. Recycling can be considered a grave-to-gate process, which takes wasted cathode active material as input and produces resynthesized cathode active material as output. The leach solution evaporates, or water is added to adjust the concentration. This ensures the smooth combination of different leaching agents to other cathode resynthesis options. Due to the variation and uncertainty of weight percentages of

different components in batteries [57], only cathode active materials (NMC, LFP, and LCO) are considered. To ensure a fair comparison, resynthesized cathode active material must be the same weight as input wasted cathode active material. However, there are losses during leaching. Thus, supplemental metal compounds are added. The goal is to find the most environmentally friendly hydrometallurgical recycling process for each cathode active material.

2.3.2. Source of inventory data

Ecoinvent 3.8 is adopted as the life cycle inventory (LCI) database for this study. It supports various types of sustainability assessments and regionalized life cycle impact assessments [58]. Because detailed hydrometallurgical recycling processes can be obtained from the literature. The bottom-up approach is adopted, using data from the literature for certain key steps within the processes [59]. The material demand can be calculated based on the L/S ratio of leaching, the O/A ratio of solvent extraction, and other detailed descriptions from the literature. The recycling rate of leaching can be used to calculate the demand for supplemental metal compounds. Besides, some processes require more supplemental metal compounds to ensure the reaction. Those are added to the demand for supplemental metal compounds. The emissions are also calculated based on the detailed descriptions from the literature, and the wastewater is calculated based on the acid usage.

2.3.3. Modeling of recycling energy demand

Because different literatures use different sizes of continuous stirred tank reactors and ovens, we adopted industrial continuous stirred tank reactors and ovens for all processes to standardize the process. The reactor has a capacity (m_0) of 10,000 kg. The energy consumed by the reactor (q_1) can be obtained by adding up the energy consumed in heating the reactor (q_{reactor}), energy consumed in stirring the reactor (q_{stir}), and half of the energy (assuming half can be recovered and reused) consumed in heating the solvent (q_{solvent}) [60]. Since the power source is electricity, the efficiency (η) of heating the reactor and solvent is 80% [46].

$$q_1 = \frac{q_{\text{reactor}}}{\eta} + q_{\text{stir}} + \frac{q_{\text{solvent}}}{2\eta} \quad (1)$$

q_{reactor} can be calculated using the thermal conductivity of the insulation (λ), the thickness of the insulation (x), the surface area of the reactor (A), reactor wall temperature (T_0), reactor temperature (T_r), reaction time (t_r) [46]. We used the reactor's capacity and calculated the total weight of the leaching process (m_{total}) to make processes have different L/S ratios comparable.

$$q_{\text{reactor}} = \frac{\lambda A (T_r - T_0) t_r m_{\text{total}}}{x m_0} \quad (2)$$

q_{stir} is based on rotation speed (ω), standard rotation speed (ω_0), and standard power consumed in stirring (q_0) [61].

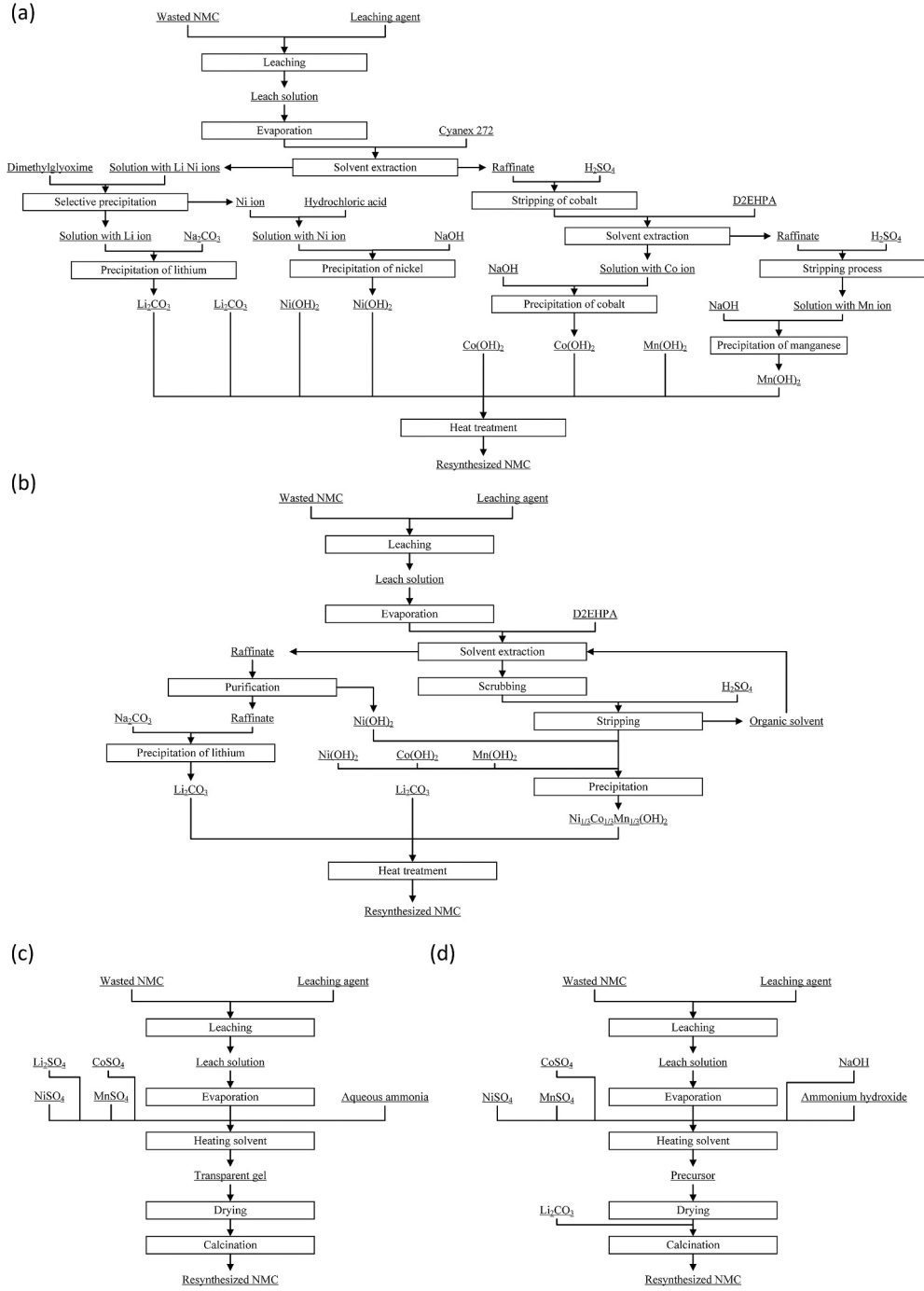


Fig. 2. NMC recycling processes (a) Recycling process option 1. (b) Recycling process option 2. (c) Recycling process option 3. (d) Recycling process option 4.

$$q_{stir} = \frac{\omega}{\omega_0} \cdot q_0 \cdot t_r \cdot \frac{m_{total}}{m_0} \quad (3)$$

$q_{solvent}$ is calculated by the heat capacity of the reactant (C_p), the weight of the reactant (m), and temperature change (ΔT) [46]. If the step includes evaporation, water vaporization heat (ΔH_{vap}) needs to be considered [46].

$$q_{solvent} = \sum C_p m \Delta T + m_{water} \Delta H_{vap} \quad (4)$$

As for the oven, an 8300L capacity (V_0) oven is chosen, with a coefficient of utilization (ϵ) of 33% [60]. So, we can calculate the energy consumption using the total volume of the reactants (V_{total}). The energy consumed by the oven (q_2) can be obtained by the oven's power (P) and reaction time [46].

$$q_2 = P \cdot t_r \cdot \frac{V_{total}}{\epsilon V_0} \quad (5)$$

After collecting the data on the oven's power and temperature from the literature [46,60], the power of the oven can be calculated by the reaction temperature (T):

$$P = 0.8579 \cdot e^{0.0063T} \quad (6)$$

According to the plan for battery plants [62], most new plants will be located in the southeast. We assumed that the battery recycling plants are close to the battery plants. So, SERC Reliability Corporation, which covers most southeast states [63], is chosen as the electricity source for calculation.

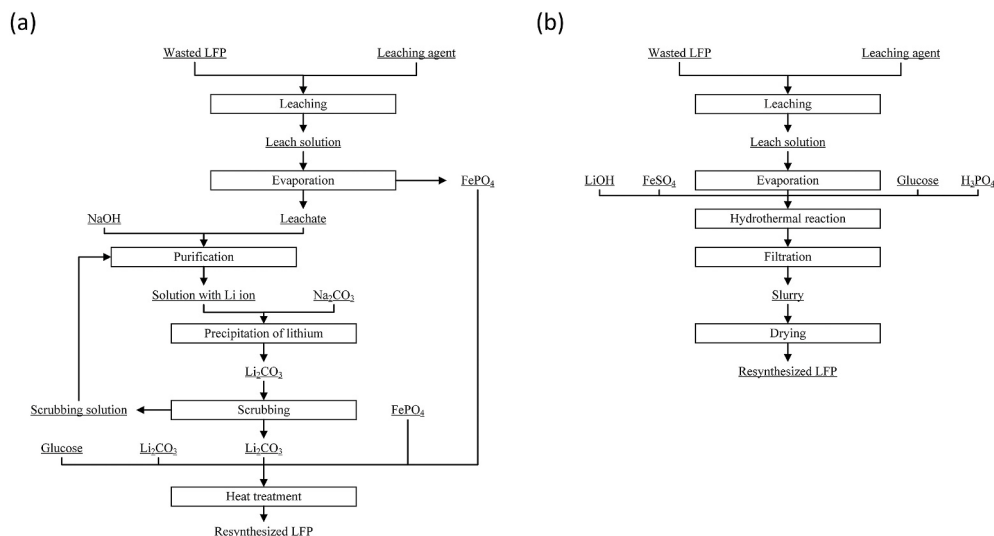


Fig. 3. LFP recycling processes (a) Recycling process option 5. (b) Recycling process option 6.

2.3.4. Applied impact assessment methodology

Based on the ISO 14040 standard, LCIA using the ReCiPe method consists of five steps: (1) characterization, (2) damage assessment, (3) normalization, (4) weighting, and (5) single score [64]. For life cycle substance, the impact categories which it contributes to are identified. Then, the contributions are calculated based on the characterization factor, which represents the relative contribution of the corresponding substance. It should be noted that each life cycle substance can contribute to more than one impact category. The egalitarian perspective is chosen due to its precautionary perspective with the longest time horizon, and it covers all impact types ranging from lightly to certainly proven facts [65]. The ReCiPe method has a midpoint level and an endpoint level for life cycle impact assessment [14]. For the midpoint level, there are 18 impact categories (e.g., ozone depletion, human toxicity, radiation). For midpoint level categories, some can contribute to only one endpoint category, while some can contribute to multiple endpoint categories. At the endpoint level, 3 endpoint categories (human health, ecosystems, resources) are calculated from the midpoint impact categories. Normalization and weighting options are adopted to simplify the impact indicator result and consequent comparison [14]. Combine these 3 endpoint categories, a single score can be obtained. Its points express the total environmental impact [56]. The total environmental impacts were calculated based on per kg of resynthesized cathode active material. Contrarily, If the process has a minor point, that means the process has a relatively low environmental impact [65].

3. Results

The ReCiPe endpoint single score (shown as environmental impact in the figures) is calculated for each hydrometallurgical recycling process of cathode active materials.

3.1. NMC

As shown in Fig. 5a, 2 M sulfuric acid with hydrogen peroxide using option 4 is the most effective hydrometallurgical recycling process for NMC. It has an environmental impact of 1.64 Pts/kg, while the average environmental impact is 18.69 Pts/kg of all evaluated processes for NMC. Options 3 and 4 have more straightforward processes and eliminate the steps for solvent extraction, so they have lower environmental impacts than options 1 and 2. Though options 1 and 2 both use solvent extraction, option 2 has a lower environmental impact due to its reduced usage of extractant. As for leaching agent, sulfuric acid and citric acid have relatively low environmental impacts, thanks to their low L/S ratio.

3.2. LFP

As for LFP, option 5 has a lower environmental impact than option 6, because option 5 requires less supplemental lithium compound. As we can find in Fig. 5b, the most effective hydrometallurgical recycling process for LFP is 3 M sodium persulfate using option 5. It can achieve an environmental impact of 1.57 Pts/kg. The average environmental impact of all evaluated processes for LFP is 4.02 Pts/kg. Besides, 0.3 M sulfuric acid with hydrogen peroxide using option 5 can also be a feasible process with an environmental impact of 1.65 Pts/kg.

3.3. LCO

As shown in Fig. 5c, the most effective hydrometallurgical recycling process for LCO is 0.7 M phosphoric acid with hydrogen peroxide using option 10. It has an environmental impact of 9.86 Pts/kg, while the average environmental impact of all evaluated processes for LCO is 45.3 Pts/kg. Options 9 and 10 have relatively low environmental impacts compared to options 7 and 8, thanks to the simplified recycling processes to eliminate the usage of extractants.

4. Discussion

A close-loop LCA is proposed to compare different cathode active materials after finding the most effective hydrometallurgical recycling processes for each cathode active material. Since the grave-to-grave process has been calculated, the gate-to-grave process is needed to complete the close-loop LCA. Their specific capacity and battery lifespan need to be considered for the gate-to-grave process of cathode active materials. The batteries are assumed to be used in electric vehicles. The literature [59,66–70] shows the specific capacity and battery lifespan listed in Table 4.

Assuming the optimal recycling condition, the maximum calculated specific capacity is adopted. The close-loop LCA environmental impact (E_c) can be calculated by specific capacity (Q), battery lifespan (L), and previously calculated grave-to-gate LCA environmental impact (E_g).

$$E_c = \frac{E_g}{QL} \quad (7)$$

The result is shown in Fig. 5d. NMC and LFP have much lower environmental impacts than LCO. There are two major reasons: On the one hand, LCO has a shorter lifespan (cycles) compared to NMC and LFP. On the other hand, LCO has a higher weight percentage of cobalt compared to NMC and LFP. During the recycling process, cobalt needs to

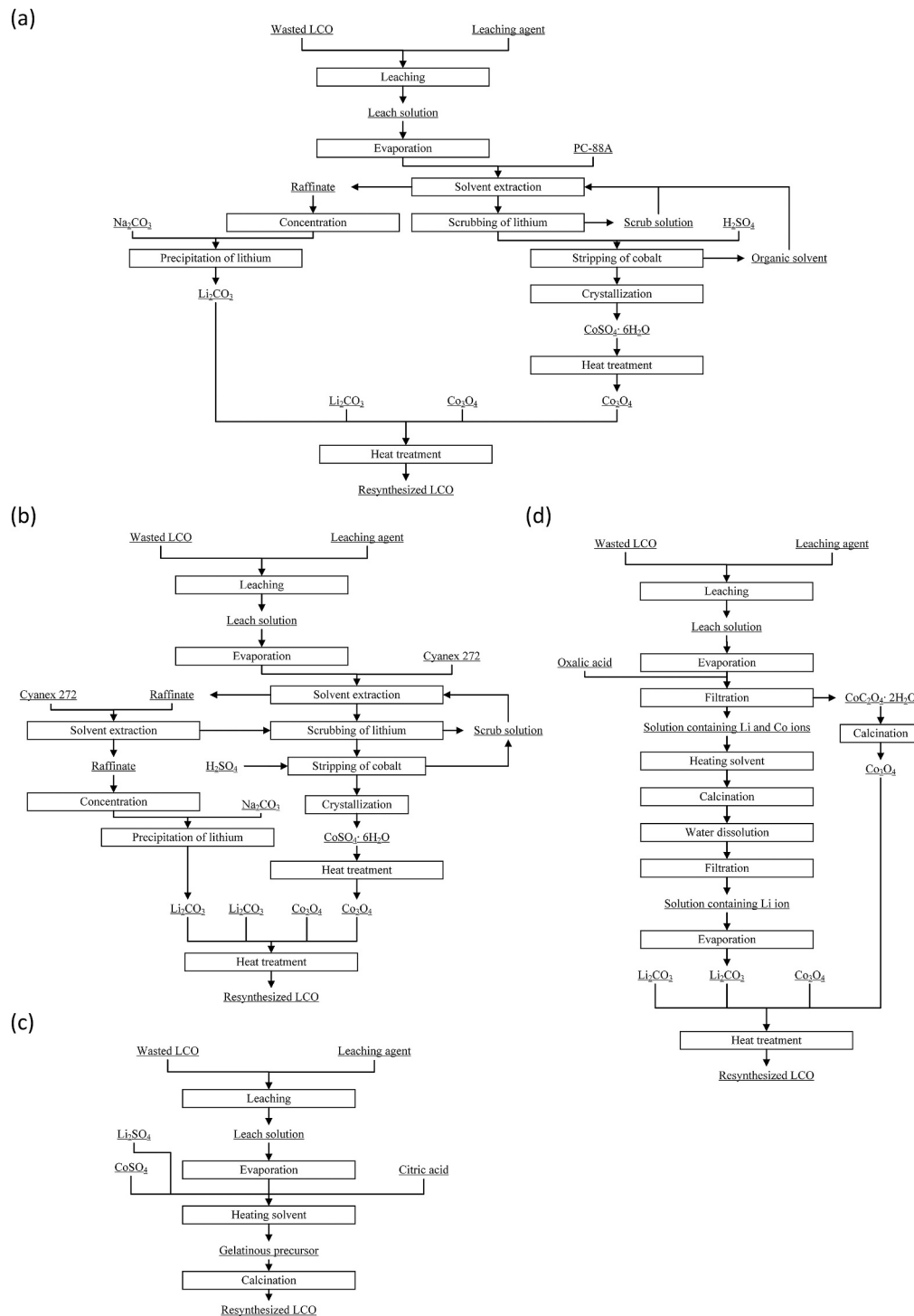


Fig. 4. LCO recycling processes (a) Recycling process option 7. (b) Recycling process option 8. (c) Recycling process option 9. (d) Recycling process option 10.

be replenished. Therefore, LCO needs more replenishment of cobalt, which significantly contributes to the high environmental impact of LCO. LFP has a lower environmental impact of 0.0029 Pts/mAh compared to NMC's 0.0034 Pts/mAh, while assuming both have the likeliest lifespan. However, due to the diversity in battery lifespan, the environmental impact of NMC is between 0.0020 and 0.0059 Pts/mAh, and the environmental impact of LFP is between 0.0018 and 0.0058 Pts/mAh.

Though from the economic aspect, the manufacturing cost for LFP is relatively low, and the economic drive for LFP recycling is compromised [71]. Regarding environmental impact, LFP recycling is important since

more and more EVs have been adopting LFP batteries in recent years [72]. In most literature, which provides the recycling process data, the purity grade of cathode active material or the electrochemical behavior of LIBs are not tested or tested on different standards. So, we only compared the environmental impact of the optimal recycling condition.

5. Conclusions

Through the literature review, we listed the hydrometallurgical recycling processes (including different leaching agents) for widely used cathode active material: NMC, LFP, and LCO. The environmental impact

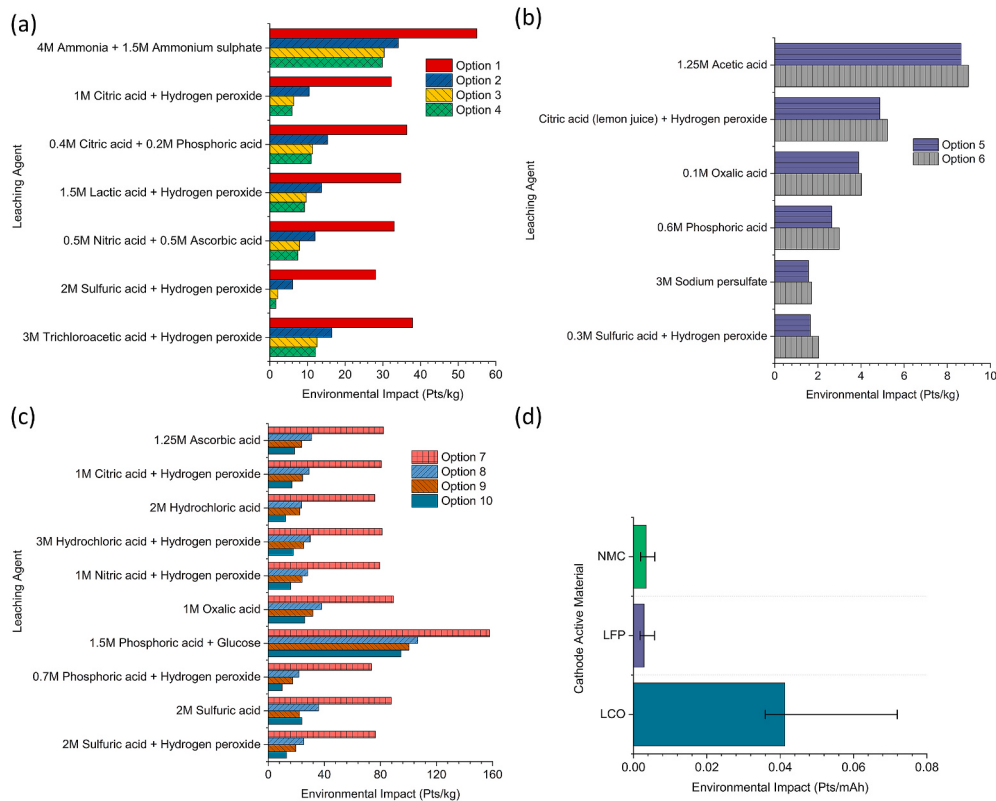


Fig. 5. Comparison of different hydrometallurgical recycling processes for (a) NMC. (b) LFP. (c) LCO. (d) Close-loop comparison of NMC, LFP, and LCO.

Table 4
Specific capacity and battery lifespan of cathode active materials.

Cathode active material	Specific capacity (mAh/g)	Battery lifespan (cycles)		
		Minimum	Likeliest	Maximum
NMC	280 [66]	1000 [67]	1700 [67]	3000 [67]
LFP	170 [68]	1600 [67]	3200 [67]	5039 [67]
LCO	274 [69]	500 [70]	871 [59]	1000 [70]

of each process is assessed based on the ReCiPe method using standardized processes and unified reactors. The processes with the lowest environmental impact are identified as follows: NMC using option 4 [48] with sulfuric acid and hydrogen peroxide [23]; LFP using option 1 [25] with sodium persulfate [29]; LCO using option 4 [51] with phosphoric acid and hydrogen peroxide [43]. The most effective recycling processes lower the environmental impact by over 50% compared to the average of those evaluated. Generally speaking, the process with more straightforward steps and fewer extractants has a lower environmental impact. Moreover, the leaching agent with a lower L/S ratio is favored for the leaching step. As for close-loop LCA comparison, LFP has a lower environmental impact than NMC and LCO. Though only cathode active materials recycling is researched due to the variation and uncertainty of weight percentages of different components in batteries, this study can help other researchers explore the next generation of advanced recycling process for battery.

CRedit authorship contribution statement

Zheng Liu: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Jarom G. Sederholm:** Validation, Writing – review & editing. **Kai-Wei Lan:** Validation, Writing –

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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.

Data availability

Data will be made available on request.

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