Enabling Sustainable Critical Materials for Battery Storage Through Efficient Recycling and Improved Design: A Perspective

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ABSTRACT:

Recent years have seen the rapid growth in lithium-ion battery (LIB) production to serve emerging markets in electric vehicles and grid storage. As large volumes of these batteries reach their end of life, the need for sustainable battery recycling and recovery of critical materials is a matter of utmost importance. Global reserves for critical LIB elements such as lithium, cobalt and nickel will soon be outstripped by growing cumulative demands. Despite advances in conventional recycling strategies such as pyrometallurgy and hydrometallurgy, they still face limitations in high energy consumption, high greenhouse gas (GHG) emissions as well as limited profitability. While new direct recycling methods are promising, they also face obstacles such as the lack of proper battery labelling, logistical challenges of inefficient spent battery collection and components separation. Here, we discuss the importance of recovering critical materials, and how battery designs can be improved from the cell to module level in order to facilitate recyclability. The economic and environmental implications of various recycling approaches are analyzed, along with policy suggestions to develop a dedicated battery recycling infrastructure. We also discuss promising battery recycling strategies and how these can be applied to existing and future new battery chemistries.

ONE SENTENCE SUMMARY: A perspective on current state of battery recycling and future improved designs to promote sustainable, safe and economically viable battery recycling strategies for sustainable energy storage.

DISCUSSION POINTS: Internationally recognized tagging system – Should every battery be tagged with standardized vital information needed for waste processing? Recycling facilities – Should there be more centralization or decentralization of battery recycling in future?

KEY WORDS: Critical Materials, Battery Recycling, Sustainable Design, Recycling Policy and Economics

■ INTRODUCTION

Since its commercialization in the 1990s, the world experienced a rapid increase in adoption of lithium-ion batteries (LIBs) in consumer electronics and portable devices.[1-2] Despite this, relatively little attention was placed on the recyclability and environmental sustainability of early LIBs.[3] However, with tumbling prices of LIBs in terms of cost per watt-hour over the past two decades (>900USD / kWh in 2011 *vs* >150USD / kWh in 2020)[4], coupled with expeditious growth of electric vehicles (EVs) sales since the 2010s[5], demands for LIBs are expected to double within a decade.[6] This directly translates into the growing battery waste problem we face today that is both more sophisticated and larger in scale compared to the past. As existing EVs on the roads approach their end of life, their spent battery packs cannot be treated as generic waste in recovery facilities, and would require specialized processes to disassemble, treat and recover materials within in a safe, sustainable and economical manner. However, due to the wide ranging chemistries involved along with safety hazards associated with handling spent batteries, this poses challenges for 3rd party waste facilities in dealing with large volumes of spent LIBs. To this end, government agencies around the world have implemented policy measures to shift the responsibility of handling LIB waste back to the manufacturers. The European Parliament and Council of the European Union (EUROPA) set out specific battery directives for manufacturers within European nations to achieve LIB collection rates of 45% and to raise efficiencies of recycling above 50% of any spent LIBs.[7] Likewise, the Chinese government now holds EV manufacturers accountable for the recovery and recycling of used LIBs, and mandating the setup of recycling and servicing centers for battery collection, storage and transport to recycling facilities.[8] In the United States (US), the US Department of Energy's ReCell Center is taking the lead by setting the core principles for future LIB recycling technologies that seeks to achieve: 1) Direct recycling of electrode materials, 2) Higher recovery rates of components and 3) Better design for recyclability.[9] These recycling policy directives largely focus on the sustainable and potentially profitable recovery and regeneration of critical materials harvested from spent batteries. In this perspective, we highlight the importance of recovering critical battery materials from both a supply chain point of view, as well as how it can enable profitable LIB recycling. We also discuss promising battery recycling approaches and advanced design considerations as well as how these strategies can be applied to existing and future new battery chemistries.

Critical Battery Materials. Critical materials can be defined by their relative specific economic value as a function of any spent LIB, as well as their supply availability risks. While material scarcity within the earth's crust is an important factor, supply chain risks also arise from the relative uneven geographical distribution of critical materials around the world. For example, Chile holds the world's largest lithium (Li) reserves at approximately 7.5 million metric tons, followed by China and Argentina collectively making up more than 90% of global Li reserves amongst these 3 countries.[10-11] Likewise, the Democratic Republic of Congo is also the source for more than 60% of the world's cobalt supply.[11-13] This naturally places Li and cobalt (Co) near the top of the list of critical materials. However, the development of high performance layered oxide cathodes using other transition metals such as nickel (Ni) and manganese (Mn), partly driven by high prices of Co, have introduced a whole new catalogue of cathode material chemistries.[2, 14] While elements such as Ni and Mn are relatively more abundant than Co in the earth's crust, their increased usage in high battery volume devices such as EVs and grid storage warrants their recovery as critical battery materials as well.[15] The recovery and regeneration of these critical materials (Co, Ni, Mn, Li) are vital to achieve both supply chain sustainability and potential profitability of battery recycling.[1] **Figure 1** compares the trend in global reserves of the above critical materials and their corresponding demand growth. The relative abundance (vertical axis) of critical materials in ascending order are Co, Li, Ni followed by Mn with the highest global reserves.[16] From the trends observed, it is clear that rates of Li and Ni usage in batteries will come close to surpassing existing reserves by 2030 and would have surpassed Co reserves by 2028. Such a trajectory in supply and demand would potentially drive prices up, further underscoring the importance of recovery and recyclability. It is worthy to note that while critical materials are also used in non-battery related industries such as in construction, ceramic production and other minor applications, their annual demand has been comparatively low compared to demands in LIBs.[17]

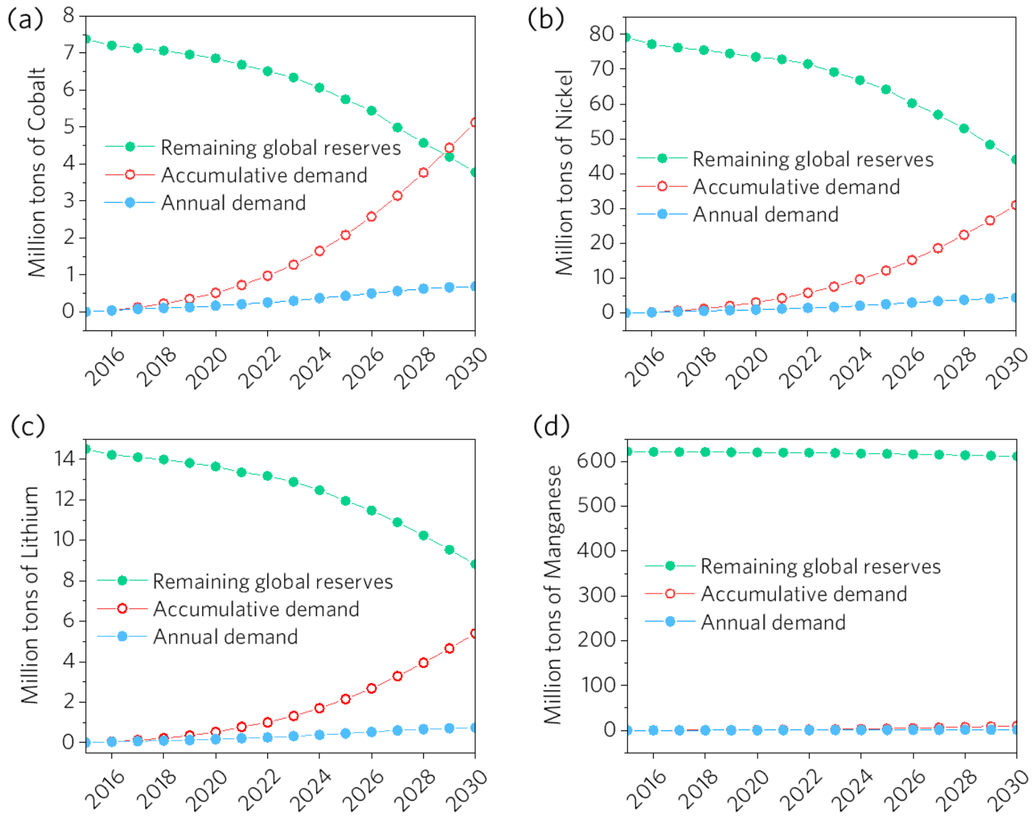


Figure 1. *Global projections of materials reserves vs current demand rate of critical materials (Co, Li, Ni, Mn). Recycling is not accounted for in the critical material projections.*[16]

Conventionally, LIB recycling is often seen as economically unfavorable, and often require government intervention to incentivize the collection, handling and treatment of battery waste.[18-19] However, as more batteries are produced, the bulk of critical materials resources would gradually move from the earth's crust toward being embedded within devices. As such, spent batteries should no longer perceived as waste, but be viewed as a valuable resource rich in critical materials that is no longer geographically limited in supply. Moreover, circular mining and harvesting of critical materials from spent batteries can be both technically simpler due to the relatively high purity of critical materials within batteries and potentially lower in costs compared to mining from natural minerals. From an ethical standpoint, recovering critical materials from spent batteries will also reduce human exposure to toxic materials during mining and the environmental damage incurred due to disposal of contaminated waste often seen in countries lacking the political or regulatory framework needed to ensure sustainable and ethical mining.[20-21]

Battery Secondary Usage. Another alternative pathway for spent batteries is the possibility for refurbishment and reuse in second life applications. As the end-of-life (EOL) batteries from EVs may still contain up to 80% of its original capacity[22-23], these cells can be used in applications with lower current and energy density demands, such as stationary storage.[3] Traditionally, these markets were served by lead-acid or aqueous type batteries that met the needs of emergency backup power supply. However, rapid growths of renewable energy deployment has forced stationary storage to extend its role from sporadic use during power shutdowns to daily high powered, high frequency switching in order to address the intermittency challenges faced by renewables.[6] As such, spent LIBs can meet both the power density and response times needed to meet such demands.[11] This was demonstrated in 2018 where Eaton and Nissan jointly incorporated a combination of new and second-life battery packs from used Nissan LEAFs in Amsterdam's Johan Cruijff ArenA to store solar power generated from the stadium's roofs for use during peak periods.[24] In another example, the Ministry of Industry and Information Technology (MIIT) in China partnered with battery manufacturers such as BYD, to replace lead-acid battery packs used in almost 2 million telecommunication towers across China with retired EV LIBs packs.[25] When directly sourced from EV manufacturers, these second life LIB packs can service markets for up to an additional 10 years after its primary use, at costs up to 50% or lower compared to new LIB packs.[26-27] However, second life applications can only serve to extend the usage and lifespans of LIBs, and critical materials from spent LIBs would ultimately still need to be recycled when the battery packs are no longer serviceable. In the following sections, we discuss conventional methods to harvest critical materials and resynthesize them into new battery materials, and how they compare against new direct recycling approaches in improving recovery efficiencies, energy costs as well as reducing environmental emissions of recycling.

■ Conventional Methods to Recover & Extract Critical Materials

Prior to any LIB recycling process, EOL LIBs must first be fully discharged to remove any residual stored energy in order to avoid potential fire or explosion hazards. This is typically done by submerging cells into aqueous salt solutions until they exhibit zero voltage.[2, 28] Next, the discharged cells undergo various pre-treatment processes such as dismantling, crushing, physical separation, milling, thermal treatment and other processes to break down the physical and chemical structures of materials (**Figure 2**). While pretreatment is typically done manually in the laboratory, the large volumes of spent batteries handled in industrial recycling often require bulk mechanochemical pretreatment instead.[6, 14] Pre-treatment of bulk battery packs serve to reduce the physical volume of materials as well as to consolidate components containing the critical materials, which help improve separation efficiencies in downstream processes. Subsequently, the collected cells are either recycled using pyrometallurgical processing or further treated with sieving and separation of sub-components before hydrometallurgical processing.

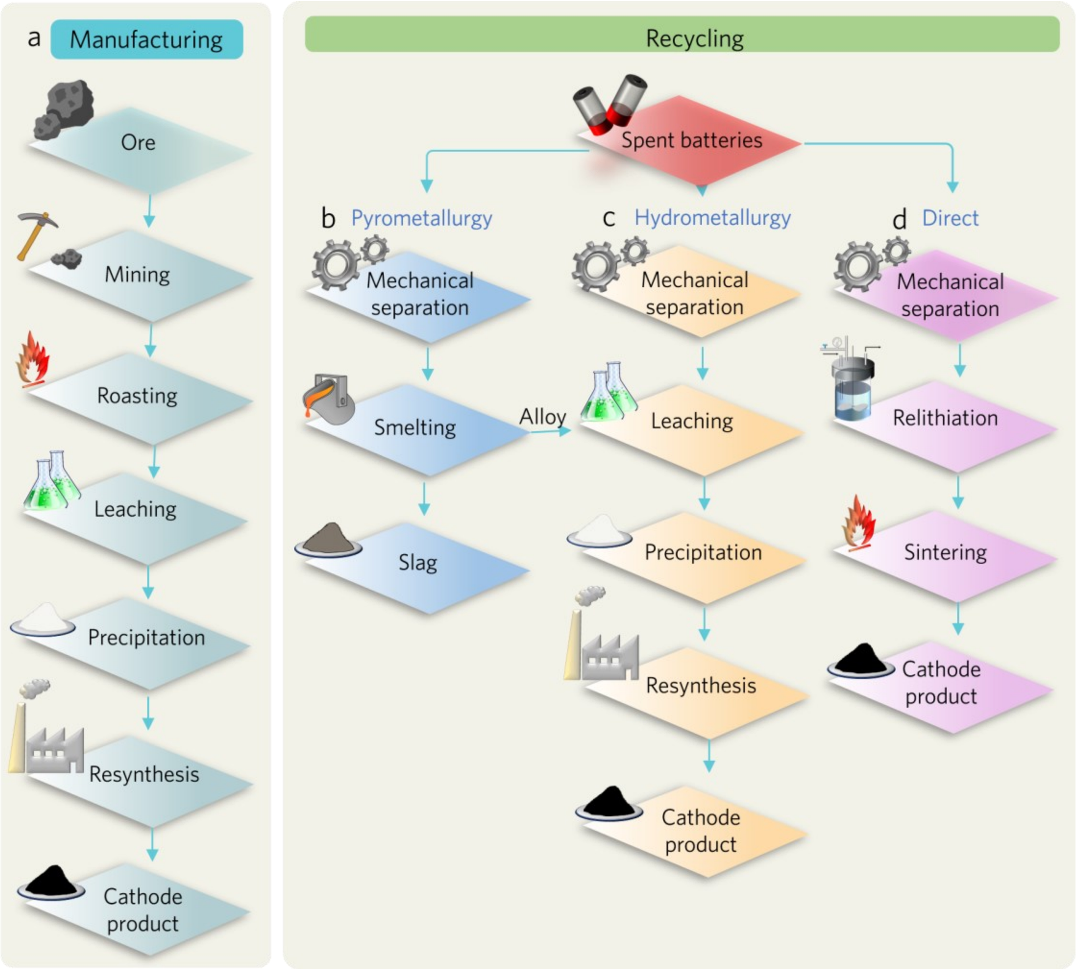


Figure 2. *Process workflows for primary manufacturing, conventional recycling methods for the extraction of critical materials, and direct recycling methods.*

Pyrometallurgy. Amongst various metallurgical processes reported, pyrometallurgy is considered relatively simpler and require fewer separation steps to treat and recover critical materials. High-temperature smelting (**Figure 2**) is first used to decompose the battery components to form a mixture of molten slag and metal alloys containing copper (Cu), Ni, Co, iron (Fe), and other metals that are later purified. Although revenue is generated from Cu, Ni and Fe recovery, Co still makes up the bulk of valuable materials recovered due to their higher prices and mass fraction in spent batteries.[1, 29] Potentially harmful chemicals such as plastics or organics are also combusted and at the same time provide additional energy during the thermal process. However, metals such as Li, aluminum (Al) and Mn often cannot be recovered as they remain trapped within the mixture of slag materials.[2, 28] As such, the future economic prospects of pyrometallurgy in battery recycling may be impacted as the EV industry continues moving toward low-Co LIB cathode materials and prices of scarce Li continue maintaining their upward trend. Additionally, pyrometallurgical processes also pose other downsides such as poorer recovery efficiencies, high energy costs as well as the need for additional toxic gas handling. To overcome this, environmentally friendly *in-situ* recovery pyrometallurgical methods have been recently developed, which have been demonstrated to reduce emissions as well as recover Li within the spent battery.[30-31] By roasting the materials between 500-1000˚C in a vacuum or inert gas environment, the cathode materials such as $LiMn_2O_4$, $LiCoO_2$, and $LiCo_xMn_yNi_zO_2$ ($x+x+z = 1$) would react with graphite powder from LIB anodes to form mixed products of Li_2CO_3 and the respective metals. In some cases, additional carbon is added based on different cathode stoichiometries to ensure complete reaction of the remaining cathode, allowing recovery efficiencies of Li and Co to exceed 95%.[31-32] In conventional pyrometallurgy, all the critical elements in the alloy and residual Li remaining in the slag can be extracted by acid leaching used in subsequent hydrometallurgical processes.

Hydrometallurgy. Hydrometallurgical processes mainly comprise of leaching and material precipitation steps after pre-treatment which does not involve high-temperature roasting (**Figure 2**). Compared to pyrometallurgy, it offers higher recovery efficiencies along with lower energy consumption and reduced toxic gas emissions. While these advantages make hydrometallurgy promising in the long term, it also faces limitations in dealing with the large amounts of waste generated.[2, 6] During leaching, critical metals in spent batteries are dissolved from solid powders typically by a strong acid.[1] Conventionally, acid-based leaching using reagents such as inorganic acids like HCl, H_2SO_4 are common due to their higher extraction efficiencies and ability to dissolve almost all transition metal oxides.[33-35] However, alkali leaching with ammonia based solutions such as ammonium carbonate ($(NH_4)_2CO_3$), ammonium sulfate ($(NH_4)_2SO_4$), ammonium chloride (NH_4Cl) allows for higher selectivity for specific critical materials such as Li, Co and Ni due to their ability to form metal-ammonia complexes (*e.g.*, $Co(NH_3)_6$) that can be easily recovered, leaving behind less valuable materials.[36-38] However, despite the high efficiencies and selectivity of either processes, both acid and alkali leaching processes still generate large amounts of waste solutions that are difficult to treat and dispose. To overcome this, use of organic acid (e.g., citric, lactic and succinic acids) leaching is instead preferred, which offers the same benefits of high leaching efficiencies but significantly reduces secondary pollution from the release of toxic fumes (Cl_2 / SO_x / NO_x) or wastewater often seen in their inorganic counterparts.[39-42] Additionally, these organic wastes are biodegradable and can be easily disposed safely after use.[6]

Precipitation and Cathode Production. After leaching, critical elements need to be extracted into useful precursors that can be then used to synthesize new cathode materials. This is commonly done via solvent extraction, where the organic and aqueous solution phases are separated to extract different metal ions based on their precipitation pH. Aside from pH, several other factors such as separation factors, extractant concentrations, phase ratio, temperature and extraction time can affect overall performance.[6] However, solvent extraction methods are often limited by their ability to extract only one metal of high purity at a time. As such, multiple extraction stages are often required to extract multiple metals that can dramatically increase capital costs of recovery.[43-44] An alternative approach is to adopt chemical precipitation instead, which utilizes the difference in solubilities of transition metal hydroxides or other compounds within the solution with the aid of specific precipitants such as sodium hydroxide (NaOH), sodium carbonate (Na_2CO_3) or oxalates ($H_2C_2O_4$) to induce metal precipitation.[45-47] In general, transition metals have lower solubilities compared to their equivalent Li compounds and can be extracted first. Once critical materials are recovered in high purities, they can then be used to resynthesize new battery materials with co-precipitation, sol-gel or solid state heat treatment steps in order to close the material recycling loop.

Despite much improvements in recovery efficiencies of critical materials in both pyrometallurgical and hydrometallurgical processes over the past decade, such destructive and re-synthesis of spent batteries materials are still energy inefficient, pollutive in nature, and consume disproportionately large amounts of input materials or energy that often result in high operation costs and additional waste generation. Given the understanding that most EOL EV batteries may still retain 80% of their original capacity, the embedded energy and economic value of the electrode materials would be lost if they are destroyed in either thermal or chemical processes. An ideal recycling process should seek to retain the core chemical structures of the materials without destroying them, and instead reconstitute or repair the deficient components at the atomic level. Such strategies are vital to produce profitable, high-value-added battery products and reduce the life-cycle energy consumption and GHG emissions of recycling processes.

■ Direct Recycling Approach

Direct recycling offers a strategy to recover critical materials without breakdown of its core structure, and without use of complicated chemical or physical treatment processes. The process allows the direct regeneration of spent cathodes and anodes into new active materials that can be directly used to assemble new batteries. Moreover, direct recycling allows the Li- containing liquid electrolytes to be extracted at the same time, increasing the overall recycling efficiency as a function of the entire cell.[48-50] In principle, dismantled spent batteries are first separated into their subcomponents and cathode powders extracted from the current collectors either by solvent dissolution to remove the binders, or thermal treatment to decompose organics within the electrodes. Solvent dissolution is typically done through the use of Dimethyl carbonate (DMC) and N-Methyl-2-pyrrolidone (NMP) to treat and separate electrodes from binders, salts and other additives used during the manufacturing process.[51] Alternatively, higher separation efficiencies can be achieved using thermal decomposition methods to remove the binder and carbon additives. However, this also introduces unwanted side reactions between the fluorine compounds generated from commonly used polyvinylidene fluoride (PVDF) based binders, and the highly sensitive transition metal oxide cathodes. To overcome this, mitigation strategies have been proposed, including the use of hydrated LiOH to counter the damaging effects of fluorine on the cathode structure.[52] After separation, the spent cathodes are then regenerated based on direct methods. As degradation of cathodes in spent batteries often occur as a result of Li loss during cell cycling, coupled with surface level structural decay (**Figure 3a & b**), the degraded cathode powders can be regenerated via re-lithiation and short sintering methods to recover their original Li content and phase purity.[51, 53] Note that direct regeneration of anode materials such as graphite is also viable, and have been reported to be highly effective using acid (HCl or H_2SO_4) along with thermal treatment to remove residual interface impurities.[54-55] Here, our discussion mainly focuses on the cathode materials that contain the majority of the critical material value in LIB materials.

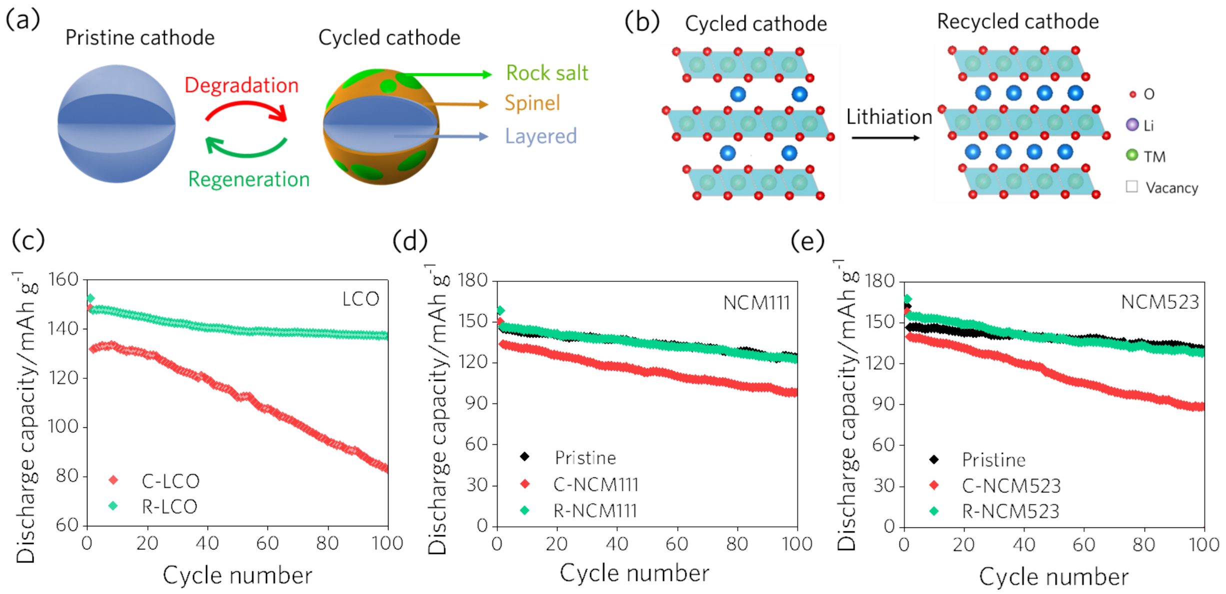


Figure 3. *Direct LIB cathode recycling using re-lithiation methods. (a) Schematic of cathode surface degradation during cell cycling. (b) Illustration of re-lithiation in spent cathodes. (c) Electrochemical performance data of before and after hydrothermal re-lithiation and short annealing of spent* $LiCoO_2$*, NCM111 and NCM523 cathode materials.*

Re-lithiation. Amongst various re-lithiation methods available, solid-state sintering with controlled amounts of Li source such as lithium hydroxide (LiOH) or lithium carbonate (Li_2CO_3) have been extensively reported.[38, 56] In a typical regeneration process, decayed cathode powders are isolated by a series of separation step as discussed earlier, then the Li amount within decayed cathodes are quantified via inductively coupled plasma mass spectrometry (ICP-MS). Subsequently, stoichiometric amounts of Li sources are mixed with the cathode powder with some excess and thermally annealed to induce re-lithiation and structural reordering. While costs of Li_2CO_3 precursors are lower than their equivalent LiOH amounts, higher sintering temperatures are typically needed to achieve re-lithiation in Li_2CO_3 due to differences in their melting temperatures (723˚C for Li_2CO_3 *vs*. 463˚C for LiOH). Nonetheless, both methods have been demonstrated to be equally effective in regenerating spent cathode materials such as $LiCoO_2$, $LiFePO_4$ and NCM111/523 (**Figure 3c**), with regenerated cathodes found to contain the original Li content and identical electrochemical performance compared to their pristine materials.[38, 57-58]

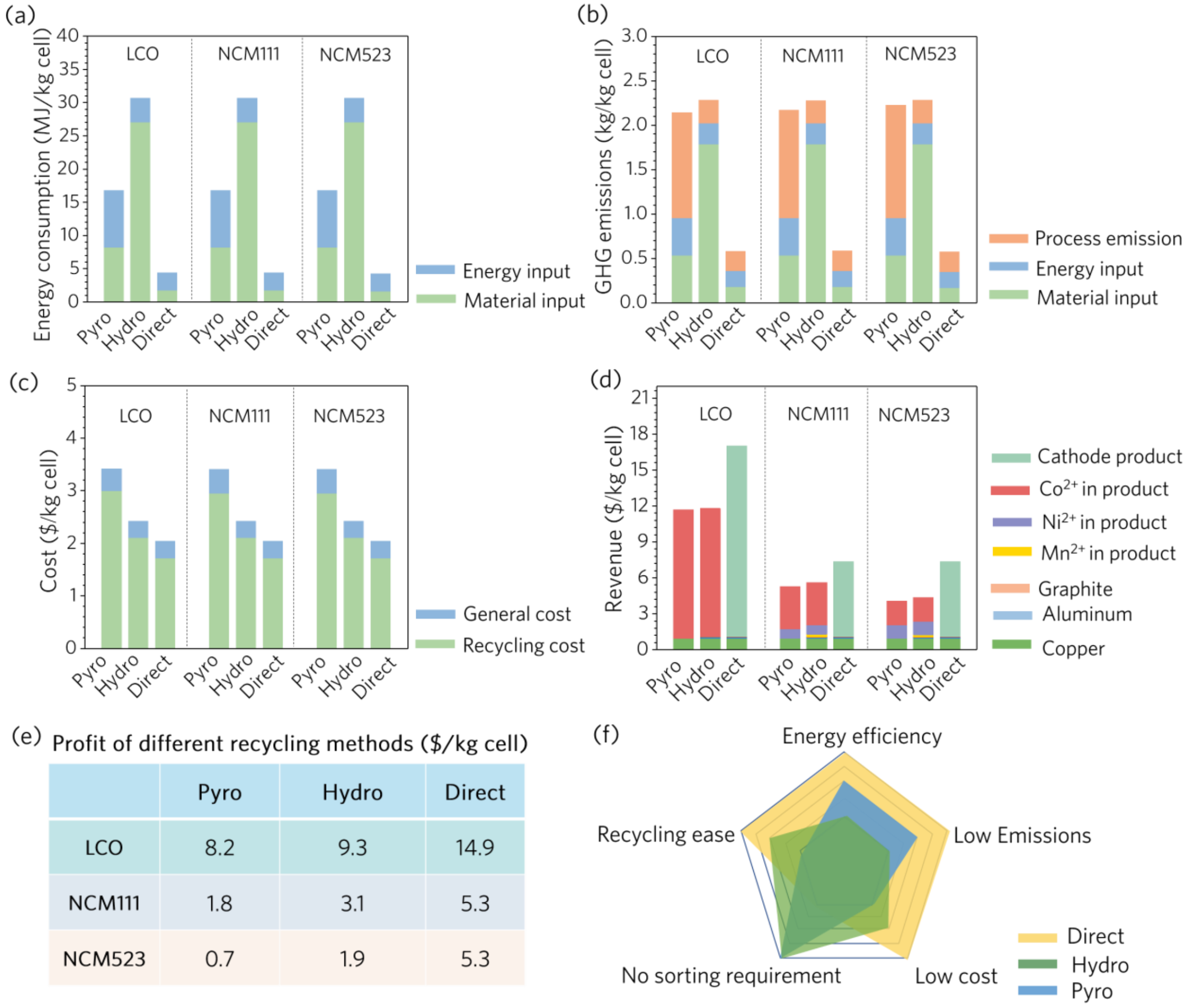
However, solid-state sintering approaches exhibit two major shortcomings: 1) Effectiveness against high-Ni type NCM cathode materials and, 2) Requirements for accurate determination of Li content in spent cathodes. Compared to high- Ni NCM materials (such as NCM622/NCM811), conventional cathodes such as those discussed earlier are more structurally stable as well as resistant to contaminative decay when exposed to moisture. As such, effective regeneration would mainly entail the re-lithiation components to compensate for Li losses during cycling as illustrated in **Figure 3b**. However, high-Ni NCM cathodes were found to break or pulverize during long-term cycling, and also undergo structural phase changes near the surfaces of the polycrystalline secondary particles, forming highly resistive spinel or rock salt phases as a result of Ni oxidation and cation mixing.[38, 51, 59-60] Additionally, spent batteries obtained for different sources and devices contain cathode materials of different degree of aging and Li loss, making it difficult to accurately determine the precise amount of Li source needed for re-lithiation in order to avoid introducing additional impurities into the sample batch.

To overcome this, hydrothermal re-lithiation would be a more robust method that can be applied to cathode materials harvested from large volumes of spent LIB instead. This method is non-destructive and can resolve the structural and Li deficiencies accumulated during long-term cycling. Conceptually, spent cathodes are immersed in an aqueous solution containing Li precursor (e.g., LiOH) and heated under pressure in an autoclave, before short annealing to achieve the regenerated materials. Hydrothermal re-lithiation can also be applied to cross chemical mixtures of cathodes.[59] For example, direct recycling process was previously applied to a 1:1 mixed ratio of $LiCoO_2$ and NCM111 spent cathodes, and found to exhibit identical electrochemical performance as its pristine form.[61] After regeneration, the aqueous solution with Li salts can also be reused for future processes, offering a low waste, energy efficient and cost effective approach for direct recycling. However, aqueous approaches may not be suitable to handle high-Ni NCM materials due to their sensitivity and reactivities toward moisture. As such, alternative methods such as electrochemical re-lithiation approaches can explored instead. While such techniques have yet to be applied to high-Ni NCM, their reported effectiveness in NCM442 and $LiCoO_2$ regeneration demonstrates the possibility to apply them for other moisture sensitive materials.[62-63] Unfortunately, the use of Li metal as the electrochemical Li source as well as additional organic liquid electrolytes presents both cost and safety constraints to such methods. To overcome this, molten eutectic salt direct chemical re-lithiation can be used instead to achieve avoid moisture and use of metallic Li sources for regeneration. In one example, re-lithiation of degraded NCM523 was achieved using eutectic solutions of $LiNO_3$ and LiOH at a molar ratio of 3:2. Such a specific formulation was chosen due to their lowest melting point at 175˚C, allowing re-lithiation to be achieved at relatively low temperature and under ambient pressures.[64] While these methods have yet to be demonstrated in ultra-high Ni NCM such as NCM811 yet, they offer a promising scalable approach to handle future large volumes of EV batteries.

Economic and Environmental Analysis. Regardless of approach taken, the key to any successful recycling strategy lies in achieving a balance of maximizing economic benefits while eliminating or minimizing waste generation. An ideal recycling design needs to have low energy consumption, low carbon footprint and require minimal capital tooling or materials inputs. Here, EveBatt model is used to conduct the economic and environmental analysis, which provides guideline to evaluate different recycling methods.[65] **Figure 4a** compares the energy consumption required to recycling 1 kg of LIBs based on various cathode material chemistries using the three main recycling approaches discussed earlier, pyrometallurgy, hydrometallurgy and direct methods. Contrary to the conventional view that pyrometallurgical methods are highly energy consuming, hydrometallurgical methods instead consume close to twice as much energy largely due to their high material inputs required. When the recycling process itself is considered (energy input component), hydrometallurgical methods would be perceived as being energy efficient. However, materials input also considers the upstream processes required to produce the large volumes of acids consumed in hydrometallurgy, as part of the entire life cycle analysis, resulting in the high overall energy needed.[2] By contrast, the main material input components of direct recycling, such as Li sources (LiOH, Li_2CO_3) are comparatively small in quantity needed to regenerate the same amount of cathode materials. In terms of GHGs, both pyrometallurgy and hydrometallurgy exhibit similarly high emissions compared to direct methods (**Figure 4b**). This is because direct recycling does not require the use of high carbon footprint smelting or toxic gas (SO_x, NO_x) emitting processes typically seen during acid synthesis. Additionally, direct recycling offers a pathway to recover and reuse liquid electrolytes from spent LIBs. This is typically done using supercritical CO_2 to enhance electrolyte dissolution properties between the liquid and gaseous phase of CO_2, allowing efficient extraction of lithium containing organic solvents within the liquid electrolytes.[48, 50] Using the EverBatt analysis, such methods were found to only introduce marginal increases in energy consumption and GHGs to the direct processes, offering a cost effective strategy to significantly increasing material recovery efficiencies as a function of the entire cell.

As shown in **Figure 4c**, both the energy and material inputs translate into the higher overall costs of recycling, where it comes as no surprise that direct recycling exhibits the lowest cost predicted. While the cost differences amongst the three methods may appear small, it is important to note that majority of battery recycling costs today come from variable components such as transport of spent batteries to centralized facilities, manual dismantling of modules, as well as other non-material level related costs.[66] From **Figure 4d**, it is clear that intrinsic value of output material is the main determinant of recycling revenue, with Co recovery making up the bulk of critical materials revenue. However, unlike the cobalt precursor materials generated from pyrometallurgy and hydrometallurgy that required additional cathode synthesis, direct recycling is able to place their output higher up the value chain, by directly producing new cathode materials that can be readily used to produce new LIBs. This translates into higher profit margins per kg of LIB cells recycled for direct methods (**Figure 4e**).

The spider chart in **Figure 4f** compares the pros and cons of each recycling method, while direct recycling is advantageous in all key aspects, it comes short in one crucial parameter: the need for a streamlined recycling infrastructure. Unlike conventional recycling methods which can be applied to LIBs of any chemistry or cell configuration, direct recycling requires spent batteries to be sorted by cathode materials (and ideally of similar age). However, LIBs today come without dedicated labelling or internationally consolidated sorting procedures needed to sort spent batteries from different devices or EV manufacturers. Additionally, a lack of regulatory framework for LIB collection along with the rapid technological developments in new LIB chemistries further complicate the prospects for direct recycling methods. Unless these challenges are addressed, adoption of direct recycling at a large industrial scale would not be practically achievable.



Profit of different recycling methods ($/kg cell)

	Pyro	Hydro	Direct
LCO	8.2	9.3	14.9
NCM111	1.8	3.1	5.3
NCM523	0.7	1.9	5.3

Figure 4. *Economic and environmental analysis of pyrometallurgy, hydrometallurgy and direct recycling methods based on recycling 1 kg of LIBs across different common cathode material chemistries. (a) Energy consumption, (b) GHG emissions, (c) Recycling costs, (d) Potential revenue from outputs produced, (e) Comparative profits generated. (f) Spider plot comparing various features of recycling methods discussed.*

Industry and Policy Impact. Challenges associated with the logistics of battery recycling typically occur before the materials recycling process itself. These include a lack of battery collection infrastructure, high costs of transport and potential safety hazards associated with long-term storage of aged battery packs.[11, 67] The high costs of transportation arises from the hazard classification of spent batteries (Class 9 Hazardous), which drastically increases costs (from \$0.14 \$/ton-mile to \$6.28 \$/ton-mile for non-hazardous) in standard trucking fees.[65] To reduce transport requirements, pre-treatment can be done at decentralized facilities to lower hazard classifications and reduce safety risks by fully-discharging collected batteries or conducting preliminary dismantling to reduce inactive mass during transport. While this can alleviate some transport and storage safety challenges, it would also pose obstacles in standardized spent battery sorting and dismantling of the wide varieties of cell chemistries without centralized economies of scale. To circumvent this, it is recognized and recommended that common standards for labelling should be mandated for used batteries, much like standardized recycling codes for plastics and metals that are internationally recognized and applicable to any recycling facility.[68-69] However, as LIB waste would require more information, such as its specific chemistry, state of health, origin manufacturer and other details, these can instead be summarized in a concise labelling system such as a bar code or tracking tag that allow facilities to sort them using automation. This would allow recyclers to immediately apply the most appropriate recycling processes to the pre-sorted batch of spent LIBs. At the same time, it can identify the "healthy" cells out of all LIBs collected that can be re-sold for secondary uses, generating additional revenue. However, such an undertaking would require the combined cooperation of original manufactures as well as regulatory institutions to incentivize both the improvement of battery collection infrastructure, as well as adopt newer recycling-friendly battery designs. Additionally, cross border applications and enforcement of such policies are also urgently needed, especially considering the reality that most of battery wastes are not generated where they are originally physically produced.

■ Improving Battery Design for Easier Recycling

Compared to the various battery recycling methods reported, relatively little attention has been placed on modifying existing battery designs to facilitate ease of recycling. At the module level, LIBs are fabricated using permanent assembly designs, making it challenging for automated dismantling and often require specialized technicians to manually disassemble.[5] Considering the multiscale barriers caused by today's LIB design towards recycling the critical materials inside, manufacturers should consider using modified assembly protocols that would ease module level dismantling, such as adoption of common quick release locking mechanisms instead of conventional metal-metal welding during pack integration. At the pack level, permanently sealed stainless steel cylindrical cans or plastic prismatic packaging cases also require tedious manual dismantling of individual customized cells by hand, posing safety hazards for workers.[5] These designs can be replaced by removable screw-off caps or incorporate perforated grooving within plastic casing similar to those used in consumer product packaging for ease of disassembly.[70] Alternatively, standardized form factors can be adopted for pack level components such that disassembly can be done with automation to mitigate hazards faced by workers. Taking such strategies will also enable ease of recycling less valuable materials in spent batteries such as plastic and metal casings used at the cell to pack level. Melting and remolding of old plastic and metals harvested into new cell parts for reuse are commonly done in lead acid battery recycling and can be extended to LIBs as well.[71] However, this will also require shared responsibility and cooperation of battery manufacturers backed by legislative mandates to encourage recycling-friendly packaging designs. While improved designs at the pack and module level can significantly improve the logistics of battery collection and dismantling, they mainly pertain to the inactive components of the spent LIBs, additional focus needs to be diverted to the cell and electrode level designs, which is where the critical materials are harvested.

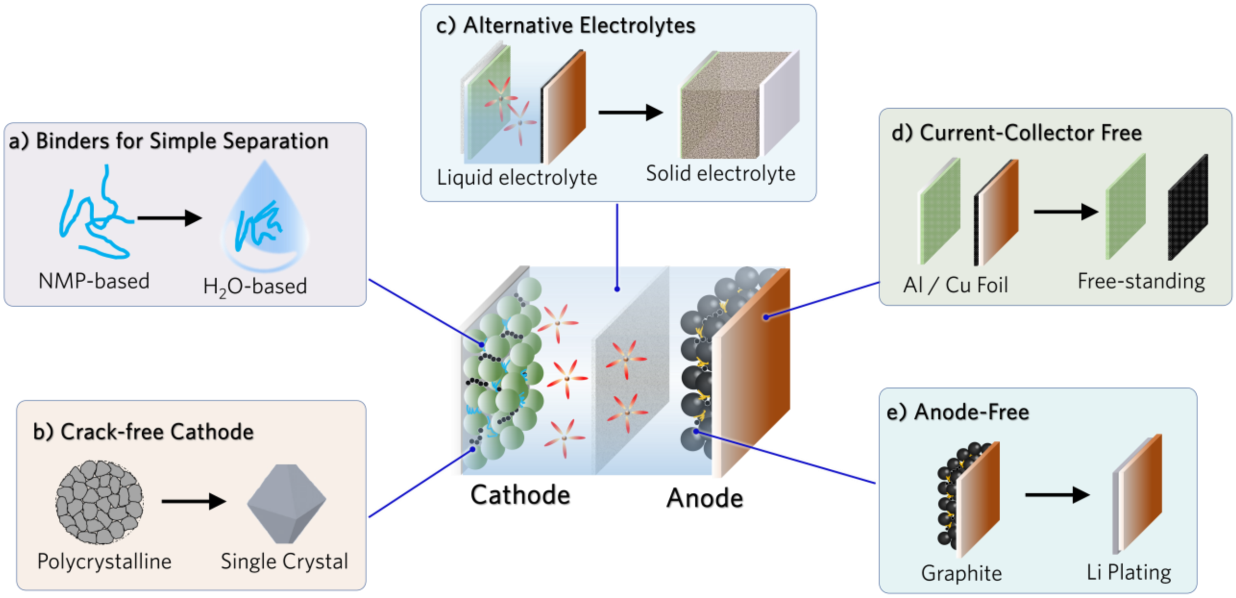


Figure 5. *New recycling friendly design for LIBs. (a) Use of carbon based current collectors eliminate need for additional metal recovery. (b) High aspect ratio carbon such as fibres or nanotubes can reduce carbon content while maintaining electronic pathway. (c) Solid electrolytes offer pathway for ease of electrolyte direct recycling. (d) Eliminate toxic chemicals in binder processability and material exfoliation during recycling. (e) Single crystals aid in re-lithiation processes. (f) Anode-free configurations to avoid graphite handling challenges in spent batteries.*

Cell and Materials Designs. For new recycling friendly designs to be successful, it must achieve both good electrochemical performances, and enable simple and cost-effective materials level separation for subsequent direct recycling processability (**Figure 5**). One potential approach would be to adopt new binder compositions that allow quick exfoliation of materials from current collectors during cell separation. These include use of water soluble binders in place of conventional NMP which is both toxic and difficult to handle.[56] While water-based solvents have been reported to be detrimental to high-Ni NCM type cathodes[72], specialized protective coatings (Al_2O_3, ZrO_2) have been also demonstrated to improve both the moisture resistance and overall electrochemical performance of these cathodes.[73] Alternatively, alcohol based binders can also be used to achieve a compromise between moisture resistance and ease of binder removal during pre-treatment.[74] Ideally, binders should avoid fluorine containing moieties to allow for efficient material separation without the need for any solvent.[52] At the cathode, promising alternative approaches include the use of single crystal type materials which are easier to directly regenerate in place of the existing polycrystalline type particles which often experience particle cracking during calendaring and cell cycling.[75-76] While understandably higher in upfront cost, higher reversible capacities and cycle performance typically achieved in single crystal type cathodes may compensate for this in the long term.[77]

To overcome stability challenges faced in aqueous based regeneration of high-Ni NCM cathodes, non-Ni based cathodes can be used as well. Doping of other transition metals such as titanium (Ti), magnesium (Mg), and Al have been shown to increase the reversible capacities of $LiM_{1-x}Co_xO_2$ (where M is dopant) to more than 200 mAh/g and voltage cut-off up to 4.6V while maintaining cycling stability, making them comparable to high-Ni NCM materials in terms of energy density.[78-79] At the anode, presence of graphite-based electrodes introduce additional steps to separation and critical material recycling especially where carbon impurities are present. To overcome this, anode free cell configurations are a highly promising approach to entirely eliminate the need for anode within the cell, as well as dramatically increase overall cell energy density and performance at the same time.[80-81] Regardless of any new materials chosen, to re-design batteries catered for recycling would require existing manufacturers to change their production protocols, potentially incurring backlash to profitability. As such, it is also important to develop potential recycling approaches for next-generation battery types as well, so that future manufactures can incorporate sustainable recycling friendly designs to new batteries at the onset.

New Battery Chemistries. All solid-state batteries that use solid-state electrolytes in place of conventional organic liquids have been gaining much popularity in recent years due to its potential for energy dense, safe and environment-friendly designs.[82-83] Additionally, all solid-state batteries offer the potential for improved recycling safety due to absence of fire hazards, and ease of material separation and recovery compared to conventional LIBs. As sulfide-based solid electrolytes can be readily dissolved in safe and low cost solvents such as alcohols[84-85], spent batteries can be fully dissolved before separation using gravity based techniques. The precipitated cathode materials can then be recycled using re-lithiation methods discussed earlier. Additionally, the recovered solid-state electrolyte can also be directly regenerated in useful formats using short annealing to regain its original ionic conductivity and crystal structure.[82, 86] Likewise, recently commercialized sodium-ion batteries (SIBs) have also shown much promise for large scale grid storage applications.[87-89] A recent study demonstrated a closed-loop sustainable recycling method to recycle high power and long cycle stable $Na_3V_2(PO_4)_3$ cathodes in a SIBs using a solid-state sintering approach.[90] Similarly, addition of sodium source such as low cost and highly abundant soda ash (Na_2CO_3) or sodium hydroxide (NaOH) combined with sintering can potentially enable direct regeneration of promising SIB cathodes such as $Na_{0.85}Li_{0.12}Ni_{0.22}Mn_{0.66}O_2$ or $Na_{2/3}Fe_{1/2}Mn_{1/2}O_2$ to compensate lost sodium during the cell's cycle life.[91-93]

■ CONCLUSIONS

Sustainable energy storage is undoubtedly becoming a core economic driver of the 21st century. With rising production of EVs and other LIB powered devices, battery recycling and the recovery of critical materials will inevitably play an increasingly important role to mitigate the shortage of raw materials. As global reserves of critical materials shrink, spent LIBs would eventually become the default resource for material harvesting, serving as the new source of continuous and diversified supply of critical materials. Conventional methods to recover these scarce resources are discussed, as well as rational direct recycling approaches to recover them in cost-effective and environment-friendly ways. The relative economic and environmental impacts of various recycling approaches are analyzed, with an emphasis on the importance of enabling direct recycling methods in achieving profitable battery recycling. Potential solutions to address the materials and logistical limitations of direct recycling are reviewed, including the importance of battery labelling and an internationally recognized tagging system. Key policy and industrial directions are also discussed to address the lack of recycling infrastructure today, including improving the transport and handling of spent batteries, as well as the need to design batteries with recycling ease in mind. Finally, promising modifications to existing LIBs are proposed to promote recycling ease, and how these can be applied to next generation batteries. We believe this perspective will offer pathways for manufacturers and researchers alike, to adopt new sustainable battery production and recycling processes in the future.

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