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Lithium-Ion Battery Recycling—Overview of Techniques and Trends

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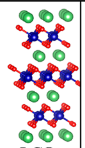
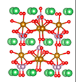
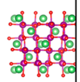
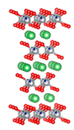
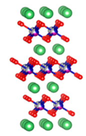
From their initial discovery in the 1970s through the awarding of the Nobel Prize in 2019, the use of lithium-ion batteries (LIBs) has increased exponentially.[1−4] As the world has grown to love and depend on the power and convenience brought by LIBs, their manufacturing and disposal have increasingly become subjects of political and environmental concerns.[5,6] World reserves of lithium, cobalt, and other metals are limited and unevenly distributed, while their mining is energy and labor intensive and creates considerable pollution.[7,8] More than 70% of the world's cobalt comes from Congo,[9] with no other country producing more than 5%. China and Mozambique produce 70% of the world's natural graphite, an important material for anodes.[10] As a result, natural disasters, war, or resource allocation decisions may also change the availability of these materials.

Resource scarcity and supply are particularly important due to the short device lifetime, whether from design obsolescence, "upgrades" to newer smartphone models, or, quite often, the LIB nearing the end of its own life. By most accounts, most discarded LIBs eventually are landfilled or stockpiled, contaminating the land while wasting energy and non-renewable natural resources. As of February 2019, there were over 5.6 million electric vehicles (EVs) in the world, a 64% increase from 2018.[11] By 2040, 58% of all cars sold worldwide are anticipated to be EVs.[12] With explosive growth in EV numbers combined with the sheer sizes of their batteries (Tesla Model 3 Long Range's battery contains 4416 cells and weighs 480 kg),[13] significant LIB waste *is and will be generated every year* which, if not recycled and reused, will exert massive environmental impacts and accelerate the depletion of mineral reserves. Adding to the recycling difficulty, LIBs are complicated structures comprising one of five common cathodes, an anode, electrolyte, a separator, and current collectors along with packaging components (Table 1, Table S1). The International Energy Agency, for example, estimates that electric vehicles produced in 2019 alone generated 500,000 tons of LIB waste, and the total amount of waste generated by 2040 could be as much as 8 million tons.[14]

In this article, we summarize and compare different LIB recycling techniques. Using data from CAS Content Collection, we analyze types of materials recycled and methods used during 2010−2021 using academic and patent literature sources. These analyses provide a holistic view of how LIB recycling is progressing in academia and industry. The benefits and challenges of LIB recycling from economic and environmental perspectives are also discussed. Finally, we provide a global overview of established and planned recycling facilities and their reported capacities.

Table 1. Cathode Materials Used in Commercial LIBs and Their Economical Recycling Methods[a15−18]

Structure	 LCO	 LFP	 LMO	 NCA	 NCM
Composition	$LiCoO_2$	$LiFePO_4$	$LiMn_2O_4$	$LiAl_xCo_yNi_{1-x-y}O_2$	$LiCo_xMn_yNi_{1-x-y}O_2$
Energy Density/ Wh kg^{-1}	624	544	410	740	592-740
Material price/ \$ (kWh)$^{-1}$	88	32	26	39	40-50
Battery price/ \$ (kWh)$^{-1}$	357	222	251	199	145-230
Publications on Direct Recovery	110	98	43	9	56
Economical Recycling Pathways	H, P, D	D	D	H, P, D	H, P, D

[a]cyan **H**: hydrometallurgy; red **P**: pyrometallurgy; black **D**: direct recycling).

PUBLICATION TRENDS

The CAS Content Collection is the world's largest repository of chemistry-related information and is particularly useful for quantitative analysis of publications against variables such as time, country, and research area, as well as their substance

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details. Here, we searched for publications related to LIB recycling (see Supporting Information (SI) for full methods and limitations). A pool of 3596 documents were analyzed based on their publication type and publication year (Figure 1). While the global scientific publication volume has been steadily increasing in the past decade, we found that the annual volume growth in publication on this topic (32%) far exceeds that of overall scientific publications (4% annually),[19] suggesting an emerging interest in recycling, especially in the last 4 years. We also found that patent applications account for 74% of the Li-ion battery recycling literature, whereas patents are outnumbered by journal articles 2:1 in the entire CAS Content Collection, showing the high commercial value of technologies and discoveries around LIB recycling. An analysis of the countries/regions associated with the affiliated organization of these documents shows that China has the highest publication volumes by far in both journals and patents (Figure S1). A further analysis of patent assignees revealed the top organizations by volume of patent applications on LIB recycling (Table S2) are primarily located in China, Japan, and France.

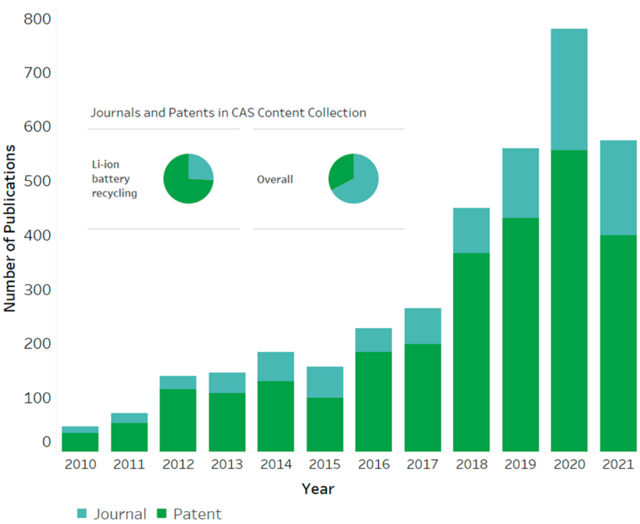

Figure 1. Journal articles and patent publications on Li-ion battery recycling (data for 2021 is partial). Inset shows relative publication volumes of journal articles and patents in Li-ion battery recycling (left) and in the chemical literature as a whole (right).

■ LIB RECYCLING METHODOLOGIES AND INDUSTRIALLY IMPLEMENTED METHODS

Due to the complex structure and number of materials in LIBs, they must be subjected to a variety of processes prior to reuse/recycling. LIBs must be first classified and most often pretreated through discharge or inactivation, disassembly, and separation after which they can be subjected to direct recycling, pyrometallurgy, hydrometallurgy, or a combination of methods, as shown in Figure 2.[18]

Direct methods, where the cathode material is removed for reuse or reconditioning, require disassembly of LIB to yield useful battery materials,[22] while methods to renovate used batteries into new ones are also likely to require battery disassembly, since many of the failure mechanisms for LIB require replacement of battery components. Reuse of LIB in stationary applications will require battery classification and the determination of charge state and capacity. Publication volume on direct methods (Table 1) shows the most frequently used cathode materials LCO and LFP have been primarily studied,

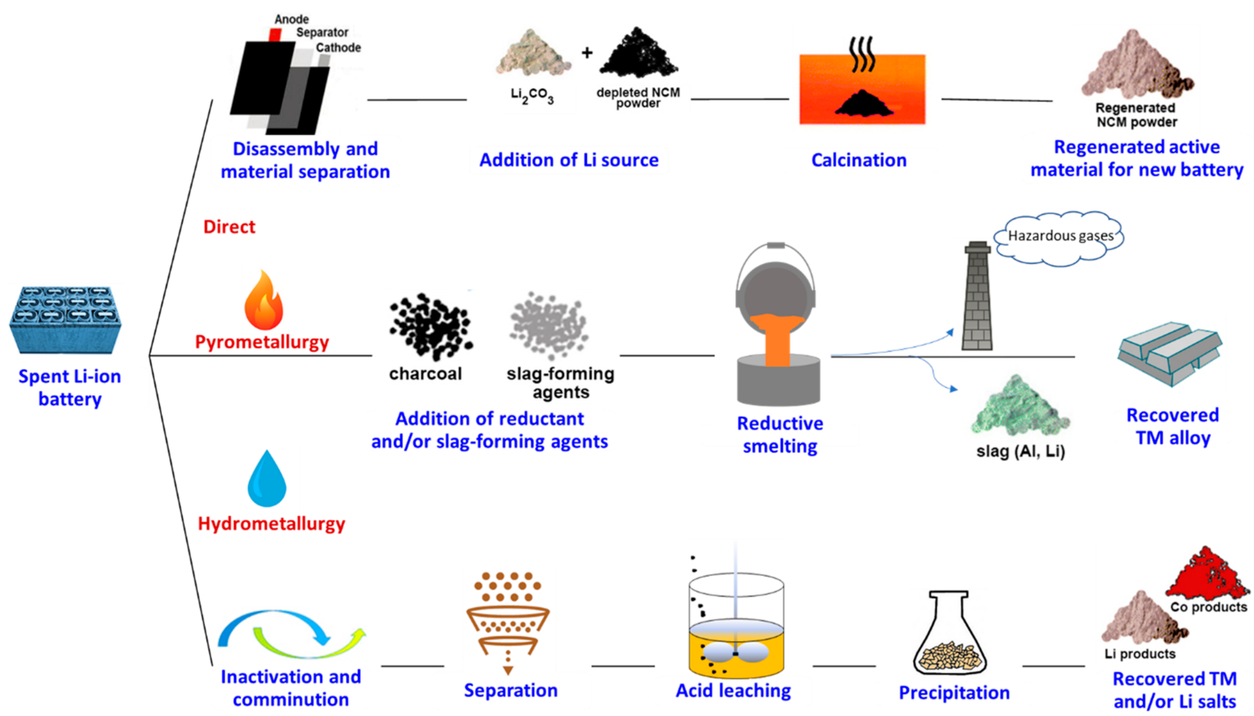

Figure 2. Typical direct, pyrometallurgical, and hydrometallurgical recycling methods for recovery of Li-ion battery active materials. From top to bottom, these techniques are used by OnTo,[15] Umicore,[20] and Recupyl[21] in their recycling processes (some steps have been omitted for brevity).

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Table 2. Companies Involved in LIB Recycling and Volumes Processed by Technique[27−42]

label	company	location	volume (tons/year)	method	status
1	ABT	Fernley, NV, US	20,000	Unknown	Planned
2	Retriev (Toxco)	Trail, BC, CA	4,500	Hydro	Established
3	Li-Cycle	Gilbert, AZ, US	10,000	Hydro	Planned
4	Ganfeng Li	Sonora, MX	Unknown	Unknown	Planned
5	Li-Cycle	Tuscaloosa, AL, US	10,000	Hydro	Planned
6	Inmetco	Elwood, PA, US	6,000	Pyro	Established
7	Li-Cycle	Rochester, NY, US	5,000	Hydro	Established
8	Li-Cycle	Kingston, ON, CA	5,000	Hydro	Established
9	Fenix	Whitehall, UK	10,000	Hydro	Planned
10	Valdi	Commentry, FR	20,000	Pyro	Established
11	Umicore Valeas	Hoboken, BE	7,000	Pyro/hydro combo	Established
12	Recupyl	Grenoble, FR	110	Hydro	Established
13	Accurec	Krefeld, DE	4,000	Pyro/hydro combo	Established
14	Glencore	Baar, CH	3,000	Pyro/hydro combo	Established
15	Redux	Offenbach, DE	50,000	Pyro	Established
16	Northvolt	Frederikstad, NO	8,000	Unknown	Planned
17	Fortum	Harjavalta, FI	Unknown	Unknown	Planned
18	Akkuser	Nivala, FI	4,000	Pyro/hydro combo	Established
19	Green Li-ion	Singapore	Unknown	Unknown	Planned
20	Brunp Recycling Technologies	Hunan, CN	100,000	Pyro/hydro combo	Established
21	Taisen	Hunan, CN	6,000	Hydro	Established
22	GEM	Jingmen, CN	30,000	Hydro	Established
23	Guanghua Sci-Tech	Guangdong, CN	12,000	Preprocessing	Established
24	Gotion High-Tech	Hefei, CN	Unknown	Unknown	Planned
25	Quzhou Huayou	Quzhou, CN	40,000	Pyro	Established
26	Tesla	Shanghai, CN	Unknown	Unknown	Planned
27	SungEel HiTech	Gunsan, KR	8,000	Hydro	Established
28	Posco Hy Clean Metal	Gwangyan, KR	12,000	Unknown	Planned
29	JX Nippon Mining	Tsuruga, JP	5,000	Pyro/hydro combo	Established
30	Dowa Eco-System	JP	6,500	Pyro	Established
31	Sumitomo/Sony	Namie, JP	150	Pyro	Established
32	Envirostream	Melbourne, AU	3,000	Preprocessing	Established

but methods for automated battery disassembly are still limited in scope and volume.[23,24]

Pyrometallurgy uses heating to convert metal oxides used in battery materials to metals or metal compounds.[25] In reductive roasting (smelting), the battery materials (after pretreatment) are heated under vacuum or inert atmosphere to convert the metal oxides to a mixed metal alloy containing (depending on the battery composition) cobalt, nickel, copper, iron, and slag containing lithium and aluminum. Pyrometallurgical methods require simpler pretreatment methods (most often shredding or crushing) to prepare batteries for recycling and require fewer different methods to recycle LIB of differing compositions, shapes, and sizes. Lithium is recyclable by some pyrometallurgical methods,[26] but the methods are most effective for particularly valuable metals such as cobalt.

Hydrometallurgical methods use primarily aqueous solutions to extract and separate metals from LIBs. The pretreated battery materials (with Al and Cu current collectors previously removed) are most often extracted with H_2SO_4 and H_2O_2, although HCl, HNO_3, and organic acids including citric and oxalic acids are commonly used. Once metals have been extracted into solution, they are precipitated selectively as salts using pH variation or extracted using organic solvents containing extractants such as dialkyl phosphates or phosphinates.

In many cases, combinations of hydrometallurgical and pyrometallurgical methods are used to process lithium-ion batteries today (Table 2).[27] Pyrometallurgical methods are likely used because they allow flexibility in battery feedstock (the Umicore method is used for both lithium-ion and nickel metal hydride batteries) and due to fixed investment in existing facilities. Methods in development, on the other hand, rely on hydrometallurgy to a larger degree, at least in part because the cost of facilities to implement the methods is smaller. Lithorec and Aalto University (Finland) both have devised hydrometallurgical methods, while Accurec, Battery Resources, and OnTo use both hydrometallurgical and pyrometallurgical methods.

To compare the popularity of different recycling methods, publication numbers over the past decade were analyzed and shown in Figure 3. Lithium is the most frequently recovered element, followed by metals in the order Li > Co > Ni > Mn > Fe. The trend agrees well with the price differences among the virgin metals except that between Li and Co. Beside economic motivations, the high publication volume for Li recovery can likely be attributed to the prevalence of Li in all LIB cathodes, resulting in studies of Li recovery being present in most publications on LIB cathode recycling. Publication volume of hydrometallurgical or pyrometallurgical methods for recovering metals follows roughly the same trend (Figure 3), while total publication volume has been exponentially rising and is now greater than 500 per year (Figure S3). Combinations of hydrometallurgy and pyrometallurgy were used in most studies regardless focused on metal recovery.

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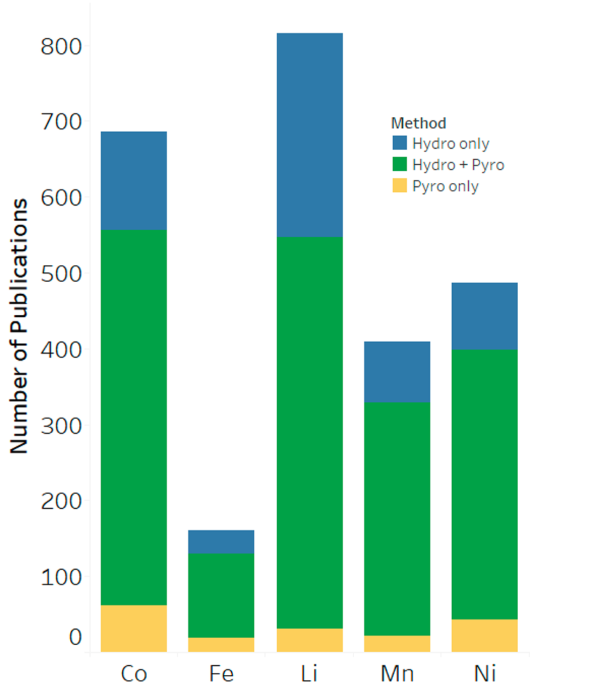

Figure 3. Prevalence of metals recovered in documents discussing hydrometallurgy and/or pyrometallurgy.

ENERGY/ENVIRONMENTAL IMPACTS

To understand how recycling may be able to decrease the effects and costs of battery recycling, the materials used in batteries and their costs should be defined, and the cost of new materials and recycled materials compared. Mining and refining of virgin materials and recycling used materials for batteries exact environmental costs. As an example, 1 ton of virgin lithium requires 250 tons of ore or 750 tons of brine.[43] While refining brine requires less energy than refining spodumene, it requires 18−24 months, yields lower grade lithium, and recovers less of the lithium present in brine than is recovered from ore.[44] In addition, water use is a concern; 65% of the water in Chile (one of the major sources of lithium) is consumed by the mining industry.

Recycling also has environmental costs including transportation, preparation, and high energy use. Kim et al., for example, noted that "the techniques that are widely commercialized or researched are also environmentally harmful".[26] Pyrometallurgical methods are implemented relatively simply, but incur environmental and significant energy costs for combustion and calcination of the batteries. While hydrometallurgical methods require less energy for processing than pyrometallurgical methods, many reagents are required and water must be purified afterward.[45]

Given the costs of making batteries, recycling battery materials can make sense. From the estimated 500,000 tons of batteries which could be recycled from global production in 2019, 15,000 tons of aluminum, 35,000 tons of phosphorus, 45,000 tons of copper, 60,000 tons of cobalt, 75,000 tons of lithium, and 90,000 tons of iron could be recovered.[46] These quantities of materials can reduce the need to mine new materials and also allow countries to reduce their dependence on other countries for battery supplies.

Several barriers exist to battery recycling with maximal efficiency, safety, environmental benefit, and economic return.[18] While the highest value is obtained from cathode recycling, a more holistic and sustainable approach considers all components of LIBs.[47] We quantified publications focusing on lesser-studied components in Figure 4. The most studied

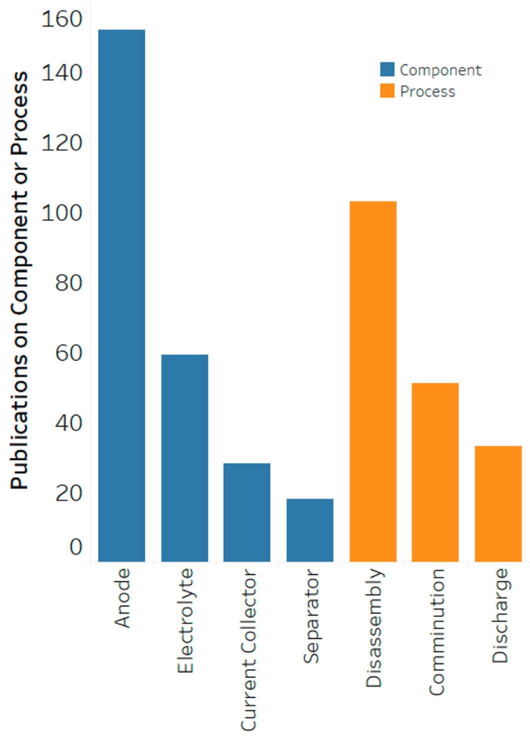

Figure 4. Publications studying recovery of noncathode materials and process optimization for recycling.

noncathode component is the anode, followed by the electrolyte, current collectors, and separator. While each of these four components comprise similar material costs in batteries,[48] the relative difficulty of purifying each respective material and their respective secondary values likely influence publication focus.

To maximize the amount of recyclable material, disassembly is preferable to comminution of the whole battery; Figure 4 shows that considerable research effort has been made toward disassembly relative to grinding/shredding. Additionally, treatment of wastewater produced by hydrometallurgy is an active area of study and invention. The safe discharge of discarded batteries has also received attention.

With a potential economic benefit, the likelihood of battery recycling on a large scale is improved. The value of materials obtained from battery recycling determines the economic benefit of recycling.[27] Offer et al.[16] discuss the economics of LIB recycling in various countries. Depending on the assumptions made, the costs of transporting LIB for recycling can make up either 2−13% or 5−70% of the costs of recycling; local recycling (for example, in Europe) has significantly lower transit costs. On the other hand, battery disassembly costs can make up 2−17% of battery recycling costs; since disassembly costs depend strongly on labor costs, disassembly is likely to be cheaper in countries with lower labor costs. While China no longer accepts materials from other countries for recycling, other countries may be willing to recycle battery materials, and the balance between transport and labor costs helps to determine the benefit of LIB recycling. Furthermore, because waste streams with complex mixtures complicate separation

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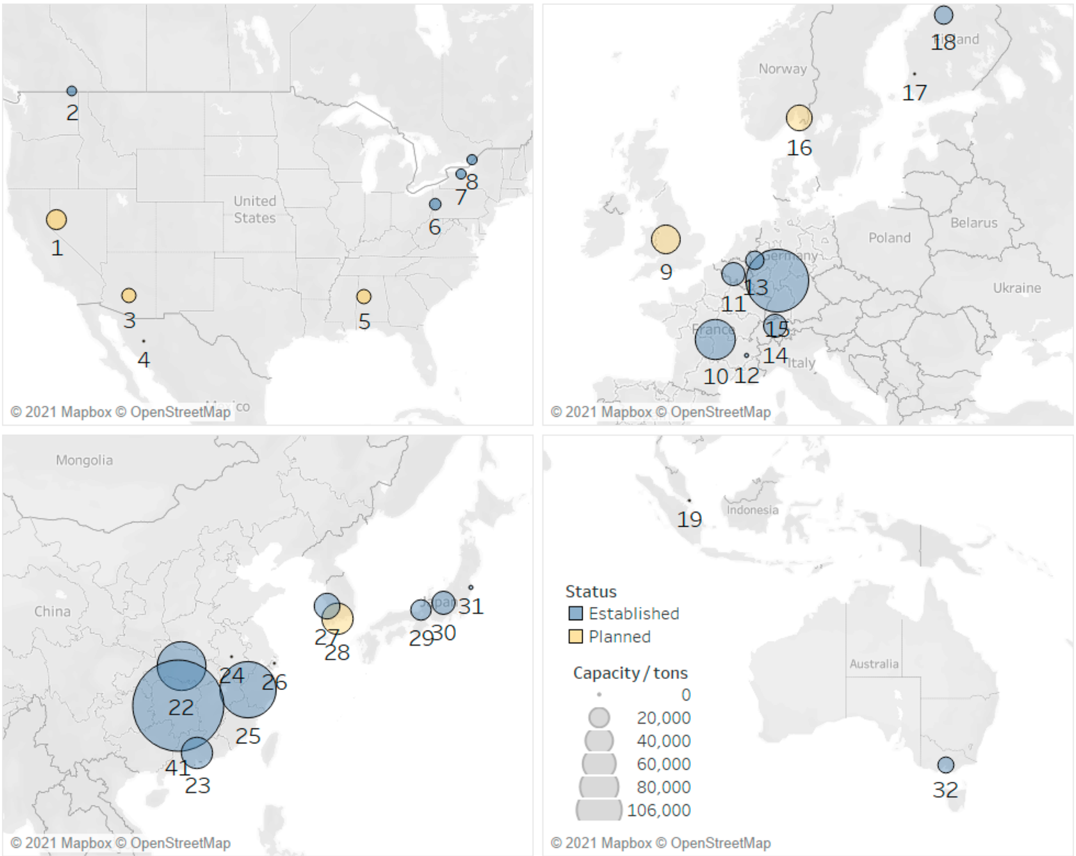

Figure 5. Established and planned global Li-ion battery recycling facilities as of November 2021.[27−42,57]

processes, the accurate classification and sorting of batteries by cathode chemistry is important for their profitable recycling.[49] These considerations aside, the methods generally found to be profitable for each cathode material are shown in Table 1.[16,18] The current LIB recycling market is estimated to be worth approximately $1700 million[50] and is expected to increase significantly over the next ten years.[51] The most commonly cited figure for LIB recycling is that approximately 5% of LIB are currently recycled at the end-of-life;[52] however, the proportion of recycled LIB may be as high as 50%.[53]

Many LIB recycling benefits, as noted by Offer et al., are not readily monetizable. Environmental and economic benefits differ over time, including energy and greenhouse gas (GHG) emissions saved by recycling, due to variations in recycling method, the development of new recycling methods, maintenance costs, changes in the costs and sources of feedstocks and energy, battery composition, and improvements in modeling.[28,47,54,55] Complicating the picture is that optimal recycling methods depend on battery type; NCM batteries are better suited to hydrometallurgical methods because of the need to separate multiple metals, while pyrometallurgical methods are simpler for recycling LMO and LFP batteries (see also SI).[18]

Direct recycling is likely to have lower energy costs and reagent costs than other methods. While calcination may be required to generate useful battery materials, the energy requirements are likely to be lower than for either pyrometallurgical or hydrometallurgical methods. Direct recycling also has lower fixed facility costs than other methods. Reconstitution of the cathode material may require added reagents, but likely far less reagent and solvent than what is required for hydrometallurgy. Direct recycling also allows recyclers to recover all components from LIB and may allow the cathode material to be retrieved with intact crystal structure appropriate for use in LIB. However, direct recycling has some disadvantages. It requires that LIB be in good condition for recycling. Different methods are required to reconstitute batteries based on the battery type and composition. Directly recycling batteries thus requires that either the selection of acceptable batteries be reduced to reduce the facility and complexity costs for multiple battery types, or increased facilities and labor must be available to recycle input LIB. The requirement for manual disassembly means that larger labor costs will be required than other methods; while automation and artificial intelligence have been studied for battery recognition, sorting, and disassembly, the methods for automated battery disassembly are limited in scope and volume.[23] Nevertheless, direct recycling is receiving significant global interest toward mitigating the negative environmental impacts of pyro- and hydrometallurgy; dedicated research efforts have been established in multiple major institutions including the ReCell Center at Argonne National Laboratory[56] and ReLiB at The Faraday Institution.[24]

One of the limitations, however, on direct recycling as an economical recycling method for LIB is the need for manual disassembly of batteries. Pyrometallurgy and hydrometallurgy use mechanical pretreatment methods but provide materials that may not be compatible with facile reuse in batteries; the methods also consume some of the battery materials so that they cannot be directly incorporated into new batteries. Direct recycling yields battery materials that can readily be reused in new batteries, requiring lower material and energy costs.

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However, LIB are used in many applications with a variety of designs and energy requirements, making standardization of chemistries and packaging difficult. In applications with similar requirements (such as automobiles),[22] LIB have a variety of sizes and are aggregated in different ways, and the variety of chemistries and designs for LIB make manual classification and disassembly necessary for direct recycling of LIB. The requirement for manual classification and disassembly of LIB is likely to be a significant contributor to the cost of direct recycling, which has been estimated to be similar to that of pyrometallurgy at >20,000 t scales.[16] Standardization of the design and labeling of LIB for use in common applications, such as in vehicles, would improve the economic viability of direct recycling and reduce the overall environmental costs of LIB recycling. This could include, for example, common designs for smaller batteries and common methods for aggregating them into larger units for use in vehicles. Common labeling specifications would also improve the efficiency of battery sorting and enabling recovery of materials in LIB recycling that might not be currently economical to recover.

OUTLOOK FOR LIB RECYCLING FACILITIES

Given an annual LIB production volume projected to exceed 1 million tons by 2025,[18] additional LIB recycling facilities will be needed. As shown in Figure 5 and Table 2, there are at least 32 established or planned facilities for LIB recycling with roughly 322,500 tons of recycling capacity (as of late 2021) and approximately 70,000 tons of planned recycling capacity (although the capacities of 4 of the 12 planned facilities are not known).

East Asia has nearly two-thirds of the current LIB recycling capacity, with 207,500 tons of battery recycling capacity and nine established and two planned facilities. Five of the established capacities are in China, with a total capacity of 188,000 tons; a facility (with unspecified capacity) is planned for Hubei Province. Three established recycling facilities operate in Japan with a total capacity of 21,500 tons, and one facility with 8000 ton capacity is operating in South Korea (with a second facility planned to bring total capacity to 20,000 tons). The largest battery recycling facility in the world, with 100,000 ton capacity, is operated by Brunp Recycling Technologies in Hunan Province, China.

Europe has the second largest set of active battery recycling facilities, with seven facilities and a total of 92,000 tons of capacity. The plants are evenly spread among the United Kingdom, France, Belgium, Switzerland, and Germany. Three recycling facilities have been planned, with one each in the U.K., Finland, and Norway and a total capacity of greater than 18,000 tons (the future capacity of the Finnish facility has not been specified).

North America has four battery recycling facilities operating with a total capacity of 20,500 tons; Canada and the United States have two facilities each, with similar total capacities. Five facilities are planned with a total capacity of greater than 40,000 tons. Three facilities with a total of 40,000 tons capacity are planned for the United States, while one facility with unspecified capacity is planned for Sonora, Mexico. Finally, Australia has one recycling facility with a 3000 ton capacity, while a recycling facility (with unspecified capacity) is planned for Singapore.

Overall, more than two-thirds of the current recycling capacity is in China, and approximately 90% of recycling capacity is concentrated in Europe and East Asia. As noted above, the planned facilities will increase total LIB recycling capacity to nearly 400,000 tons of batteries; while East Asia and Europe will have the largest battery recycling capacities (with more than 219,500 and 110,000 tons of capacity, respectively), the battery recycling capacity of North America is likely to more than double to more than 60,000 tons.

In conclusion, while LIB recycling technology is partially established, additional technical discoveries are being researched today and are needed to continue to improve recycling processes. For researchers in the field, reading both the patent and academic literature is important due to the prevalence of industrially relevant work. New discoveries will feed the rapidly expanding recycling industry to help conserve resources and provide global sustainability.

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ASSOCIATED CONTENT

Supporting Information

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Schematic diagram of lithium-ion battery (LIB), description of LIB components, background on aging, LIB recycling publications by country/region, top LIB recycling patent assignees, costs and benefits of LIB recycling, methods for recycling LIB, LIB recycling publication volume by year and method, and search strategy and limitations (PDF)

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Notes

Views expressed in this Energy Focus are of the authors and not necessarily the views of the ACS.
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