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Economics and Challenges of Li-Ion Battery Recycling from Endof-Life Vehicles

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Abstract

This study sheds light on current and future recycling methods for spent Li-ion batteries from retired vehicles. The demands of Li-ion batteries for automotive applications and power electronics are expected to increase significantly in the next 15-20 years. Recycling cathode materials from end-of-life batteries provides a sustainable source of materials, and offers an economic alternative for some of the high value elements such as cobalt and nickel. Insights and directions for future R&D will be presented in this paper based on the results of the supply chain and techno-economic analyses made for end-of-life li-ion batteries.

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Keywords: Cathode Materials; Environment; Li-ion Battery; Recycling

1. Introduction

Lithium ion batteries (LIB) are a category of batteries of various chemistries that include lithium as a primary cathode and electrolyte component. The primary use of LIB has, until recently, been in consumer electronics. However, the use of LIB in battery electric vehicles (BEVs) and large-scale electricity storage applications has led to rapid growth in the market for LIB, and consequently, their constituent materials. In addition to lithium, critical materials used in LIB include cobalt and graphite. Lithium and cobalt are scarce and mined in only a few countries, potentially creating risk in their supply and availability. The increased demand represented by the widespread adoption

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of electric vehicles, grid electrical storage, and other uses could increase prices impacting electric vehicle and other markets. Closed-loop systems with recycling at the end-of-life provides a pathway to lower environmental impacts and a source of high value materials that can be used in producing new batteries.

Currently, consumer electronics make up the bulk of spent LIB. Most of these batteries are landfilled or disposed in some other way (e.g., in a drawer) because environmental regulations concerning end-of-life batteries are not fully developed or implemented in many countries including the United States. Only a small number of spent batteries are currently sent to the existing recycling facilities [1]. However, as electric vehicles begin to reach their end-of-life, the volume of the spent LIB waste stream is expected to grow rapidly. With proactive regulations regarding collection and disposal of spent batteries and innovations in recycling technologies, end-of-life batteries could supply a significant fraction of the materials needed for manufacturing of new LIB. This paper briefly reviews the current economics and challenges in the supply chain of virgin materials for LIB manufacturing in Section 2. In Section 3, we address the efforts that have been undertaken in various countries to increase recycling of end-of-life vehicles (ELVs) and section 4 discusses how management of ELVs could impact LIB recycling success. The "reverse supply chain" for spent LIB is discussed in Section 5. Recycling methods, world recycling capacity and benefits of recycling are discussed in section 6.

Nomenclature

BEV	Battery electric vehicle
ELV	End-of-life vehicle
EV	Electric vehicle
HEV	Hybrid electric vehicle
LCO	Lithium-cobalt oxide
LIB	Lithium-ion batteries
LFP	Lithium Iron Phosphate (LiFePO ₄)
NMC	Lithium Nickel Cobalt Manganese Oxide (LiNiCoMnO ₂)
LFP	Lithium Manganese Phosphate
NCA	Lithium Nickel Cobalt Aluminium Oxide (LiNiCoAlO ₂)
LMO	Lithium Manganese Oxide (LiMn ₂ O ₄)
LCO	Lithium Cobalt Oxide (LiCoO ₂)
PHEV	Plug-in hybrid vehicle

2. Challenges in the LIB Supply Chain

Consumer electronics are currently the largest LIB application. However, LIB have emerged as the battery of choice for electric vehicles because of their high energy and power density and long life [2]. Sales of electric vehicles are expected to increase rapidly in the next years. For example, the compound annual growth rate (CAGR) for plug-in hybrid electric vehicles (PHEVs) is forecasted to be 56% in the period 2017 to 2020 and the CAGR for fully battery electric vehicles (BEVs) is expected to be 42% for the same period ([3,4] and NREL analysis 2018). The total global capacity for vehicle LIB manufacturing was more than 31 GWh in 2016 [3-5], and demand is expected to exceed 120 GWh by 2020. This rapid growth is projected to require more than 550,000 metric tons of the battery materials—lithium, cobalt, manganese, nickel, and graphite—by 2020. LIB are also the leading battery technology used for grid-scale electricity storage; a critical component in integrating increasing amounts of variable renewable energy into the electricity supply. In the third quarter of 2017, deployment, in MW, of stationary energy storage (residential, non-residential, and utility) in the U.S. was up 46% over the previous year and lithium ion technologies continued to make up more than 94% of the installed MW [6]. Figure 1 below shows projections of LIB for different applications. This figure shows that transportation (i.e., electric vehicles and buses) are expected to dominate the LIB market in the coming years.

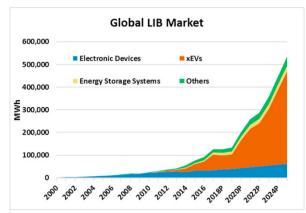


Figure 1. Trend capacity of LIB and their key applications (projections for 2018-2025 are based on data from [3-5])

Current mine production of materials for LIB is limited to a few regions around the world potentially creating availability and price issues. The increased demand represented by the widespread adoption of electric vehicles and large-scale stationary storage combined with limited supply could put upward pressure on prices for these materials and potentially interrupt manufacturers' plans and the projected growth of electric vehicle markets.

According to data from U.S. Geological Survey (USGS) [7], in 2017, 110,000 tons of cobalt were produced globally, with about 60% coming from the Democratic Republic of Congo. China accounted for 67% of the 1.2 million tons of natural graphite produced globally. Lithium extraction was concentrated in Australia (44%) and Chile (34%), with global production totaling 43,000 tons. Sixteen thousand tons of Manganese were extracted primarily in South Africa (33%), China (16%) and Australia (14%). Global nickel production totaled 2.1 million tons, with the Philippines accounting for 11%, Canada 10%, while Russia, and Australia each accounted for 9% of the total. In 2017, 32 countries accounted for all global production of these elements (Figure 2).

3. lithium-ion battery research

Improving Li-ion battery performance and reducing cost have become increasingly active areas of R&D as Li-ion battery technology has become the leading technology for vehicle batteries. Research efforts are concentrated around six primary areas: reducing the dimensions of active materials to improve ion transport and increase mechanical stability; improving the mechanical properties of conductive media; adjusting battery chemistries to improve electron transport; increasing chemical and thermal stability; tuning particle morphology; developing coatings to reduce decomposition of active materials, and modifications of electrolyte solutions [2].

Battery types currently under investigation include lithium metal (lithium metal anodes), solid state batteries that employ solid inorganic or polymer electrolyte, and lithium sulphur with high capacity sulphur-containing cathodes, among others [8]. Whatever path battery technology takes, battery chemistry will likely change significantly over the next decade. Potential changes in battery chemistry, such as developing low-cobalt and cobalt-free cathodes, are important to the supply chain because they may have a significant impact on the demand for critical battery materials and cost. The evolution of NMC and NCA cathode chemistries—NMC and NCA are the main cathodes in automotive LIB—are centered on developing nickel-rich, cobalt-free cathodes [3].

According to Aviceene Energy [3], lithium-ion NMC cathode chemistry is shifting from high cobalt content (e.g., NMC 333) to lower cobalt and higher nickel content (e.g., NMC 622 and NMC 811). NCA, the main cathode chemistry used in Tesla cars, is also shifting toward higher nickel contents (and lesser cobalt content). Unlike NMC and NCA, both LMO and LFP have zero cobalt; therefore, most R&D efforts are directed toward improving their performance (e.g., specific capacity, volumetric energy density, and lifetime). Graphite is still the dominant anode material in most LIB, but recently some researchers have introduced silicon as a cheaper alternative to graphite. Silicon has higher energy capacity and relative abundance in the earth's crust.

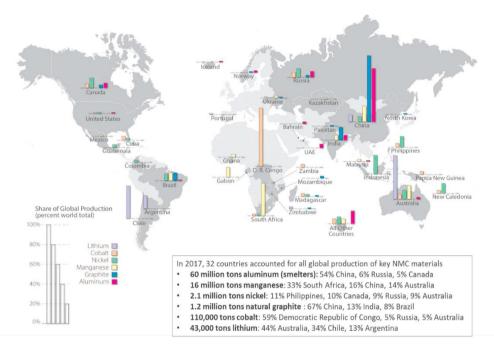


Figure 2. World mining industry production for materials used in LIB in 2017 (data source: USGS 2017 [7])

4. End-of-Life Vehicle Management

Effective collection of LIB from electric vehicles would begin with effective collection mechanisms for end-of-life vehicles (ELVs). ELVs have been recycled to recover valuable parts and materials for many years. Recycling of ELVs, mostly to recover useful metals, had reached 95% by mass in the EU by 2015 [9]. Currently in the U.S., nearly all cars are recycled and, in the recycling process approximately 86% of the vehicle is recovered or used for energy production [10]. Current waste reduction directives in the EU set aggressive goals for recycling of ELVs. Under the European Union End-of-Life Vehicle Directive[†], vehicle manufacturers are responsible for collecting and recycling of ELVs. The Directive required that by 2015, only 5% (by weight) of an ELV could be sent to landfill. In 2016, between 8 and 9 million tons of ELVs were generated in the European Union, and between 80% and 100% of materials from collected vehicles were recovered or recycled[‡] However, not all vehicles taken out of service are collected through regular channels that direct them to recycling/recovery processes and not all countries have reached the recycling goals set by the Directive. A 2007 study commissioned by the European Parliament found that compliance with the ELV collection and recycling directives was difficult for many member states [11]. Collection problems included export of second-hand cars to countries with less stringent disposal regulations, unlicensed operators that only remove economically valuable parts, and abandonment or "garaging" of ELVs by owners. Burdensome administrative requirements, and reluctance to charge vehicle manufacturers for costs associated with take-back programs also impacted compliance.

Japan's End-of-Life Vehicle Recycling Law [12] was promulgated in 2002 and entered into force January 1, 2005 [12] The law is similar to those in Europe except that car owners are required to pay a fee for recycling when

[†] Directive, 2000/53/EC of the European Parliament and of the Council of 18 September 2000 on end-of life vehicles (EPC), 2000. (https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:02000L0053-20130611&qid=1405610569066&from=EN)

[‡] http://www.europarl.europa.eu/legislative-train/theme-new-boost-for-jobs-growth-and-investment/file-end-of-life-vehicles-batteries-andelectronic-waste (accessed 4/12/2018)

purchasing a vehicle so that responsibility for ELVs is shared between owners, ELV-collecting and recycling businesses and vehicle manufacturers. In the same timeframe, South Korea promulgated a law (South Korean RoHS/ELV/WEEE Act, 2007) addressing collection and recycling of vehicles[§]. China has had policies and regulations regarding ELVs in place since 2001 [13]. These policies have been updated and improved in subsequent years, but effective tracking of ELV collection and recycling is lacking and implementation of ELV recycling mechanisms has suffered from some of the same problems encountered in Europe and elsewhere.

There are currently no federal laws in the United States implementing Extended Producer Responsibility (EPR) for ELVs^{**}. However, there are a number of voluntary consortiums (e.g., the ELV Solutions consortium^{††}) addressing recycling and design for recycling. There are also state programs and laws addressing mercury switches and tires.

Because the market for LIB has been dominated by consumer electronics, collection of spent LIB has so far been governed by waste electrical and electronic equipment (WEEE) policies. Several studies have investigated the effectiveness of these programs; however, it is likely that policies and regulations affecting recycling of vehicle batteries will be similar to, and build upon, other vehicle related programs such as the ones discussed above rather than WEEE policies, which are designed for a much different disposal model.

5. The Reverse Supply Chain for LIB

The economics of recycling ELV LIBs must account for all stages in the "reverse supply chain" of collecting ELVs, dismantling and recycling them to recover useful and valuable materials and energy including but not limited to LIB, and finally the economics of the battery recycling process itself. Gradin et al. [14] compared the recovery rates for copper, steel and aluminium and energy use (including energy recovery from incinerating some of the materials) for the two available ELV recycling methods; shredding and manual disassembly. They found that disassembly had both lower greenhouse gas emissions (primarily due to the energy recovered from incinerating energy-rich polymers) and lower metals depletion (due to better recovery of copper) than the shredding option. From a regulatory standpoint, only disassembly could meet the waste reduction goals under the EU ELV Directives. At least partial disassembly of the vehicles would be required for EVs to recycle the batteries and mitigate the safety hazard of shredding the batteries.

Collection of ELVs would probably occur at dealerships or scrap yards where cars are first taken out of service. If disassembly was not co-located with the vehicle collection point, the vehicles would then be transported to a disassembly plant where they might be stored for a period of time before being disassembled. After the car is taken apart, the constituent materials would be transported for further processing at a recycling facility or energy recovery site. Cost components associated with each step include transport costs, energy for operation of the disassembly plant, labor and costs associated with the logistics of storing and handling of the vehicles. Optimization of networks of collectors, dismantlers and recyclers has been undertaken by a number of researchers [8, 10, 15] Golenbiewski et al. [15] investigated the optimal placement of vehicle dismantling facilities in Poland. They found that transportation costs made up 70% of the total cost of vehicle recycling, which includes collection and dismantling of the ELVs and processing of the remainder (shredding and incineration) to recover energy and materials.

Recycling of LIB from vehicles would begin with a similar dismantling process to remove the battery system from the vehicle. The batteries would then be discharged to render them safe to handle, further disassembled, and then processed in one of the processes described in Section 5. Wegener et al. [16] examined the process for disassembly of the battery system for an Audi Q5 and VW Jetta hybrid car. They outlined 24 individual steps in the disassembly of the Audi Q5 battery system including removal of the battery management system, removal of coverings and casings, wiring, connectors and cables. All of the steps were done by hand with minimal tools (e.g., a screw driver). However, because of the complexity of the process and expected variations between the configurations of different cars, the authors suggested that there was minimal opportunity for automation. Therefore, the process is likely to remain time

[§] http://www.step-initiative.org/south-korea-resource-recycling-of-electrical-and-electronic-equipment-and-vehicles-act-no-8405-2007.html (accessed 4/13/2018)

^{**} https://archive.epa.gov/oswer/international/web/html/200811_elv_directive.html (accessed 4/13/2018)

^{††} http://elvsolutions.org (accessed 4/13/2018)

consuming and expensive at least in the near term. The potential for automation of this step would be less influenced by variations in chemistry that become more relevant in the recycling of the battery modules themselves [16].

6. LIB Recycling Processes

6.1. Battery Recycling Methods

Lithium battery packs are generally composed of a cathode, an anode, an organic electrolyte and a separator that are laminated and compressed together to create an electrical contact between them [17]. The cathode is made of active metal powders that possess certain electrical qualities. Natural and artificial graphite are common anode materials. Separators are usually made from thin plastic sheets. An electrolyte acts as an inert component in the battery and must demonstrate stability against both the cathode and anode surfaces. Several chemistries and several states of electrolyte materials are used in LIB, including a non-aqueous electrolyte that is made from lithium salts solubilized in organic solvent, an aqueous electrolyte of lithium salts solubilized in water and polymeric electrolytes [18].

Three technologies—used alone or in combination—are employed commercially for recycling LIB from vehicles, consumer electronics, and other: pyrometallurgy, cryo-milling (recovery of Li) or other mechanical processes, and hydrometallurgy. Direct recycling, an emerging recycling method, is a solvent extraction process where supercritical carbon dioxide (CO₂) is used to extract cathode and anode materials [19]. Table 1 provides an overview of the advantages and disadvantages of the primary LIB recycling processes.

Recycling Method ^a	Pros	Cons	Recovered Materials	Examples
Mechanical Processes	Applicable to any battery chemistry and configuration. Lower energy consumption. Enhance the leaching efficiency of valuable metal	Must be combined with other methods (mainly hydrometallurgy) to recover most materials	Li ₂ CO ₃	Toxco process
Hydrometallurgy	Applicable to any battery chemistry and configuration	Only economical for batteries containing Co and Ni	Copper, Aluminum, cobalt, Li2CO3. Anode is destroyed	Shenzhen Green Eco-manufacturer Hi-Tech Co. (China); Retriev Technologies (Canada); Recupyl S.A. (France)
Pyrometallurgy (smelting)	Applicable to any battery chemistry and configuration	Only economical for batteries containing Co and Ni; Gas clean-up required to avoid release of toxic substances	Cobalt, nickel, copper, some iron. Anode is destroyed	Umicore (Belgium); JX Nippon Mining and Metals (Japan)
Direct Recycling (supercritical CO ₂)	Almost all battery materials can be recovered	Recovered material may not perform as well as virgin material, mixing cathode materials could reduce value of recycled product	Almost all components (except separators)	OnTo Tech ^b (USA)

Table 1. Comparison of Main Recycling Methods Used to Recover Battery Materials

^a Recycling pros and cons from [18, 20]. ^b OnTo is a small recycling R&D company that did not announce any recycling quantities in 2016

6.2. World Battery Recycling Capacity

Recycling efforts of the end-of-life LIB are not comparable to the recycling of lead acid batteries [20]. Recycling of lead acid batteries is profitable because recycled lead, which has high purity, can be recycled back into new batteries, while the electrolyte solution is drained and subject to further chemical processing [20]. Recycling capacities for non-lead acid batteries are shown in Figure 3. We can see that recycling efforts in Europe and Asia are far ahead of that in in North America. This is partially due to the stringent environmental regulations set by European Environmental Agency (particularly 2006/66/EU directive on waste batteries and accumulators and 2000/53/EC directive on end-of-life vehicles). The recycling efforts in China are supported by the economic value of recovered materials making China the top country in terms of the recycling capacity.

In 2016, the world battery recycling capacity exceeded 94,000 [9, 21, 21]. These estimated capacities included all electrochemical storage batteries except lead acid batteries. European countries host about 50% of these capacities, and China alone hosts about 33% (see Figure 3).

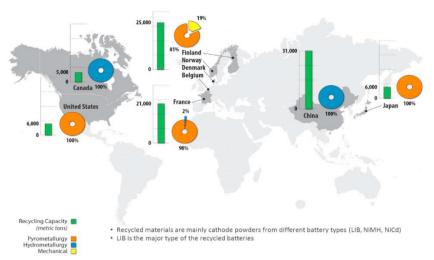


Figure 3. Recycling capacities of the spent batteries in metric tons (MT) (source of data: [9, 22, 23])

6.3. Environmental and economic impacts of LIB recycling

Lower cost of recycled materials is expected to be the main driver for recycling end-of-life LIB. Environmental and social aspects could also contribute to the need for more recycling of end-of life batteries. The key areas where we can see savings are cost, energy and greenhouse gas emissions. Table 2 summarizes some values from several studies [21 - 24]. Cost, as a key economic driver, is expected to decrease with economies of scale. The cost values provided in Table 2 are not fixed especially with the dynamic prices for raw virgin cobalt, lithium and other materials used in LIB. Significant savings in energy and emissions are also expected from recycling end-of-life LIB. However, direct recycling is an emerging recycling method and appears to be more environmentally friendly with lower levels of energy consumption and emissions relative to pyrometallurgy and hydrometallurgy.

	Cost			Energy CO _{2-Eq}						
	LCO	NMC333	NMC811	LFP	LCO	LMO	LCO	NMC333	LMO	LCO
Pyrometallurgy	38%	6%	5% more		35%		70%	78%		70%
Hydrometallurgy	41%	13%	1%		38%	18%			5%	
Direct Recycling	43%	27%	16%	15%	5%	76%		94%	10%	
Virgin Raw					77	34	200 kWh/kg	9 kg CO ₂ .	5 kg CO ₂ .	11 kg CO ₂ /kg-
Materials	\$ 62	\$ 45	\$ 40	\$ 32	MJ/kg	MJ/kg	material	_{Eq} / kg cell	_{Eq} / kg cell	material
Source	[23]			[22]	[22]	[24]	[25]	[23]	[24]	

Table 2. Savings from recycling cathode materials from the end-of-life LIB relative to the use of virgin materials

7. Concluding Remarks

Continued growth in the use of LIB batteries for consumer electronics, electric vehicles and grid electrical storage will increase the demand on critical materials such as cobalt, lithium and graphite. The increase in demand for electric vehicles will be partially but not fully offset by changes in LIB chemistry with lower cobalt content and new anode chemistries using silicon. Competition between original equipment manufacturers for scarce resources has highlighted the importance of recycling of the end-of-life batteries.

All recycling methods have been shown to be economic at high volume with current raw material prices and battery composition. However, the reverse supply chain for ELVs and LIBs must be optimized to fully realize the economic benefits of recycling. Recycling also has lower environmental impacts compared to the mining of virgin materials. Direct recycling could result in less energy and emissions compared to pyrometallurgy and hydrometallurgy and can be used to recondition old cells to recover pure cathode and anode powders which need minimum processing before putting them back in the cells. Previous cost studies suggest cost savings that could reach 43% of the cost of cathodes made from virgin materials.

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