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Lithium Battery Reusing and Recycling: A Circular Economy Insight

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This study is dedicated to Dr Maria Lea Ziino for all she has done for the health of younger generations in 20 years of child psychology work carried out in Sicily

Abstract: Driven by the rapid uptake of battery electric vehicles, Li-ion power batteries are increasingly reused in stationary energy storage systems, and eventually recycled to recover all the valued components. Offering an updated global perspective, this study provides a circular economy insight on lithium-ion battery reuse and recycling.

Keywords: lithium-ion battery • battery recycling • battery electric vehicle • circular economy

1. Introduction

Driven by the electric vehicle (EV) boom,[1] which led to a 3-fold increase in the price of lithium[2] and a 4-fold increase in that of cobalt[3] between 2016 and 2018, reclaiming lithium, cobalt, manganese and nickel (along with other valued materials like copper, aluminum and graphite) from spent lithium ion batteries has lately become profitable. Perhaps not surprisingly, numerous new lithium battery recycling plants started operation across the world, and those existing are expanding capacity.

In about 2 years, the recycling of lithium batteries which still in 2016 was claimed in Europe to lack economic viability as "only 3% of the material mix in batteries is made of lithium"[4], became profitable and convenient.

Investments started to flow targeting opportunities not only for recycling but also for refurbishing and reusing retired EV lithium-ion batteries (LIBs) in energy storage systems. "Certain companies" reads a working document of the European Commission dated mid 2018, "have already begun investing in recycling of used EV batteries in Europe (e.g. in Belgium and in France). Some have teamed up with car manufacturers to collect and recycle batteries".[5]

Hence, as lately emphasized by Melin, a reputed consultant in lithium-ion battery life cycle, "when it is not rare to read about recycling rates of 3 or 5 per cent… 58 per cent will be recycled this year".[6]

Recycling, in general, relies on first generation recovery technologies in which a physical treatment to obtain different streams of raw materials is followed by a hydrometallurgical process (leaching and extraction) to extract metals.[7]

The environmental benefits of this powerful economic trend are significant. As the lithium-ion recycling industry consolidates and the demand for spent LIBs increases, the old practice for which small batteries used by portable electronic devices were hazardously stockpiled in generic materials recovery facilities causing fires due to thermal runaway from damaged or short circuited batteries,[8] will become a thing of the past.

This trend, we argument in this study, will further evolve and eventually first generation LIB recycling processes will be replaced by green chemistry processes producing highly pure ("battery-grade") lithium, cobalt and manganese compounds along with graphite, copper and aluminum.

Lithium-ion batteries, indeed, generally use a graphite anode and a cathode made of lithium metal oxides generally comprised of lithium-iron phosphate (LFP), lithium-nickel manganese cobalt (NMC), lithium nickel cobalt aluminum oxide (NCA), lithium-manganese oxide (LMO), or lithium-titanate oxide (LTO). First generation LIBs mainly used in portable electronics used lithium-cobalt oxide (LCO).[9]

The cells are assembled in modules and modules further assembled in battery packs. The voltage from "power" batteries supplying current to the motor of electric passenger cars or buses, can respectively top 300 V or even exceed 600 V.

2. Technology and chemistry aspects

By weight percentage (g material/g battery), a typical lithium-ion battery comprises about: 7% Co, 7% Li (expressed as lithium carbonate equivalent, 1 g of lithium = 5.17 g LCE), 4% Ni, 5% Mn, 10% Cu, 15% Al, 16% graphite, and 33% other materials.[10]

In a recent patent,[10] seven main components (cobalt, lithium, copper, graphite, nickel, aluminum, and manganese) were reported to comprise >90% of the economic value of a spent lithium-ion battery: Co (39%) and Li (16%, as LCE equivalent) followed by Cu (12%), graphite (10%), Ni (9%), Al (5%) and Mn (2%).

Besides so called "calendar ageing", a lithium-ion battery becomes "spent" (reduced ability to store and deliver electricity) mainly because during the charge and discharge cycles taking

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place in the battery cells a solid product forms due to reaction of the lithiated anode with the alkyl carbonate comprising the electrolyte solution.[11]

The resulting solid electrolyte interphase mainly consisting of stable (such as Li_2CO_3) and metastable components (with the latter polymers, $ROCO_2Li$, $(CH_2OCO_2Li)_2$, and ROLi prone to decompose exothermically at >90°C, releasing flammable gases and oxygen)[8] progressively deposits on the anode surface forming a passivating film which limits the electrochemical reaction by making graphite sites inaccessible for Li^+ to intercalate and thus leading to an increase in internal ohmic resistance.

A typical EV lithium ion battery pack has a useful first life of 200,000-250,000 km,[12] even though increasingly adopted fast-charging at >50 kW reduces the battery pack duration since battery degradation rapidly accelerates with charging current.[13]

When, the automotive battery pack loses 20% (15% for certain EV models) of its initial capacity it becomes unfit for traction as the lower capacity of battery affects acceleration, range and regeneration capabilities of the vehicle.[14]

2.1 Second-life batteries

Besides the beneficial effect on the price of grid electricity due to the concomitant expansion of EVs utilization and renewable energy generation (particularly solar photovoltaics),[15] a second synergistic effect of battery electric vehicle on renewable electricity uptake lies in the possibility to reuse the batteries at the end of their automotive lifecycle for utility-scale energy storage.

Compared to use in EVs, stationary applications demand lower current density from the battery pack. Hence, batteries retaining between 80-85% of their original capacity are rearranged into racks suitable for stationary applications other than automotive such as utility-scale grid, building or telecommunication tower storage applications, nicely fulfilling the key “refurbish, reuse, recycle” circular economy principle.

Battery modules found to have similar power and life are sorted out and re-assembled in new “repurposed” battery packs, ready for stationary usage.[16]

A significant public demonstration of the ability of repurposed batteries to provide energy storage and grid services (regulation of the alternating current frequency in the grid) is the 3 MW (nominal power)/2.8 MWh (nominal capacity) energy storage system installed in 2018 at Amsterdam’s “Joahn Cruyff Arena”, (Figure 1).[17]

During events at the stadium, the demand for electricity lighting, powering broadcasting, information technology equipment, catering, and security services increases from a baseload of around 200 kW to more than 3000 kW, for the entire duration of the event.[17]

Figure 1. Amsterdam’s “Johan Cruyff Arena” multipurpose stadium. [Photo courtesy of Eaton].

The new energy storage system installed at Amsterdam’s Arena is comprised of 590 battery packs (340 new and 250 second-life batteries with each EV second-life battery having slightly less than 20 kWh capacity from the original 24 kWh directly supplied by the EV maker certified to last 10 years), namely the equivalent of batteries included in 148 used exemplars of the first generation world’s best selling EV.[18]

The batteries are now contained in 61 battery racks (Figure 2). Four bi-directional inverters manage the energy flows from the 4,200 rooftop PV modules, from and to the grid, and from the batteries (we remind that the grid accepts and supplies only alternating current, whereas the PV modules and the batteries supply direct current only).

Figure 2. Racks with part of the 2.8MWh energy storage system at the “Johan Cruyff Arena”. [Photo courtesy of Eaton].

The new energy storage system enables optimal use of both solar PV and electricity retrieved at low cost from the grid during the night hours.

Now the PV energy generated during the day, rather than being fed into the grid and sold to the grid operator at low price, goes to charge the 2.8 MWh battery pack whose nominal capacity was chosen to meet the energy demand for 1 h during the most important events with maximum power absorption; and for 3 h when accessory services such as catering are not in use.[17]

Flattening ("shaving" in the jargon used by electricity practitioners) the peak demand with free PV or low cost grid electricity stored in the lithium batteries *i*) cuts the diesel fuel cost, *ii*) avoids peak demand charges, and *iii*) generates a revenue stream when the energy storage system is used to provide well paid grid-balancing services, such as frequency control.

Similar energy storage systems combining second-life EV battery modules with battery and power management digital technology for both residential, commercial and industrial applications are increasingly commercialized across the world by a number of companies.

Similarly, in China the world's biggest operator of telecommunication towers, since 2018 ended purchase of lead-acid batteries. All existing and rapidly ageing lead-acid batteries currently installed at 98% of its 2 million telecom towers across China (54 GWh battery storage demand for back-up power for their base stations) will be replaced by second-life LIBs.[19] Partnership agreements were signed with more than 16 EV and battery manufacturers as second-life LIBs in 2018 were reported to be priced at less than \$100/kWh, namely the same price of new lead-acid batteries.[19]

For comparison, this translates into forthcoming demand for up to 2 million retired EV batteries only from China's telecom base station back-up, since one single tower needs about 30 kWh back-up battery.[19]

According to a thorough analysis conducted in 2017 by Melin, by 2025 about three-quarters of spent EV batteries will be reused in second-life solutions for several years after retirement from vehicles, after which they will be sent to recycling to recover all the valued components.[20]

2.2 New green chemistry technologies

Reviewing first-generation metal recovery processes using pyrometallurgical or hydrometallurgical methods, scholars in China lately emphasized the need for new "selective leaching of most of the valuable metals from the spent LIBs".[7]

Discovered in 2015, one such green process for the recovery of metals from spent Li-ion batteries makes use of citric acid (H_3Cit) and aqueous H_2O_2 affording Co and Li in excellent recovery yields (98% Co and 99% Li).[21]

In detail, the spent batteries are first discharged and then manually dismantled to recover the Al and Cu foils in metallic form and the separator, all directly recycled after dismantling (Figure 3).

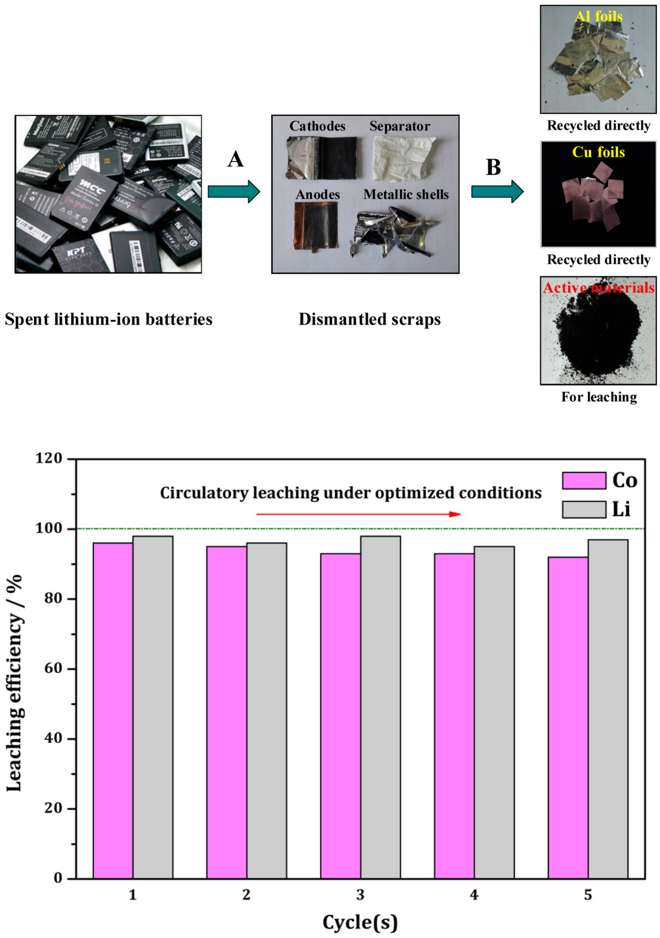

Figure 3. Simplified pretreatment process of spent LIBs based on citric aicd/hydrogen peroxide oxidative leaching of Co and Li: (A) manual dismantling; (B) peeling off Al/Cu foils and recycling of Al and Cu (*top*); and circulatory leaching experiments under the optimized conditions using citric acid and H_2O_2 (*bottom*). [Reproduced with permission from Ref.21, Copyright American Chemical Society].

The waste cathode materials ground into finer fractions for the subsequent extraction process is obtained by calcining at 700 °C for 2 h the cathode materials to remove carbon.

The powder of cathode material thereby obtained is used as raw material for the leaching process under optimized and mild extraction conditions (80 min, 70 °C, 2.0 M H_2O_2, with reductant dosage of 0.6 g/g, and slurry density of 50 g/L).

Aqueous H_2O_2 acts as efficient and clean reductant during metal leaching (Eq.1), with both metal ions and waste citric acid being simultaneously recovered by selective precipitation.

$$\begin{aligned} &H_3Cit + LiCoO_2 + H_2O_2 \\ &\quad \rightarrow Co_3(Cit)_2 + Co(HCit) + Co(H_2Cit)_2 + Li_3Cit \\ &\quad + Li_2(HCit) + Li(H_2Cit) + H_2O + O_2 \qquad (1) \end{aligned}$$

Co and Li ions dissolved in the lixivium are treated with oxalic acid and phosphoric acid solutions to recover Co and Li, in a total reaction equation (Eq.2) showing that water and oxygen are the only byproducts in the whole recovery process.

$$LiCoO_2 + H_2O_2 + H_2C_2O_4 + H_3PO_4 \rightarrow Co(C_2O_4)_2 + Li_3(PO)_4 + H_2O + O_2 \qquad (2)$$

In a truly closed-loop route typical of the circular economy, about 99% Co and 93% Li could be recovered as $CoC_2O_4{\cdot}2H_2O$ and Li_3PO_4, whereas the recycled citric acid shows similar leaching capability as fresh acid (Figure 3, bottom).

LFP batteries, we have discussed elsewhere,[1] will remain for many years the dominating lithium battery technology used by electric vehicles. It is therefore particularly relevant the recent discovery of a green and economically viable process for recycling entire spent $LiFePO_4$ batteries to battery grade (99 wt%) Li_2CO_3 ready for manufacturing new LFP batteries.[22]

The process is based on the selective leaching of lithium based on oxidation of $LiFePO_4$ to $FePO_4$ with aqueous sodium persulfate ($Na_2S_2O_8$), forcing lithium deintercalating from the cathode (Eq.3), while neither Fe (0.048% leaching) nor P (0.387%) leach out from the cathode structure whose olivine crystal structure is retained during the leaching process.

$$LiFePO_4 + Na_2S_2O_8 \rightarrow FePO_4\downarrow + Li_2SO_4 + Na_2SO_4 \qquad (3)$$

In detail (Figure 4), cathode scrap of LFP powder attached on Al foil obtained by a local battery recycling company is first separated from soft-package batteries via discharging and dismantling, and then cut into small pieces.

More than 99% of Li is leached without the addition of acid and alkali under the optimal conditions of 1.05 times the stoichiometric amount of persulfate, under remarkably mild conditions (25 °C, 20 min stirring a 300 $g{\cdot}L^{-1}$ suspension of powdered cathode plates) with nearly no wastewater and solid waste generation.

No prior separation of cathode active material and Al foil (the most demanding procedure in the present recycling process of spent LIBs) is required because in the strong oxidative environment, Al is passivated (formation of a thin layer of Al_2O_3) resulting in an extremely low leaching of Al.

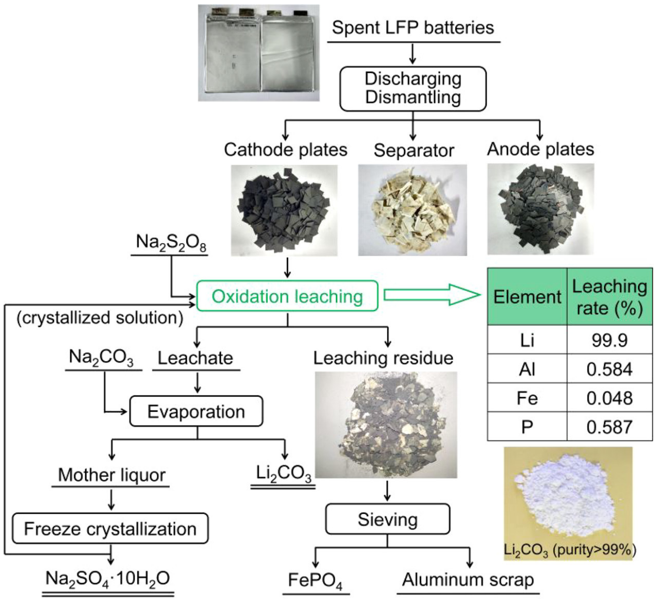

Figure 4. Flowsheet of the method for treating entire spent $LiFePO_4$ batteries using aqueous sodium sulfate under neutral conditions for Li leaching. [Reproduced with permission from Ref.22, Copyright American Chemical Society].

The leaching of lithium is very rapid with >90% of Li leached into the solution in only 5 min, an further increases to 99.8% by prolonging the leaching time to 20 min. As a result, the most valuable element in spent LFP batteries is directly recovered as Li_2CO_3 of high purity (>99%) by simple addition of Na_2CO_3 to the leachate followed by evaporation.

In a closed-loop method with great potential for industrial upscale, the mother liquor obtained after evaporation is used to prepare valued $Na_2SO_4{\cdot}10H_2O$ product via freeze crystallization, whereas the crystallized solution is returned to leach another batch of raw cathodes with the addition of fresh $Na_2S_2O_8$..

Besides Li recovery, the process enables direct cathode recycling to make new LFP cathodes for new batteries using the well-crystallized orthorhombic $FePO_4$ leaching residue whose XRD pattern is close to that of raw $LiFePO_4$ cathodes (the reverse of the phase transformation occurring in the charging of LFP battery, in which $LiFePO_4$ releases Li^+ ion and turns into $FePO_4$).

Affording highly pure (>99.5%) lithium, the aforementioned processes solve the main problem which so far has limited the industrial uptake of green chemistry processes in LIB recycling, namely the "lengthy processing and purification processes of the raw materials to reach battery grade"[23] which determines "the true cost to manufacture"[23] Li-ion batteries.

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3. Economic aspects

The economic (and environmental) advantages of EVs are so large (electric buses, for example)[24] that, regardless of rapidly growing output from new large factories in China, the demand of Li-ion batteries currently overcomes supply. This is especially the case for countries and regions like Europe where limited Li-ion battery manufacturing takes place. One Germany's bus manufacturer, for example, by early 2019 was reported to be unable to get the batteries needed to start manufacturing electric buses in 2020.[25]

New regulation in China now holds EV makers responsible for the recovery of batteries, requiring them to set up recycling channels and service outlets where old batteries can be collected, stored and transferred to recycling companies. By the end of February 2019, 393 carmakers, 44 scrapped car dismantling enterprises, 37 cascade utilization enterprises and 42 recycling enterprises had already joined the new traceability platform to track origin and owners of discarded batteries.[26]

Furthermore, since 2017 new legislation forbids to import in China electronic waste, including batteries, which is leading China-based companies formerly supplying lithium carbonate, cobalt and nickel sulfates obtained from batteries retired from large consumer electronics manufacturers to establish new recycling plants "overseas" (in South Korea for example);[27] as well as foreign EV battery makers to open recycling plants in China.[28]

Figure 5. The automatic control system of GEM power battery precursor material production line. [Photo courtesy of GEM Co., kindly reproduced from gem.com.cn].

Industrial LIB recycling companies in China include Taisen Recycling, Zhejiang Huayou Cobalt, Brunp, Jinqiao Group, Jiangxi Ganfeng Lithium and GEM. The latter company, for example, operates in China 13 automated battery dismantling and recycling facilities where it manufactures the cathode precursors (Figure 5), with an annual production capacity of cobalt, nickel materials of lithium ion batteries and cathode material exceeding 50,000 tons.[29]

The products resulting from battery recycling are sold to battery manufacturers. Hence, it may not be surprising to learn that large battery manufacturers own recycling companies, as in the case of Brunp, a lithium-ion battery recycler in Hunan.

Though smaller, from Singapore (TES-AMM operating a plant using an hydrometallurgical process developed in France), through South Korea (SungEel) and Belgium (Umicore), from the U.S. and Canada (Retriev Technologies) through Australia (Envirostream Australia) and Great Britain (Belmont Trading), several other companies across the world operate LIB recycling facilities.

The list above is far from being exhaustive. What is relevant here is that, driven by dramatically growing uptake of LIBs for electric vehicles, recycling companies are rapidly expanding their facilities and new companies are entering the market.

For instance, by early 2019 when the company recycled over 8,000 tons of retired batteries annually through an hydrometallurgical process, a Korean firm was undergoing a 5-fold expansion with three new plants due to start operations in Hungary, India, and in USA.[30]

Market forecasts for the LIB recycling industry agree on significant growth, though forecasted figures vary. According to the aforementioned 2017 report,[6,31] recycled lithium will reach 9 percent of total lithium battery supply in 2025 (namely 5,800 tonnes of recycled lithium, or 30,000 tonnes LCE), and that of cobalt almost 20 percent of the demand, with >66% lithium-ion batteries being recycled in China.

4. Conclusions

In early 2017, Zhao published one of the first comprehensive books on the reuse and recycling of lithium-ion power batteries.[32]

Referring to the "uncertain performance and service life of retired power batteries", ending the "Market Development of Reuse and Recycling of Power Batteries" chapter, he wrote:

> "The profit margin in the reuse of lithium-ion power batteries is unclear. Although data on batteries provided by lithium-ion power battery producers state that the batteries removed from new energy vehicles retain 70‑80% valid energy and appear competitive in costs, there are still many challenges when energy storage is focused in the field of battery reuse". [33]

Two years later, a large electric power company started construction of a 268.6 MWh energy storage plant in the east China's Jiangsu Province. The PV+storage plant will use retired EV batteries of 75,000 kWh residual capacity (45,000 kWh from LFP batteries and 30,000 KWh from lead-acid batteries), with additional storage capacity of 193,600 kWh from new LIBs.[34]

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This single example renders the pace of innovation in energy storage (and renewable electricity storage in particular), and reinforces the need to broaden and renew the education of energy managers,[35] particularly in the field of solar energy[36] whose 2018 photovoltaic output in China grew by 50 per cent in one year only, to the outstanding figure of over 177 TWh.[37]

Regardless of ongoing reports for which, citing data going back to 2010, lithium-ion batteries would be "currently recycled at a meagre rate of less than 5% in the European Union",[38] this study refers to actual figures for which, globally, 58% of the world's spent LIBs will be recycled only in 2019.[6]

Narins has lately shown that access to lithium is limited more by logistical issues (cost-effective access) and quality than actual availability,[38] with the huge lithium reserves in Bolivia's salt flats not yet exploited (agreements between with Bolivia's State mine company and China's and Germany's companies were signed, in 2018 and in early 2019).[39]

Yet, with several large factories under construction or announced and global capacity projected to reach over 1,100 GWh by 2028 from 202 GWh in 2018,[40] the recycling of LIBs is no longer an option but an inevitable need for both battery and EV manufacturers (as shown by the scarcity of battery grade lithium lately recorded by the company willing to expand electric bus manufacturing).[25]

Helping to further streamline and automate the recycling process, the circular economy companies recycling lithium-ion batteries already work with battery makers to adopt easily dismantled product designs, and will shortly uptake the new green chemistry processes lately developed for the waste-free recovery of all valued battery components.

In conclusion, as energy storage in lithium-ion battery is essential to expand the uptake of clean and renewable energy, this study providing and updated global perspective on lithium-ion battery reuse and recycling will be useful to scholars, for their teaching and research needs, as well as to policy makers and citizens engaged in the energy debate.

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