Closing the Loop of High-added-value Materials (CloseLoop)

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Narrative #1: Tools for circular economy of lithium-ion batteries

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1. Background

Lithium-ion batteries are powering our portable electronics including mobile phones, tablets and laptop computers. They are also considered as the major candidate for electric vehicles (EVs) and stationary electricity storage for storing variable renewable energy (wind, solar) and stabilizing smart electricity grids. The consumption and market value of lithium-ion batteries is expected to increase rapidly as the use of EVs and stationary electricity storage become more and more common. An example of market estimates is given in Figure 1.

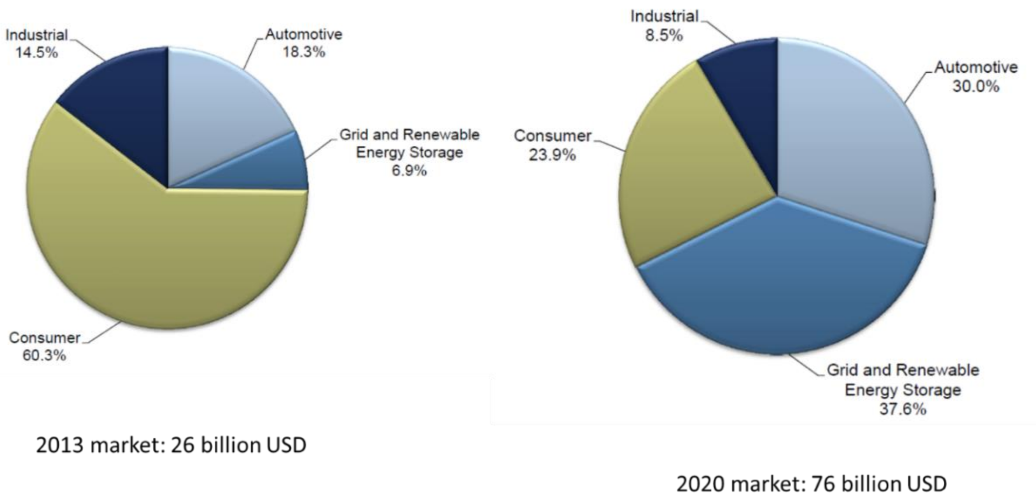

Figure 1. *Estimates for the market shares of different Li-ion battery applications in 2013 and 2020 (Frost & Sullivan, 2014).*

As thousands of individual battery cells may be needed for each battery electric vehicle or grid scale energy storage unit, the fast growth of these application raises concerns about the sustainability of lithium battery production and availability of raw materials for them (Petersen, 2016) (BMI, 2016). 13.000 Tons of lithium battery chemicals worth 380 M$ were used for electric vehicles in 2013. This is expected to grow by a factor of 20 by 2020 reaching 270.000 Tons and 7.8 B$ (Frost & Sullivan, 2014b). The consumer electronics market was three times the size of the EV market in 2013 and is expected to double by 2020. This means that the lithium battery chemical market can grow from a few ten thousand to one million Tons by early 2020´s.

A typical lithium ion battery contains a graphite anode coated on copper foil, an oxide cathode coated an aluminum foil, a polymer separator and an electrolyte consisting of a lithium salt dissolved in organic solvent, tightly enclosed within a battery case usually made of steel, aluminum, plastics or nickel alloys, Figure 2. The cathode oxide contains lithium, cobalt, manganese, nickel and/or aluminum. As an alternative, lithium-iron-phosphate (LiFP) can be used as the cathode material but the energy density is lower than for the cobalt containing cathodes (Petersen, 2016).

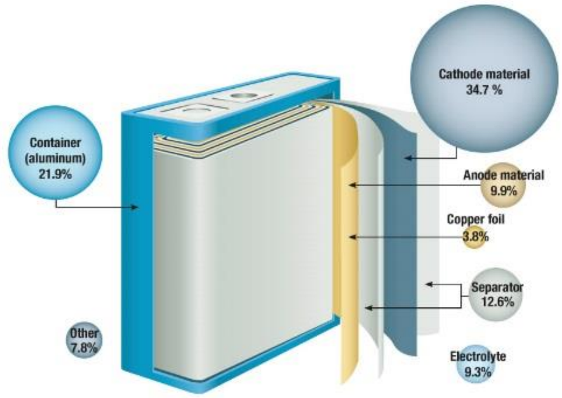

Figure 2. *Lithium ion battery construction.*

An estimate of the composition of an average electric vehicle battery is given in Table 1.

Table 1. *Composition of an average electric vehicle battery (Worrell & Reuter, 2014).*

Element	**%**
Aluminum	25
Iron	21
Copper	11
Carbon	9
Cobalt	0-4
Manganese	0-4
Nickel	0-4
Lithium	<0.5

As far as availability and prices of raw materials are concerned, the critical lithium battery materials are lithium, cobalt and graphite (Petersen, 2016) (BMI, 2016). Cobalt can be replaced in stationary and high power application but it is critical for high energy application in portable electronics and battery electric vehicles. Estimates for lithium and cobalt production, useful reserves and global resources as well as share of battery application in the end use are given in Table 2.

Table 2. *Production, reserves, share of battery use and recycling rate of lithium and cobalt (Jaskula, 2016) (CDI, 2016) (UNEP, 2011).*

	Lithium	Cobalt
Annual production (Ton/a)	32.500	99.000
Useful reserves (million tons)	14	7.1
Global resources (million tons)	34	120
Share of battery use (%)	35	42
Main reserves	Chile, Argentina, Bolivia China, Australia	Democratic Republic of Congo
Recycling rate (%)	< 1	68

In order to facilitate sustainable growth of lithium battery use, different circular economy (CE) concepts should be implemented within the industry.

2. Objectives

Lithium batteries have been selected as a case example to study different circular economy concepts for high-value-added materials. The main objective is to develop a holistic CE approach for the battery use combining the top-down (circular design and new business models) and bottom-up (critical material substitution and new recovery processes) approaches.

In the top-down approach the objective is to design new products, business concepts and ecosystems for sustainable use of lithium batteries and the materials within. The development of a sound method to evaluate the materials life cycle from a holistic point of view, including the evaluation of battery manufacturing and recycling technologies, is also necessary.

In the bottom-up approach, the objectives are to analyze the design, performance and degradation mechanisms of state-of-the-art lithium batteries as well as the existing second life, refurbishing, reuse and recycling processes in order to improve them. Especially, the objective is to facilitate safe reuse of automotive batteries in stationary applications and efficient recycling of all the battery components after this secondary use. Research in battery metal recycling focus on improved tailoring of mechanical, pyrometallurgical and hydrometallurgical unit processes and process flow sheet, aiming at recycling of all metallic elements. Firstly the focus is on the state-of-art battery waste whereas later in the development stage high-added-value batteries. A final objective of the project is to replace all the critical raw materials in lithium ion batteries by earth abundant ones, for example organic compounds, and recycle them. Such a battery would not be raw materials limited provided that lithium is recovered and recycled from used batteries.

Information regarding the different approaches is shared with industrial and municipal stakeholders and government officials as well as with the general public with the objective of creating new high value industries, jobs and export opportunities.

3. Tools

The top-down approach is based on multidisciplinary “Modelling factory” simulation approach by VTT combining eco-design, material databases, life cycle assessment (LCA) and business models with social and political impacts. The energy flow based LCA tools are further refined by exergy analyses by Aalto University and TU Bergakademie Freiberg (Reuter, 2016) .

Mathematical models are developed and refined to optimize the dismantling of lithium ion batteries and metallurgical recovery of all the metal components in a form usable for further processing into new batteries. These models as well as process simulation is verified using well designed and executed experiments using spent Li batteries from consumer electronics. The experiments consist of test series related to unit processes such as mechanical separation, hydrometallurgical leaching, solvent extraction and metal precipitation. The models produced should allow to predict in a reasonable extent the recycling potential and mass balance of the battery components, with a correlation to economical parameters whenever possible. New batteries are produced using the recycled materials and their performance is compared with similar batteries based on pristine raw materials.

Tools for analyzing the battery state-of-health (SoH) of used automotive Li-ion battery packs are developed in order to refurbish them for safe secondary use in stationary applications, Figure 3. Li battery ageing phenomena are studied under selected battery use scenarios. The results are used to develop non-disruptive methods to estimate the SoH. Initial tests are performed for individual battery cells.

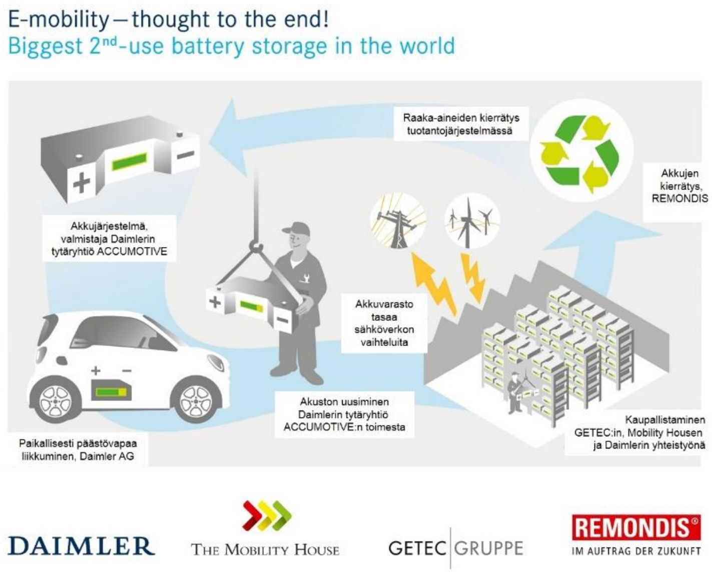

Figure 3. *Circular economy of Li batteries by Daimler (Daimler AG, 2016).*

Lithium battery structures that are easy to disintegrate and dismantle are proposed. After dismantling damaged components can be removed and healthy components reused which is more efficient than scrapping the whole battery and recycling of the raw materials. Easy to disintegrate interfaces are developed for the battery electrodes and easy to open or dismantle methods for the battery cans, packages and electronics (BMS: battery management system).

Atomic (ALD) and molecular (MLD) layer deposition methods are used to grow organic electrodes and lithium micro batteries based on these electrodes, Figure 4.

The results are disseminated to the CloseLoop Stakeholder group with industrial, municipal and governmental participation. Co-creation workshops are organized on regular basis for effective involvement of these Stakeholders. Further domestic and international cooperation is sought with relevant players in every part of the battery cycle life, i.e. material producers, cell and pack manufacturers, automotive companies, second life service providers and recyclers.

Figure 4. *Characterization of ALD/MLD grown microbatteries. Photo by Mikko Raskinen, Aalto University.*

The results are further disseminated in scientific and industrial journals, magazines, conferences and workshops as well as project web pages and social media. The regulatory needs to promote circular economy of lithium batteries will be discussed with relevant ministries and the European Commission officials.

The project results are implemented in the curricula of Aalto University and University of Helsinki both for under and post graduate students. 6 post graduate and 2 post doctoral students are working in the project.

4. Impacts

The use of lithium and cobalt produced only from primary resources will likely face difficulties in coping with the fast demand growth of the battery industry. 35% of the global lithium and 42% of the cobalt production is already used in batteries (Jaskula, 2016) (CDI, 2016). The expected tenfold increase in demand is not possible in short term and not sustainable (Petersen, 2016) (BMI, 2016). Starting new mine operations is capex intensive and time consuming and the main reserves are outside Europe.

Lithium batteries are recycled in dedicated recycling plants or as base metal classified fractions at the primary metals plants. Umicore, the leading European recycler, recycles 7000 tons of mixed Li-ion and metal hydride battery waste. In the state-of-the-art pyro metallurgical recycling process only cobalt, nickel and copper are recovered (Reuter, ym., 2013). Lithium and aluminum are lost in the slag. Altogether, less than 1% of the world lithium is recycled and only small minority recycled back to the battery chemicals. According to some estimates however, high recycling rates of lithium would be needed to ensure an adequate supply for the 21st century (Gruber, ym., 2011). Cobalt is the other critical raw material used in lithium batteries for it is mainly produced in unstable countries like Congo (EC, 2014).

Economic recovery of lithium or lithium-components from battery waste would drastically decrease the concerns about future availability and price of lithium as the demand increases. Furthermore, the recovered lithium and other metals could be further processed to battery raw materials and chemical precursors by Finnish companies represented in the CloseLoop Stakeholders Group. While some researchers have claimed recovery efficiency for lithium of more than 74% (Zhang, ym., 1998), its implementation is presently limited by its currently low commercial value.

The uncertainty regarding the state-of-health (SoH) of used batteries is a major constraint for the secondary use applications. Improved methods to estimate the SoH of battery packs and individual battery cells will facilitate new methods for refurbishing battery packs, estimating their remaining useful life and warranting their performance. This is especially important to guarantee the safety of the refurbished packs which is a prerequisite for successful development of the second life application.

Replacement of graphite anode and metal oxide cathode materials by organic compounds would eliminate the resource scarcity in Li battery manufacturing. Moreover, the batteries could be made thin, flexible and transparent which would open completely new application fields for micro batteries in healthcare, sensors and electronics.

5. Desired impacts

The Finnish companies working on cobalt (Freeport Cobalt, Norilsk Nickel), nickel (Norilsk Nickel) and copper (Boliden) are represented in CloseLoop Stakeholders Group. Freeport Cobalt using mine tailings from Congo as the primary raw material has 8.6 % of the global cobalt refining capacity. All the companies are already today processing recycled materials. Development of more efficient recycling processes for low grade and mixed recyclates can improve their competitiveness in the market or bring new metal circular economy SME operators next to the plant facilities.

The collection rate of the old batteries present e.g. in cellphones and laptops is still not satisfactory, but also recycling rate of some metal fractions such as Li and rare earth metals is low. . The battery types used in the laptops and cellphoness are valuable in metal value and thus desired raw material for the recycling operators. In order to get the collection working properly new business models are needed. Consumers have concerns about their private information in laptops and phones and therefore trustable practices to deal with the concern are needed. Furthermore, consumers are expecting to get benefits when giving away their old gadgets, otherwise they will easily remain in drawers just in case one will use it later on.

6. Secondary impacts

The hydrometallurgical processes developed to recover battery metals from battery waste where the metals are in dilute concentrations and in the form of mixed oxides could be further applied to metal recovery from other secondary raw materials. Also the process models built can then be adjusted for different types of raw material feed to evaluate the mass flows and environmental impacts.

The modelling tools developed using Li-ion batteries as working case should include simulation of recycling processes and their impact on the entire materials value chain. Therefore, it is our aim to develop robust simulation principles and recycling parameters upon which the development of recovery processes for other metals for example from electronics waste (WEEE) could be based.

Replacement of cobalt and nickel in Li batteries could have a long term impact on the businesses of companies involved in the value chain of Co and Ni battery chemicals. However, as the alternative technologies emerge first in small scale low power application and the main growth is expected in large scale high power applications the companies are expected to have excellent prospects for market growth and sufficient time to adjust for any longer term transitions.

7. Research background

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