State-of-the-art in reuse and recycling of lithium-ion batteries – A research review

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Commissioned by The Swedish Energy Agency

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Preface

Less than 5 per cent of the lithium-ion batteries in the world are recycled. The few processes that are available are highly inefficient and the costs to recycle lithium is three times as high as mining virgin lithium. With the rapid growth in e-mobility we will end up with a mountain of waste with millions of tonnes of batteries dumped in landfills.

Statements like these are found both in the news and social media every week, all over the world. And usually they are supported by research from NGO's, corporations and the academia.

There is only one problem. The statements are not true.

Although there is no official statistics over recycled volumes in the world, for somebody who visits collectors and recyclers around the world it becomes clear that significantly more batteries than only 5 per cent are recycled. The processes are many times very efficient and you can be fairly sure of that in the battery in your phone, laptop, or electric car if have one, chances are there is recycled lithium and cobalt inside.

But why is this not mentioned in the research? Or, do we actually know that it's not? Maybe there is just no-one reading it.

This was a feeling I had when trying to understand how there could be so many companies in China and South Korea that recycled lithium-ion batteries while the general perception was that this is barely happening at all. When The Swedish Energy Agency, which is subordinate to the Ministry of Environment and Energy and responsible for the funding of battery research in Sweden, saw my early work they commissioned my company Circular Energy Storage to make an independent review of what's actually out there.

What we found was that many areas are far better understood than what most people believe. We have found hundreds of published papers covering primary research in areas such as material recovery and reuse of batteries. However, we also identified important areas which have attracted very little attention with the consequence that valuable research results often becomes useless because there are other bigger problems that still are not addressed.

We also identified one of the reasons of why statements like those above continuously get more fuel is that far too many researchers are using bad secondary data and rarely check their references. Let's give an example. In an editorial in the scientific journal Nature Energy in April 2019 the "5 per cent" recycling rate is used. The cited source is an article in The Guardian from 2017 which in turn has found the number in a report from the NGO Friends of the Earth, published in 2013. Friends of the Earth used a press release from the European Battery Recycler's Association, EBRA. In the press release, which is from 2011, the collection numbers among its members for 2010 are reported, which for lithium-ion showed a 27 per cent decline that year. According to personal communication with a recycler Friends of the Earth conclude that this is "around 5 per cent" of what has been placed on the market. Hence, the number used in 2019 is nine years old and from a time when there were no electric vehicles, tablets and barely any smartphones on the market. Still Nature adds "currently less than" to the number.

This is not an isolated phenomena. We have found several reviews which are listing recycling processes that can be traced back to research papers or even student thesis's from the early 2000s and which since then have never been checked whether they are still valid. For instance there are two British recycling facilities which are listed in paper after paper, all over the world, despite the fact that they never existed. Even prices of raw materials are sometimes referred to as high or low based on previous research although prices for materials like lithium and cobalt can be highly volatile and have changed signficantly over the last years. Worse still is that there is rarely any new primary data added to this which means that research papers in 2019 continue to describe a situation from around 2010.

Often the reason behind this is that an overview of the market or even current technologies wasn't the primary purpose with the research. Instead it was used to put the research in context. But if the context isn't correct then there is a risk that the relevance of the research is not as high as the researchers state.

Another phenomena is that there is so much primary research done especially in China and South Korea that rarely is used in Western research. If that is because it is considered irrelevant or because it just hasn't been found is not for us to say but if it was the former reason there should at least be more discussions in the papers why researchers have chosen not to refer or use the results as basis for their own work.

The purpose of this report has been to create a tool for both researchers and government officials when prioritising projects to fund in the future. Besides the report The Swedish Energy Agency has also published a data base with all sources and their abstracts that have been identified in this work. We hope that this will facilitate future research in the area, both for the academia and for companies. Of this reason the report has now also been made available in an English version in order to further advance the area on an international basis.

London, June 2019

Hans Eric Melin
Circular Energy Storage

Summary

This report aims to give an overview of the current knowledge about reuse and recycling of lithium-ion batteries. The work has been commissioned by the Swedish Energy Agency with the purpose to identify any gaps in knowledge and technical know-how required to make the end-of-life chain for lithium-ion batteries more efficient in order to ensure that future funding of research in the area is used effectively and where it is mostly needed.

The research review mainly comprises peer-reviewed research articles published in English, as well as known research projects in Sweden, the EU and the US in the areas of collection, reuse, recycling, batteries' environmental impact and design for recycling. To put the findings in context the report also includes a background section with data on the global market for reuse and recycling of lithium-ion batteries.

Lithium-ion batteries differ from other battery types in several ways. They usually have longer lifespan than other batteries such as lead-acid batteries. They are also more often built into equipment and cannot easily be disposed of separately by the user. This contributes to the fact that it takes longer time for the batteries to reach recycling and that they are exported to a greater extent than other batteries for reuse purposes, which means that they are not recycled in Sweden or not even in Europ. This has caused a perception that recycling is underdeveloped while it's in fact works well. At least in the countries where the batteries end up, primarily in China and South Korea.

That the public perception is out of sync with reality could have to do with the fact that research within both generation and collection of end-of-life batteries is very sparse. Only a few studies in China, Japan and the United States have focused on how batteries reach recycling and what prevents more of them from doing so. In addition, most research has been done on the collection of mixed portable batteries. Little has been written about how batteries reach end-of-life or what life span they can be expected to have.

Reuse, or Second Life, is considerably better covered. As a rule, these studies have one of three different focuses: economic potential, technical possibilities and limitations, and which environmental consequences reuse of batteries entail. Most studies are fairly small and there is a clear need to study larger systems. The conclusions are positive.

Recycling is the area which is best covered. More than 300 studies featuring primary research have been found where various types of methods have been tried to separate different substances from waste batteries and to re-produce cathode material or precursors. More than 75 percent of the studies have been done on hydrometallurgical processes and 70 percent were carried out by scientists in China or South Korea. Most of these have been focused on the treatment of LCO and NCM batteries, while only a few studies have been done on LFP, LMO and NCA batteries. The results show that all active materials including lithium can be recycled with high efficiency.

There are a number of areas where research is completely lacking. Several of these have great significance for the entire recycling chain, especially from an industrial point of view. For example, there is very little done on sorting and classification of batteries, about discharging and pre-treatment, and not least in design for reuse and recycling. What is remarkable is also how absent areas such as safety, work environment and transport are in the overall research.

Research ahead should focus on issues that currently prevent effective recycling. It should also promote an increased system understanding among all players involved. In reuse, there is still an opportunity for the research community to be pioneers and obtain data and experiences that can form the basis for strategic, long-term decisions for the industry. Finally, it may be justified in Sweden to conduct research in areas that are already well covered globally with the aim of strengthening the knowledge in the country.

Background

Since their commercialisation in the early 1990s, lithium-ion batteries have become an increasingly important energy storage technology thanks in particular to their high energy density. This has enabled a rapid development of portable electronics such as mobile phones, laptops and tablets. Around 2010, lithium-ion technology began to be used in electric and hybrid cars and soon thereafter in buses and energy storage systems.

The rapid development has meant that manufacturers of batteries have been able to scale up their production and become more and more efficient. This has led to lower costs which in turn have further increased the demand for batteries and the applications in which they are used. Together, this has created a huge growth in the market, driven primarily by different types of vehicles but also by the possibility of replacing lead-acid batteries in e g base stations and data centers.

With ever-increasing volumes in the market, the requirement for recycling of lithium-ion batteries is also increasing. The batteries contain substances, especially compounds of cobalt and fluoride, which should not be dispersed in nature. Several substances are also relatively rare or extracted in countries with high political instability. Through their value chain, the batteries have also caused climate-impacting emissions whose burden can be reduced if the materials in the batteries can be reused.

However, unlike lead-acid batteries, of which over 90 percent usually are claimed to be recycled in Europe[1], the recycling of lithium-ion batteries is often considered insufficient[2]. Both because European recyclers do not receive a particularly large proportion of the batteries which can be assumed to have reached end-of-life, and because many of the processes are neither sufficiently efficient nor profitable.

Research about lithium-ion battery recycling

An overall insufficient recycling efficiency is often the stated purpose for many research projects in which new methods or technologies are expected to improve the situation. Thus, in today's research projects, there is a large focus on the actual recycling process in which batteries are converted into different types of materials for use in the same or other applications.

Research is also done on the actual collection of the batteries and how it can be improved. In fact collection, or rather the lack of, is often considered to be a limiting factor for recycling as both individuals and companies are hoarding used batteries for too long or allegedly throwing them in ordinary household waste.

An additional area that gets more and more attention in the industry, as well as in the research world, is the potential to reuse lithium-ion batteries in new applications, so-called Second Life. This is mainly about reusing batteries from electric vehicles in energy storage solutions. This research differs from that about recycling as it is more focused on the battery's functions, such as ageing and degradation, monitoring of the battery's health and which commercial benefits reuse can possibly have.

Research in these areas, collection, reuse and recycling of lithium-ion batteries, is within the scope of what The Swedish Energy Agency has as mission to finance. It's complex areas that are closely linked to each other where one area can have consequences for another.

The purpose of this study is to create an overview of how the state of knowledge looks in these areas in order to create better conditions for using funds for new research as efficiently as possible. The goal has been to produce a basis for answering above all two questions:

- What can be considered state-of-the-art in research on recycling and reuse of lithium-ion batteries?
- What gaps are there in knowledge and technical know-how required to make the recycling chain for lithium-ion batteries more efficient?

In order to be able to answer above all the first question, we also need to better understand how the recycling chain looks and what driving forces for how volumes, processes and legislation look like today. This is done in the first chapter where we have described the area from an industrial perspective.

Then follows a review of the research done in the field. The goal has been to describe what research that is available, more than describing the actual results which would have required a much larger approach.

Finally, we summarise the conclusions we have drawn and try to put this into perspective for how it should affect the The Swedish Energy Agency's continued work.

About the author

Hans Eric Melin is a consultant at the London-based company Circular Energy Storage. He has extensive experience from collection and recycling of batteries. Before founding Circular Energy Storage he served as Vice President Market Development at Battery Solutions in Michigan, which is largest battery recycler in the US. Before that Hans Eric was CEO of Refind Technologies, which is a leading company in the sorting of waste batteries. Hans Eric is also the author of Circular Energy Storage's annual report "The Lithium-ion battery end-of-life market" from which data has been included in this report's introductory chapter.

Hans Eric has a BSc in Media and Communication Studies from the University of Gothenburg, which in the early 2000s led him to areas such as the collection of waste, sorting and waste minimisation.

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Recycling of lithium-ion batteries put in context

An introduction used multiple times in research articles about recycling of lithium-ion batteries is a reference to how the battery type was commercialised by Sony in 1991 after being invented in 1986 by John Goodenough, then at the University of Oxford, UK. What is not so often mentioned is that the first patent for recycling of lithium-ion batteries was filed only one year later, in 1992, by the company Valence Technology[3], headquartered in Austin, Texas, which by coincidence is the same city John Goodenough today calls home.

In the year 2000 recycling of lithium-ion batteries was mentioned in a published research article[4] for the first time. The article was written by researchers at the Argonne National Laboratories in the United States, who made an overview of available recycling processes which were three at the time.

Unlike the development of the lithium-ion battery itself, which was gradually developed in several different academic research environments, the development of recycling has long been industry-driven. The same applies to the reuse of batteries, which is also mentioned the first time in the article from Argonne.

The fact that lithium-ion batteries were recycled before the turn of the millennium, and that different recycling methods have been available for as long time, is an important reminder when trying to understand the causes and drivers of how recycling and reuse look today and how it will develop in the future .

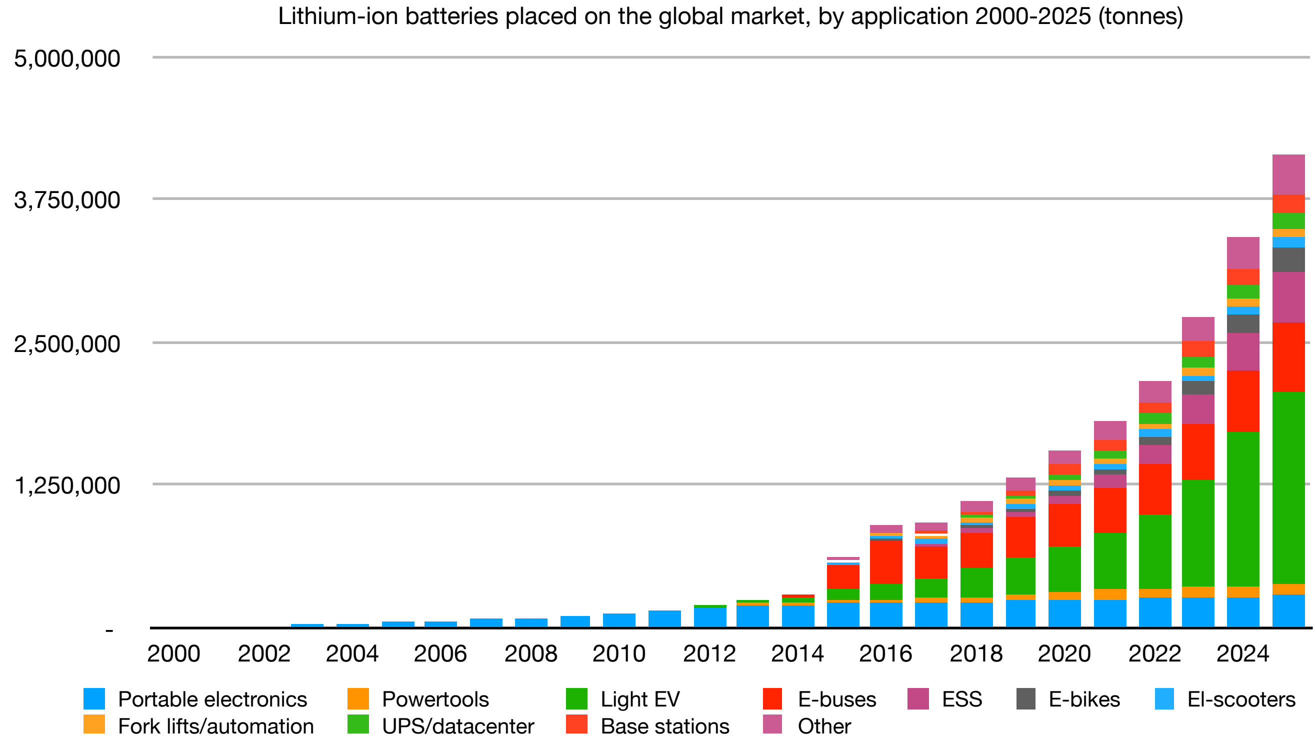
In recent years, the attention to these issues has increased radically. Similarly, research on both recycling and reuse has increased, as have many closely related areas such as manufacturing of batteries from recycled materials. The obvious explanation for this is the increase in the volume of batteries that are now being put on the market and which also have an increasing impact on the extraction of the substances required to manufacture them.

The development of the lithium-ion battery market

From first being used in video cameras, the applications that really made a real impact for the lithium-ion battery were the mobile phone and the laptop[5]. With these two applications, the growth of two main types of cells also started: prismatic and cylindrical where the prismatic cells were mainly used in single cell applications such as mobile phones, while cylindrical cells were used in packs for computers and cameras.

Volume growth has since the start been strong, first generated by increased popularity for mobile phones and computers, and during the second half of the 00's by smartphones and tablets. At the same time, the amount of other applications using lithium ion batteries has increased sharply. From 2010, lithium-ion batteries began to dominate the power tool market and after that, new products such as hover boards, drones and electric bicycles had the lithium-ion battery to thank for their rapid development.

In 2010, the first serial manufactured electric cars began to enter the market[6]. This was the start of the enormous development we are now seeing. Cars, and shortly thereafter buses, have grown strongly during the 10’s and are now the dominant segments of the market. It is worth noting, however, that it was only in 2016 that cars and buses surpassed portable electronics as the largest application and had it not been for the enormously fast growth of Chinese electric buses then portable electronics had still been the largest area of use.



Source: Circular Energy Storage

Like the rapid growth of mobile phones and laptops, economies of scale in battery manufacturing which subsequently enabled their use in cars now push the development for other applications - thanks to the large investments made in EV batteries. In particular, applications that have traditionally used lead-acid and nickel-cadmium batteries, such as backup power for base stations and data centers, have started to use lithium-ion batteries. The same applies to energy storage systems that can be used in everything from batteries connected to solar power plants to balancing the entire power grid instead of, for example, pumped hydropower. Even fork lift trucks and other industrial vehicles are increasingly turning to lithium-ion batteries. These new segments will soon also be larger than electronics and will be an important market for battery manufacturers.

The situation in Sweden is similar. In the Swedish Environmental Protection Agency statistics it is not possible to discern which applications have been placed on the market other than whether batteries are portable (electronics and power tools) or industrial (larger built-in batteries including electric vehicle batteries). But the tendency looks the same. From having completely dominated the area, the growth of portable batteries has leveled out and since 2015, industrial batteries have been the largest category put on the market[7].

The shift from a market dominated by electronics to a much more diverse market has major consequences for recycling companies. Above all, it means three major changes[8]:

1. Different cathode chemistries

Batteries in mobile phones and computers were initially exclusively of the type LCO ($LiCoO2$). Cells of this type contain between 17 and 20 percent cobalt, which is the most valuable substance in the batteries. With power tools, energy storage, electric bikes, buses and the majority of all cars, especially in China, the LFP battery ($LiFePo4$) came. This type does not contain cobalt at all, which means that the recovery from recycling from a value standpoint is very low. The same applies to LMO ($LiMn2O4$) which is used in electric scooters, power tools and in many of the first large-volume electric cars such as Nissan Leaf and Chevrolet Volt, although in a combination with NMC (Li (NiMnCo) O2). NMC, together with NCA (Li (NiCoAl) O2) has subsequently become the dominant battery chemistry for electric cars in

Recyclable materials in different lithium-ion battery types

Material	USD/kg	% Content in a cylindrical cell (18650)							
		NCM111	NCM523	NCM622	NCM811	NCA	LFP	LMO	LCO
Casing									
Steel	0,29	10.0%	10.0%	10.0%	10.0%	10.0%	10.0%	10.0%	10.0%
Aluminium	1,8	10.0%	10.0%	10.0%	10.0%	10.0%	10.0%	10.0%	10.0%
Current collectors									
Aluminium	1,8	5.0%	5.0%	5.0%	5.0%	5.0%	5.0%	5.0%	5.0%
Copper	6,0	7.0%	7.0%	7.0%	7.0%	7.0%	7.0%	7.0%	7.0%
Anode material									
Graphite	1,2	18.1%	18.1%	18.1%	18.1%	18.1%	18.1%	18.1%	18.1%
Cathode material									
Manganese	2,4	6.1%	5.5%	3.6%	1.8%			19.4%	
Lithium	70,0	2.3%	2.3%	2.3%	1.9%	2.3%	1.4%	1.2%	2.3%
Cobalt	30,0	6.5%	3.9%	3.9%	1.9%	2.9%			19.3%
Nickel	12,0	6.5%	9.7%	11.6%	15.4%	15.6%			
Aluminium	1,8					0.4%			
Iron	0,4						11.3%		
Total value per kg		**5.42**	**5.02**	**5.19**	**4.77**	**5.32**	**1.97**	**2.26**	**8.30**

Source: Circular Energy Storage

the western world. Both chemicals contain cobalt but only between 2 and 6.5 percent in NMC and less than 3 percent in NCA.

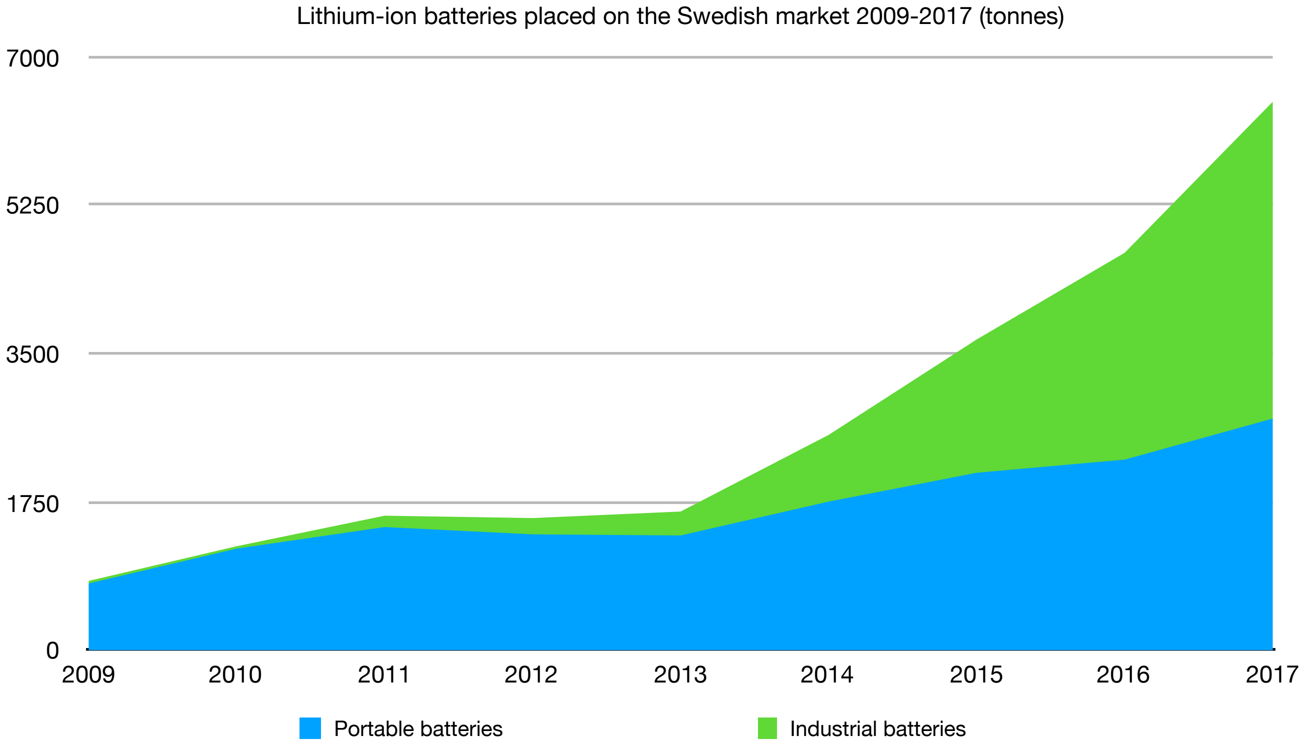
Apart from the fact that the new battery chemistries have reduced the average value of the batteries, the complexity has increased for the recyclers to keep them apart. This is important both from a technical and an economic perspective. However, the problem should not be exaggerated since cell types can often be associated with their respective applications or products they have been in, which means that they can usually be identified relatively easily.

2. Batteries are getter larger and are assembled

While the challenge for portable batteries lies in sorting the batteries, the challenge for EV batteries and other industrial batteries is that they are assembled in larger modules, which in turn are assembled in battery packs. This means that the amount of work needed to prepare the batteries for recycling is considerably larger than for portable batteries and at the same time requires higher skills.

3. Batteries are following their applications

Unlike old portable batteries, lithium-ion batteries today are almost always built into their applications and are not designed to be separated by the user. In applications such as electric cars and energy storage, this is obvious, but since two to three years back the same thing applies to smart phones and laptops. This means that batteries rarely come directly to companies specialized in battery recycling, but rather to recyclers of electronics and cars. Another consequence is that batteries in many cases follow their applications when they are sold and exported, which means that they never reach recycling at all, at least not in the market where they were first sold.



Source: The Swedish Environmental Protection Agency*

Lithium-ion batteries reaching end-of-life in Sweden and in the rest of the world

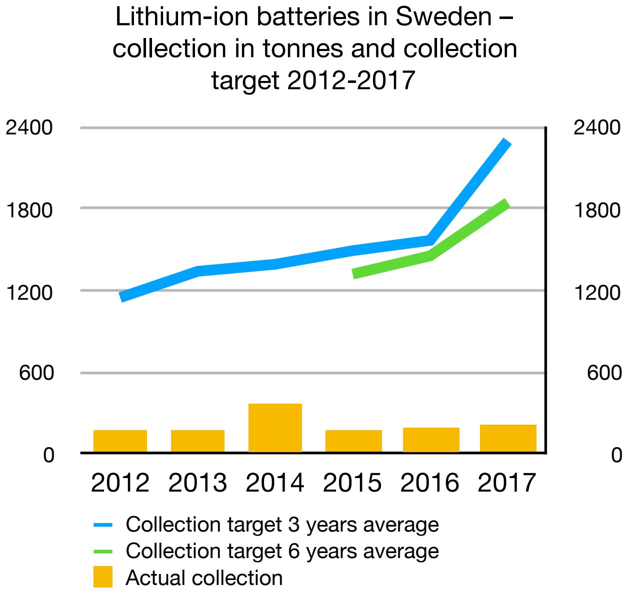
The capacity of a lithium-ion battery decreases both as it is used and because it gets older. How fast this goes depends on a number of factors such as how the battery has been used, whether it has been used as a single cell or in a larger pack, and what type of battery it is. LCO batteries used in electronics are usually estimated to have between 500 and 1000 cycles while an LFP or NCA battery in an energy storage system has been shown to be usable for over 2000 cycles before losing more than 20 percent of its capacity. Finally, however, all batteries will stop working.

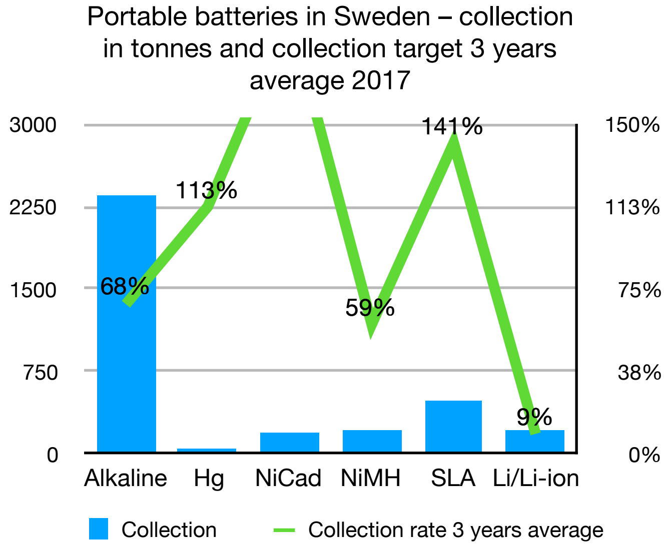
Still, it is not always the battery life that makes it reach "end-of-life" - EOL. In many cases, it may be the equipment that the battery sits in which is considered consumed while the battery itself is still working well. There are currently no statistics available on how many batteries in Sweden that each year reach EOL. The only information available is how much is put on the market and how much is collected to be recycled, and what is finally recycled. However, what is collected by compliance schemes and recyclers is not the same as how many batteries have reached EOL.

One way to calculate the amount of portable batteries that have reached EOL is to use the baseline value in the calculation of the annual collection target in the EU Battery Directive. The directive states that 45 per cent of an average of the three previous years' volume placed on the market should be collected. An assumption could thus be that this average would correspond to the amount of spent batteries and 45 percent corresponds to what the legislator have found reasonable for the member countries to be able to collect from this amount.

Translated to the Swedish Environmental Protection Agency's statistics, this would mean that the amount of portable lithium-ion batteries in Sweden that 2017 reached EOL would be 2297 tonnes. This should be compared with the amount of portable lithium-ion batteries collected the same year which was 206 tons, corresponding to a collection rate of 9 percent. However, a study conducted by the European association of national collection schemes for batteries, EUCOBAT, has shown that the period of three years is too short as people rather keep their batteries for an average of

* The chart has been adjusted for an abnormal volume in 2016 most probably due to a reporting error to The Environmental Protection Agency. The adjusted volume has been obtained by moving 1300 tonnes from 2016 to 2015 and 2017.





Source: The Swedish Environmental Protection Agency*

six years[9]. The difference in the collection rate for the Swedish part however would not be radically different, but then amount to 11 percent.

The low collection rate for lithium-ion batteries becomes even more visible when compared to the collection rate for other battery types. In some cases, these are affected by phase-outs, especially when it comes to mercury and nickel cadmium, which means that more batteries are collected than what is put on the market. Other types of batteries, such as alkaline and lead-acid batteries, also have a shorter life than lithium-ion. But the difference is nevertheless remarkable, not least in comparison with nickel-metal hydride which has a similar life span and use as lithium-ion batteries.

For industrial batteries, it is reasonable that the time between the battery being put on the market until it is consumed is even longer since these batteries in many cases are designed to last for many years, in some cases over 10 years. But even if you use an average of what was put on the market 6-8 years ago, the collection rate is no more than 16 percent. This is far from Sweden's collection target, which is 95 percent.

The figures are in line with the situation in many other European countries[10] as well as in the US[11] and Australia[12], which report from 2 to 11 percent collection and recycling rates.

There is currently very little academic research that has tried to explain the low collection. One common explanation is that the batteries are instead disposed of in the household waste and that EV batteries should also be placed in landfills[13]. However, we have not found any studies that support this by examining how much batteries that are actually put on landfill but the claims are only assumptions that the batteries that are not collected are discarded in another way.

Another explanation that is often referred to is that the batteries are hoarded in homes and companies, primarily in the equipment that they have initially been installed in, such as mobile phones[14]. Several studies have been done in this area, confirming that in many cases people keep their old phone when they buy a new one. However, there is no study in the area that is even close to explain the whole great discrepancy between collection and EOL.

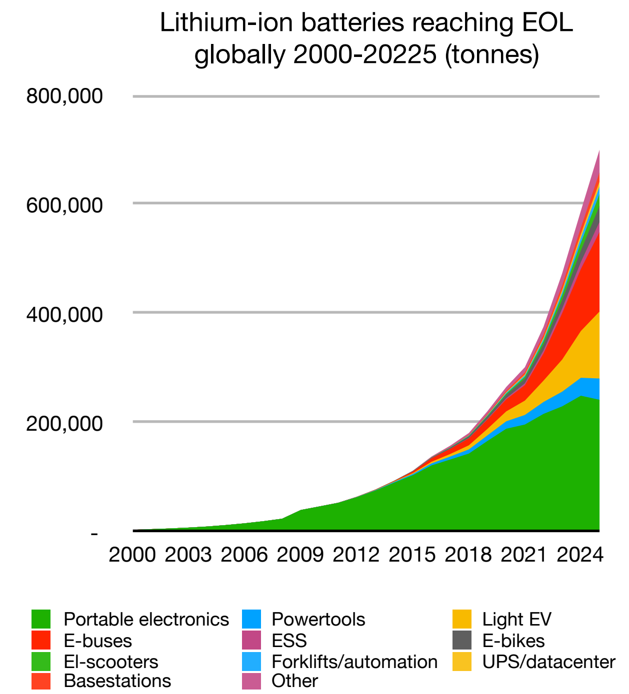
Circular Energy Storage's explanation is that large quantities of batteries are exported instead. Partly because smartphones and tablets, but also laptops, are mainly exported to China for reconditioning and reuse. In these cases, the batteries are included as part of the equipment as an export product. The batteries are also exported because batteries collected by electronics recycling companies are sold to recyclers in mainly South Korea or China for recycling or to reconditioning companies that use the batteries in e g power banks, as these companies pay much higher prices than recyclers in Europe or US. This, increasingly extensive, trade means that large quantities of batteries that would otherwise have been recycled in Europe will instead become available to recyclers in Asia.

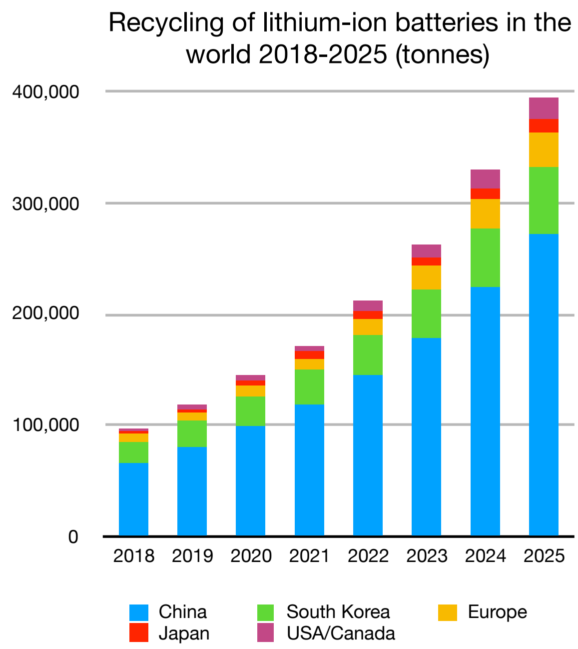
This means that just because the recycling rate of lithium-ion batteries is low in Europe it does not mean that the recycling rate is low globally. Instead, Circular Energy Storage has estimated that as much as 97,000 tonnes were recycled in 2018, of which 67,000 tonnes in China and 18,000 tonnes in South Korea.

The reason why so few industrial batteries are recycled is a combination of the fact that the batteries have simply been good for use longer than expected, and that a large proportion of batteries are reused in new applications. In particular, EV batteries are used in energy storage systems. There is also a large proportion of batteries that are only kept in stock while waiting for companies to make decisions about how to use them. Still over 80 percent of the batteries that reach EOL come from electronics.

Recycling of lithium-ion batteries

The picture that lithium-ion batteries cannot be recycled which is often conveyed in both media and in many research reports is not true. Today, there are over 50 companies around the world which recycle lithium-ion batteries on some scale, from small laboratory plants to full-scale factories[15]. Most companies are located in China followed by South Korea, EU, Japan, Canada and the United States. In principle, each market has greater capacity than the currently available supply of batteries to recycle.





Source: Circular Energy Storage

The structure and methods look different in the different markets. The largest market for recycling lithium-ion batteries, China, consists of over 30 companies that to a varying degree recover both waste batteries and production waste. The methods are almost exclusively hydrometallurgical and the end products are generally different chemical products such as cobalt sulphate, nickel sulphate or lithium carbonate. Many companies also produce complete blends to produce, for example, NCM or NCA cathodes, so called precursors. More than one third of the companies are associated with producers of cathode or anode materials. The large volumes come from large quantities of production scrap and discarded new products, as well as from waste batteries collected from the refurbishment market, where batteries are replaced and sold to traders who in turn sell to the recyclers.

Several of the large lithium producers also have, or plan to, invest in recycling facilities and use recycled materials in their products. The competition for batteries to recycle is very high, which also means that prices are higher than anywhere else. In principle, all types of cells, including LFP, have positive values in China – the recyclers pay to get hold of the material.

In South Korea, recyclers also work closely with materials companies. Several companies have for many years imported material mainly from North America to produce battery chemicals for South Korean battery manufacturers. In recent years, South Korea has also become a road into China where batteries cannot be shipped as waste. The connections to China have also increased through Chinese acquisitions of South Korean recyclers and through joint ventures. The methods are, as in China, mainly hydrometallurgical and the main products are precursors and battery chemicals.

Europe has about ten companies that treat lithium-ion batteries in different ways. The recovery efficiency is not as good as in China and South Korea, which is mainly due to lower volumes which does not make it profitable to extract all substances. Both pyrometallurgical and hydrometallurgical methods are used as well as combinations thereof. Although several existing plants have very small volumes in relation to their capacity, additional facilities are planned in different parts of the Union. Volumes in Europe come mainly from compliance schemes for portable batteries, but also through contracts with producers that put industrial batteries on the market. Unlike South Korea and China, no recycler in Europe produces finished products for the battery material market, but instead sells so-called black mass or chemical compounds that require further processing in order to be used in the production of new batteries or other applications. This also means that the values of the waste batteries in Europe are considerably lower or even negative, which further contributes to the low volumes.

The US and Canada are among the pioneers in recycling, but today the volumes are limited as the largest quantities are exported to South Korea, China and to some extent to Europe. Several start-up companies are about to start processes, especially in Canada. From having both hydrometallurgical and pyrometallurgical methods, the remaining, as well as the new processes, are only hydrometallurgical in combination with mechanical processes.

Japan, too, belongs to the pioneers but, like Europe and North America, the country suffers from volumes being exported to China and South Korea. Despite a large market for cathode materials, the low volume has meant that the methods have developed slowly over the last decades. Other growing markets for lithium-ion recycling are Malaysia, Indonesia, the Philippines and Singapore, which also import batteries from Europe and from where the processed materials can be exported to China.

Reuse of lithium-ion batteries

The biggest reason for the low volumes of industrial batteries is that they are still used in their original applications. But as the batteries for various reasons get classified as EOL, many are reused in new applications.

The pioneers in this segment are the companies that were early on put electric cars on the market: Nissan, Renault, BMW, and Daimler. A large number of initiatives have been taken by these companies, which includes energy storage systems in homes as well as industries and commercial premises, larger energy storage systems in regional or national electricity grids or support for charging of electric cars in both home and public charging stations.

In many cases, thet vehicle manufacturers have created solutions that combine new and used batteries. One reason for this is that the amount of batteries coming back is still very small. There is currently greater capacity in energy storage installations to fill than there are batteries available.

The same situation prevails in China where batteries from mainly buses and early electric cars have started to come back. In a government-run initiative, there is an agreement signed by 16 of the largest battery and vehicle companies and the country's operator of telecom towers to use the batteries as backup power. With 1.9 million base stations with an average requirement of around 100 kWh, there is thus also a greater need in China than there are currently available EOL batteries. So far, 10,000 tons of EOL batteries have been reused in backup solutions, corresponding to a capacity of 800 MWh or 2 percent of the need.

In addition to reuse, used batteries are re-manufactured as replacement batteries for the same car model they previously have been used in, in order to service the aftermarket. This, in turn, is a reason why several manufacturers are not more active in second life solutions, as the batteries they come across simply are used in their cars.

An obstacle for car and battery manufacturers to invest in second life, or to otherwise take advantage of the reuse market, is that they in many cases do not have control over the batteries. For industrial batteries, the law in Europe says that anyone who puts industrial batteries on the market is also responsible for the battery being taken back and recycled, unless otherwise agreed. However, there is nothing that says that an owner must send the battery back to the manufacturer. This means that batteries, for example, acquired by car salvage companies, rather go to the one who is prepared to pay the most for the battery. Only when the battery is damaged and can be assumed to have a negative value is it likely that the legal right to return the battery will be exercised as long as nothing else has been agreed between the seller and the buyer.

The market for second life solutions based on reused batteries is expected by several analysts to increase sharply[16]. But it's not the only the industrial battery market. As mentioned earlier, a large amount of portable batteries are also reused[17]. This is mainly done in China, which is also how spent batteries come into the country without being classified as waste, but there are also companies in both the US and Europe who test and sell batteries overseas. The batteries can then be used in their original products such as mobile phones or laptops or mobile chargers and smaller packs.

Research on recycling and reuse of lithium-ion batteries

As mentioned in previous chapters, recycling and reuse of lithium-ion batteries were initially driven by industry and individual contractors. It took until the year 2000 when the first research articles were published. It was then in the form of an overview of the already existing market[18], a description of an industrially developed process[19] and a first scientific experiment with the goal of recovering substances from an LCO battery[20].

Since then, research on the entire EOL area for lithium-ion batteries has increased almost every year. The research done in the field can be divided into four categories:

- Industrial research and development
- Academic reviews and system analysis
- Academic experiments, modelling and development of new technology
- Academic Life Cycle Assessments

In this review we have mainly focused on academic research but also looked at what was done in the industry by studying different patents or going through various publicly funded research projects.

Purpose

The purpose of the compilation has, as previously mentioned, been to answer which research is available within the EOL area for lithium-ion batteries and to identify potential blanc spots. Specifically, three issues have been central:

1. Which research is available in the different sub-areas?
2. What are the main findings of this research?
3. Where has the research been conducted?

The goal has been to give a picture of how much we know, or rather could know, based on what is available in publicly published research.

Method

The focus of the work has been to identify as many research sources as possible and to divide them into a rough structure. Articles have then been searched using Google Scholar with a number of fixed keywords in four different areas:

1. Generation and collection of lithium-ion batteries: "collection lithium-ion batteries", "collection EV batteries"
2. Reuse of lithium-ion batteries: "second life lithium-ion batteries", "reuse of lithium-ion batteries"
3. Recycling of lithium-ion batteries: including recycling lithium-ion batteries, recycling EV batteries
4. Life cycle analysis for lithium-ion batteries: eg life cycle assessment lithium-ion batteries, life cycle analysis lithium-ion batteries, life cycle analysis EV batteries, life cycle analysis EV batteries

The searches were sorted by relevance and ended after three consecutive pages without relevant articles came in sequence.

After the searches, the sources of the 10 most cited articles in each category were scanned to find articles that were not captured by the search terms.

The articles in Google Scholar are for the most peer-reviewed research published in various scientific journals. There are, however, also extracts from scientific textbooks represented as well as in a few cases reports issued by institutes and research companies. Only research published in English has been included in the compilation, although articles published in other languages have been detected as result in the searches.

After the articles have been identified, abstracts have been studied to classify the texts in different sub-areas within their respective areas. Approximately one-quarter of the texts have been fully or partially reviewed for further study.

In addition to Google Scholar's searches, Google's open search engine has also been used to identify Swedish research in the field. This has been supplemented by a review of projects financed by e g the Swedish Energy Agency, Mistra and Vinnova. Searches have also been made at all Swedish universities and research institutes.

Finally, Google has also been used to identify completed and ongoing EU projects and, where appropriate, national projects in countries other than Sweden, including the United States and the United Kingdom. Further searches have also been carried out on the relevant authorities' project lists, such as the EU or Innovate UK.

Research about generation and collection of lithium-ion batteries

Generation and collection includes all events and activities that affect or are required for batteries to reach both reuse and recycling. The processes that exist in this area seldom have a higher technical level, even if there are elements, mainly regarding safety, where technical solutions play a major role.

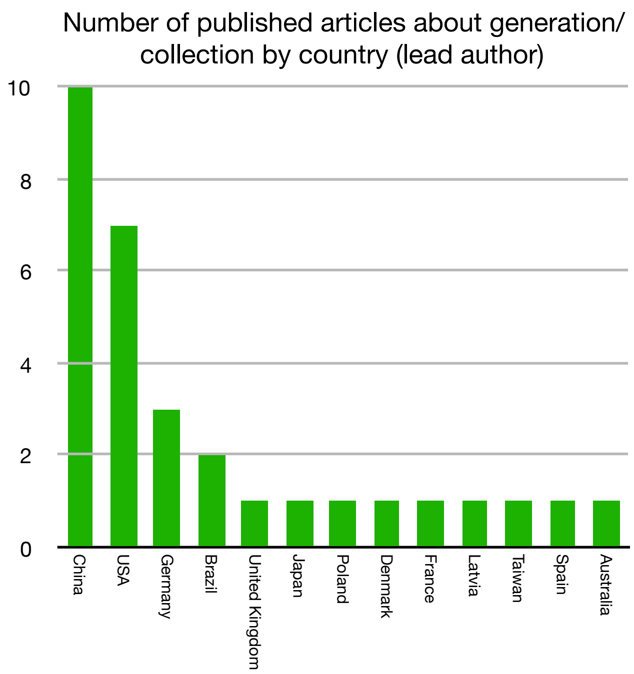
However, the area is extremely important because the result of both generation and collection affects the next step in the process. If no batteries are collected, there is no great need to recycle and the batteries that are collected risk inefficient recycling due to insufficient volumes.

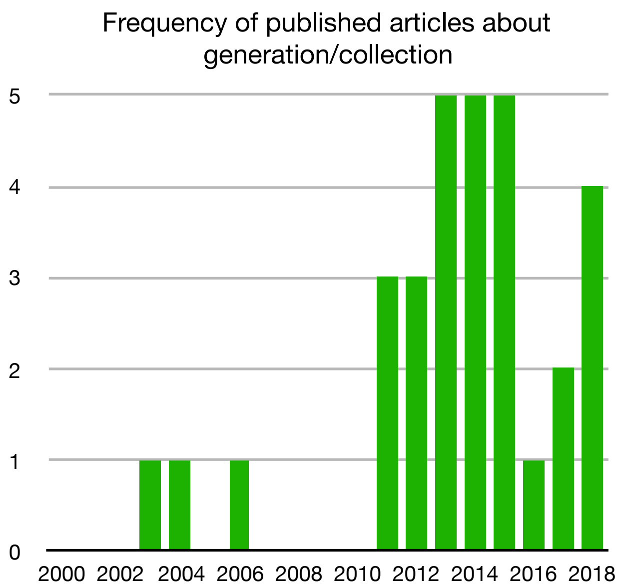
A related area is research about companies or countries' overall EOL strategy, where alternatives such as recycling and reuse are discussed based on how they can be used to optimise the entire EOL chain. The results in this type of study above all make clear what economic but also environmental incentives exist for collecting batteries.

Sub-areas that are relevant to study are:

- Longevity and nature of batteries put on the market
- Origin of and marketing channels for spent batteries
- Collection and business models as well as end-of-life strategies
- Collection and transport models
- Dangers and safety aspects of collection and transport
- Sorting, classification and decision on next use

Despite the area's importance for the entire recycling chain, there are only a few studies covering the areas above. In our searches, we have only found 31 reviewed research articles that concern the collection of lithium-ion batteries including articles on electronics and mixed waste batteries. The articles are spread around the world with 10 from China, 9 articles from the EU, 7 from the US and 5 from the rest of the world.





Longevity and generation of EOL batteries

While there are a large number of studies on the lifetime of lithium-ion batteries from a technical perspective (more in the section on reuse and LCA), it is surprisingly sparse with research around the lifetime of batteries from an EOL perspective. There is a large number of studies on eg life expectancy for mobile phones[21] and at what rate they reach recycling[22] [23] [24] [25] [26]. None of these studies, however, discuss how the collection of mobile phones affects the collection of the included batteries. When it comes to studying other electronic equipment with batteries, we have not been able to find any study that can be used as a starting point for how long it takes for, for example, laptops or tablets to reach EOL.

The only academic research on the emergence of EOL lithium-ion batteries can be found in a study on how much rare metals from batteries are generated by electronic waste, partly as a result of the EU project ProSum[27]. The goal of the project has been to create a data base with all available data such as volumes, material flows and treatment of electronic waste, EOL vehicles, waste batteries and mining waste. The data base which is called Urban Mine Platform[28] is available for anyone and contains very valuable information for forecasting or pure understanding of how the material flows look.

In addition, the recycling company Accurec has conducted a similar study[29] where estimates and forecasts are made of how much batteries have been put on the European market and how much is estimated to reach EOL and finally being recovered. In a report by CEPS made within the framework of the EU project Circular Impacts, forecasts have been made for how much raw materials will be available from recycling of electric car batteries in Europe 2040. As part of this, forecasts have been made for EOL flows of vehicles.

In the preparatory work for The Urban Mine Platform, flow analyses have been carried out taking into account both exports and imports of both batteries and equipment with batteries. Recycling has also been taken into account. However, in the database itself, it is not clear how much that becomes waste in Europe and how much is exported. The studies from Accurec and CEPS do not take into account exports at all. However, CEPS has taken into account reuse in its forecast.

Collection of batteries

Of the articles that specifically study the collection, most reviews describe the current situation in each country and, in many cases, what the situation is in the rest of the world. All of these studies are focused on collection of portable waste batteries of all different categories such as alkaline, nickel cadmium etc. In most cases, the studies are made by research institutions in countries where the collection has a short tradition or relatively low acceptance such as Latvia, Spain, Poland and Brazil. Among the studies are several where the researchers conducted simulations and modelling of various collection models where, for example, a deposit has been a component. Among the results, researchers highlight the importance of both financial incentives and a well-developed collection infrastructure.

These studies have equally high relevance for lithium-ion batteries as for other battery types in cases where the batteries are collected in a similar way. However, none of the studies highlights why lithium-ion batteries have a lower collection rate than other battery types or how these batteries differ from the others in the way described in this report's background chapter.

Only one study has been found that specifically examines the amount of batteries that are disposed of in household waste[30]. The study done in Denmark in 2013 shows that the amount of batteries that are disposed of in household waste is relatively high (4g per household and week) but that the amount of lithium-ion batteries is very small and only occurring in electronics. Instead, it is disposable batteries that account for the large amount.

Outside the internationally available research, a project has been carried out in Sweden on mixed waste batteries at Chalmers University of Technology[31]. Among the results was a model for calculating the collection rate of batteries and data showing three years of median age of collected AA and AAA batteries, which means that 50% of the batteries collected are older than that. The project has been continued in a second phase where the purpose is to develop better collection methods for both loose and device-bound batteries[32].

Research on lithium-ion batteries

Articles that focus on lithium-ion batteries usually do so from a more holistic perspective where the entire EOL chain is discussed. This chain looks different depending on whether it is portable or industrial batteries. Here, the studies can be divided into three different categories; portable batteries, electric car batteries and lithium-ion batteries in general.

The most complete study of lithium-ion portable batteries is done by Gu et al[33] and covers the entire recycling chain of batteries in electronics in China. The study describes in detail the various routes from consumer to recycler. The authors' conclusion is that less than 10 percent of the batteries are recycled and that extensive policy changes are needed to increase the collection. Another analysis of the Chinese market has been made by Zeng et al.[34]. The study that studies both portable and industrial batteries is also an overview of the entire Chinese recycling market for lithium-ion batteries and, as in the prior study, proposed policy measures, especially for the collection and handling of EV batteries.

Wang et al[35] has done a comprehensive analysis of how changed material values in the lithium-ion batteries will affect the collection and handling of batteries in the US. As the cobalt content decreases, the incentives for recyclers to collect batteries deteriorate, which, according to the authors, gives reason to change legislation regarding requirements for collection and recycling. The questions are also included in two of the authors' doctoral theses, where they make in-depth studies of the economic and environmental consequences of handling the batteries[36].

In a study by Winslow et al.[37] a similar analysis is made in which they suggest that lithium-ion batteries should be classified as hazardous waste, thereby being able to direct even less valuable cell chemistries to recycling. None of the studies look at how the reuse market affects the flows.

An additional analysis, which is based on material values in the Japanese market, has been made by Asari and Sakai[38]. The researchers have studied how batteries with high cobalt content come from users to recycling in Japan and investigate, among other things, what knowledge and attitude people have to recycling and what they actually do with their batteries.

In a report written by Boxall et al[39] the collection and recycling of lithium-ion batteries in Australia is studied. The study concludes that most of the batteries are put on landfill and that there is a great economic potential if the batteries instead are recycled. However, the report does not investigate whether the batteries are put on landfill, but the data comes from a previous consulting report where it is assumed that everything that has not been collected instead is disposed of. The authors conclude that the collection rate can increase through policy measures and improved communication.

Studies on industrial batteries that are not EV car batteries are with only one exception absent. The exception is a study of the collection of electric bicycle batteries in the Chinese market and is interesting not least because it addresses the difference in the collection rate between lead-acid batteries and lithium-ion batteries based on their respective material values.

There are no studies of the European market regarding the collection of portable lithium-ion batteries. Nor have we found any study that addresses the reuse of portable batteries and how it affects the flows.

Research on collection of EV batteries

When it comes to EV batteries, several studies have been done in China, the US and Germany, which were also the early countries with larger volumes of electric cars. Often the entire chain is studied, from the fact that the battery is considered to have reached EOL until they reach reuse or recycling.

A number of studies have been made which, based on batteries' values, both positive and negative, suggest different types of standards, goals and organisational structures. Hoyer et al[40] propose a higher recycling requirement as a means of control to ensure a high collection rate of electric car batteries while Xu et al[41] suggest standards for creating structure in the Chinese market for EOL batteries.

The importance of reuse is included in several studies in the US and China where it is pointed out that the values for batteries intended for reuse are considerably higher than if they are directly recycled.

In a study by Quiao[42] et al the economic consequences of recycling and how it might affect the collection was covered. The study found that recycling of electric cars with NMC batteries generates a gross profit of USD 474 after purchasing the vehicle for USD 569. The authors concluded that the cathode material in the battery accounted for over 60 percent of the total revenue. Although the study does not take into account reuse which generally generates even higher values, it is important as it points to the importance of the battery's value for recycling the car, which gives a clear indication of how the flow will look when the cars reach EOL .

There are two studies that specifically look at the life of EV batteries from a recycling perspective. A study by Natkunarajah et al covering the German market comes up with a number of scenarios how quickly the batteries come back and also takes into account that batteries will be reused in energy storage solutions. A study made by Richa et al[43] analyses the same thing for the US market. Both studies are very useful for understanding the rate at which batteries can be recovered.

In addition to the reviewed articles, the collection area has been studied and results have been published within the framework of a few projects. In the European research project ELIBAMA[44] with the aim of strengthening and accelerating the creation of a European electric car battery industry focused on large-scale production of electric car batteries, the comprehensive logistics chain around EOL batteries was studied in a work package. The project proposed various standardisation measures in three different areas: EOL logistics, recommendations for easy disassembly of batteries and standardised eco design for improved recycling.

There is a need for further research, not least because the area is changing so quickly. However, it must be said that within this particular area, a knowledge base is being started with several studies that can be further developed when comparisons are made in other countries and when more primary data from eg reuse installations or more full-scale recycling can be evaluated.

Additional areas within generation and collection

What is most remarkable in this area is the total lack of studies on transport or physical handling (before disassembly) of lithium-ion batteries. There is only one article about safety, from the research project Lithorec[45]. Other available documentation is presentations of corporate experiences and innovations from different conferences. This despite the fact that the area has a very important importance for both costs and flows in the recycling chain and often gets a lot attention in industrial conferences.

What has also not been found is some form of experiment or pilot project that tests ideas or hypotheses within collection, but all research is descriptive.

Together with the relatively few studies that still exist, as well as the lack of studies on the export and re-use of portable and other industrial batteries, the conclusion is that generation and collection as a research area is strongly underdeveloped. Although several studies contribute with very useful information for both the industry and the research community, it is not possible with such a small basis to determine what is state-of-the-art in the area. Here is a huge need. Not least, since it is not possible to exclude the fact that it is precisely the sparse knowledge about collecting and trading which is a major contributing factor to why researchers in other parts of the EOL area draw completely wrong conclusions about collection levels and reasons why the recycling volumes are so low in Europe and North America.

Research about reuse

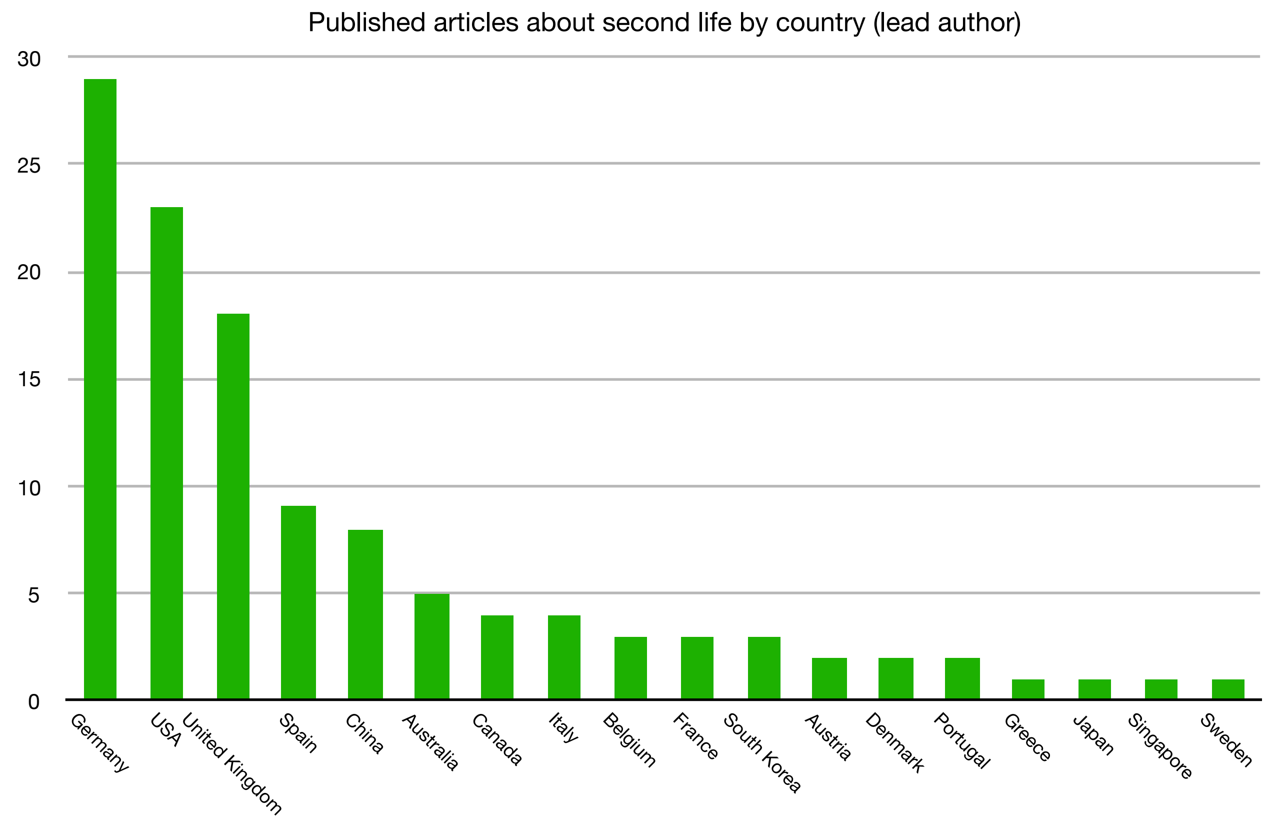
As the market for electric cars has matured, research on battery life and alternative uses has also increased significantly. The search phrase "Lithium-ion second life" in Google Scholar gave 2010 21 hits and has since increased annually and gave 2018 the search 252 hits. Many of these studies, however, are not specifically about reuse but, for example, about batteries' ageing or about general conditions for EVs.

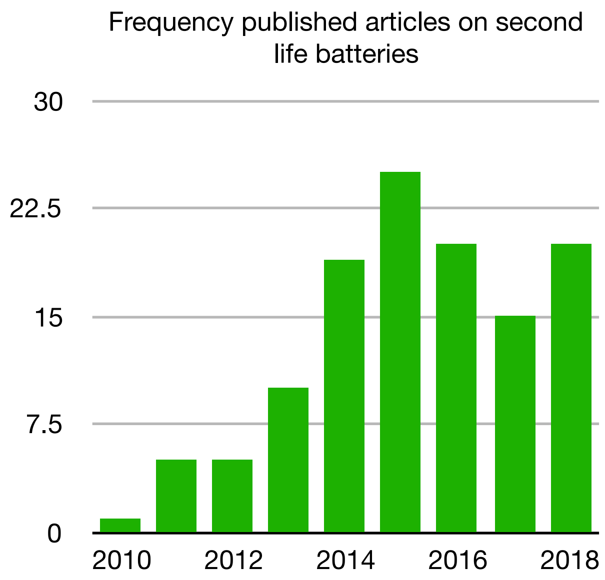
In our compilation we have found 166 published studies which are focused on reuse of lithium-ion batteries or which are particularly relevant to the area. Examples of the latter are studies dealing with ageing, monitoring and control, and where the authors have specifically pointed to the possibilities of reuse.

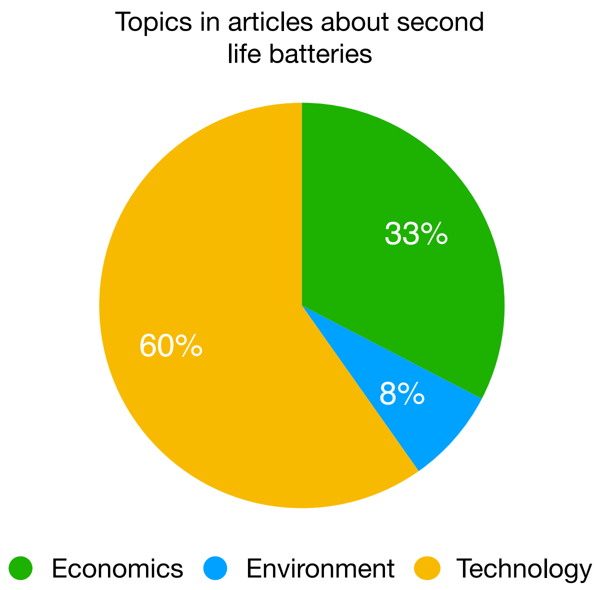
Unlike the collection and recycling area, the development of reuse has been much more driven by academic research institutions. Several of these studies, however, have been parts of larger industrial projects, not least within the EU's Seventh Framework Program and in national research projects. In total, six EU and national projects have published their results, while two are ongoing, which are presented in a separate chapter later on. Several of the EU projects have produced research that has been reviewed and published.

There is therefore a large number of studies with results that a player with ambition in the reuse or Second life industry can use directly to make decisions and directly utilise in technological innovations.

Judging from where the research has been done, it can be stated that it is primarily a European area of interest with Germany, the UK and Spain in the top four, though with the USA in second place. Then comes China and Australia. These countries are also the leading countries in energy storage, which has a very clear connection to the use of Second life batteries.







Interest in the area has increased sharply, but compared to, for example, the recycling area, reuse is still a new phenomenon. It also becomes clear in the actual research content which follows a fairly clear pattern and from a (short) historical perspective one can observe partly a number of hypotheses that have been confirmed and falsified, as well as a number of common research topics.

Above all, the recycling area has been studied from three different perspectives:

- Technical possibilities and limitations
- Economic potential
- Environmental impacts

As already mentioned, reuse is also included as an important aspect of research on the batteries, mainly in the context of the battery's ageing and in studies and development of systems for monitoring and controlling batteries.

Technical opportunities and limitations

From a technical point of view, the focus in the Second life area has predominately been on:

- Quality and remaining capacity in used electric car batteries
- Test and verification methods for determining reuse potential for batteries
- Control of batteries in recycling applications
- Applications for recycle batteries
- Potential lifetime of batteries in recycling applications

Studies on the quality of the batteries and remaining capacity primarily deals with how the life of the batteries in the car can be optimised and what potential they have for reuse in Second life applications. Overall, the results are positive with a general view that the batteries are both expected to have a long life in the cars and that they will then also have sufficient capacity to be used in new applications. Neubauer et al[46] notes that batteries are certainly degraded during their life in vehicles in several cases more than the often assumed 80 per cent, but in spite of that, there are great possibilities to use them in new applications. At the same time Martinez-Laserna et al[47], who conducted a comprehensive literature review on the reuse of electric car batteries, points at a lack of research that takes into account how quickly batteries are degraded when they are then used in the actual reuse application.

For a buyer of batteries, this is, of course, absolutely crucial in order to be able to verify the potential for e g an energy storage application. In their literature study, the authors also point out that there is still no knowledge of how age affects the performance of the batteries, as this in principle cannot be tested in forced experiments. The authors also showed that batteries that reach the so-called "ageing knee", where the degradation accelerates, cannot be used in neither vehicle nor energy storage applications.

In line with these comments, it can also be noted that the vast majority of reuse studies are don through modelling and simulation of data. There are still no published studies of batteries that have been followed over a long period of time and especially not under actual conditions.

An area that can be of great importance for the battery's degradation is its control and monitoring, both when the battery is in the car and when it is then used in Second life applications. We have identified several studies showing positive results on both the actual wear and tear of the batteries by, for example, active balancing[48] [49] [50]. Likewise, there are a number of articles describing new technologies for monitoring the cells and thus gaining better knowledge of how the battery has been used [51] [52] [53].

As far as applications are concerned, many previous studies have not only asked whether it is possible to make money from Second life batteries in energy storage applications, but even seen reused batteries as an absolut enabler for the applications. Early studies therefore had, in many cases, focused on applications that were previously not profitable with new batteries such as "off-grid" solutions[54] and backup power in rural areas or in developing countries. The most common applications here are integration with solar cells.

The focus has gradually been broadened and today there are research results on the integration of reused batteries into larger systems that support national power grids [55] [56], as well as smaller applications such as EV charging or backup power. What is clear and what is also mentioned in the literature study from Martinez-Laserna et al is that there are no studies on larger systems and not least systems used by the major energy storage actors.

An area that is followed with great interest by the players in the reuse area is the testing of batteries. There are studies that look at different methods for this [57] [58]. However, this is an area where one could have expected more research, since in many cases it is considered a crucial factor for efficient use of batteries in new applications.

Economic potential

Over 30 studies have been identified on the economic potential of recycling applications. These studies, as in several cases, are based on modeling, mainly with regard to the degradation and longevity.

An early aspect that was studied was whether the revenues from recycling applications have the potential to reduce the cost of the electric car. This should be seen in perspective from the fact that an electric car battery accounts for between 30 and 50 percent of the car's cost. If part of this cost could be charged in subsequent applications, the price for the car could be lowered. In the literature study from Martinez-Laserna et al[59] it is noted that there are certainly opportunities to make more money on the battery in new applications, but that the constantly reduced battery price will make the difference less.

Great emphasis has been placed on assessing whether there is sufficient revenue-generating services and product - market-fit for the batteries. Here it is important to take into account that many of the studies have been done during a time where one tries to find and clarify the business models also for new batteries in e g energy storage systems for home use.[60]. The results in most studies are overwhelming, where it is believed that there are great opportunities to make money on reused batteries, especially in energy storage applications[61] [62]. Even more studies have focused on the cost structure for reuse and compared this with the costs for recycling. Here, several researchers note that it is important to work with an efficient production chain in order to bring down the cost which is required as the batteries must be sold with a proper discount in relation to new batteries due to their shorter lifespan. Studies and modelling that investigate the costs for dismantling and testing of the batteries have therefore direct interest for the industry.

An additional economic aspect is different business models for both selling used batteries and making money from reusing them, but also how this is integrated with the ownership of the electric car.[63].

The overall result is that researchers believe that there is a great potential in reuse of batteries from electric cars and that there are currently established revenue models for this. However, the area is moving very fast, which means that both cost and revenue calculations can soon only be outdated, which can also affect the actual dynamics in the market.

Environmental consequences

We have identified seven studies that carry out life cycle analyses focusing on Second life batteries. The answers in the studies are relatively unambiguous[64] [65] There are great advantages in reusing batteries in energy storage solutions and other applications as it increases the life of the batteries, which means that a smaller part of the batteries' climate footprint will be attached to the car.

It is also positive to replace new batteries with used ones, which makes the climate footprint for renewable energy even smaller. As in the case of the economic benefit, second life batteries contribute to less emissions by being part of, for example, energy storage solutions that allow renewable energy to be utilised more efficiently and even more when it replaces gas or diesel-powered power plants used for creating temporarily increased capacity in the power grids or to regulate the frequency on the network.

Reuse of other kind of lithium-ion batteries

As shown in the introductory chapter, electric car and electric bus batteries are only one of many applications for which the lithium-ion battery is used. So far, batteries from portable electronics are the largest stream of spent batteries, and the reuse market for these is both large and driving the global volumes. Nevertheless, there is very little research on reuse of batteries other than those from cars. The only study we found is a Korean article where cell phone batteries in combination with photovoltaic cells are used to supply lamps with electricity[66].

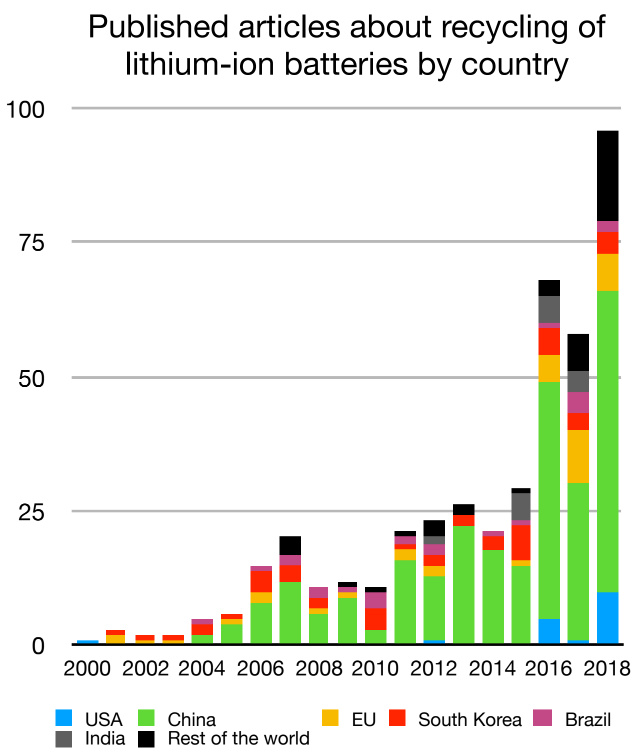
Research about recycling of lithium-ion batteries

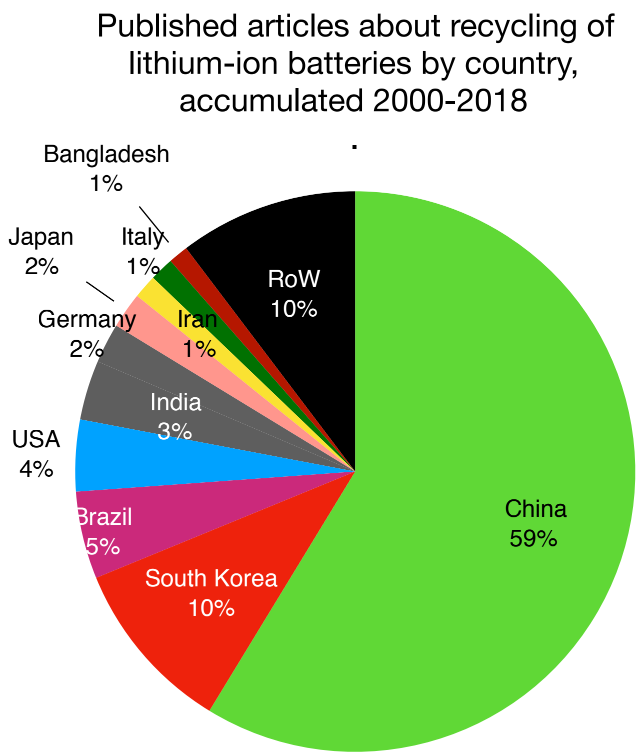
If research on the collection and re-use of lithium-ion batteries is fairly sparse, the research on recycling processes is considerably more extensive. Since the first articles on recycling methods were published at the beginning of the new century, extensive knowledge has been built up on various processes for treating batteries, extracting materials and reusing the material in new batteries. A total of 432 articles has been identified in this compilation ranging from individual recycling processes to studies on the characterisation of batteries for recycling. There are also a large number of reviews in the form of literature studies that effectively summarise the area.

At an overall level, two things become very clear. First of all how early the research and collective knowledge about recycling was established. Secondly, how much of this research that is dominated by primarily China and secondly South Korea. Of the research published between 2000 and 2018, 59 per cent of the articles have a lead author from a university or institute in China. In many cases, exclusively Chinese research groups are behind the studies. The diagram below clearly shows that this is not a new phenomenon but China and South Korea already dominated the area after a few years into the 21st century. What is also interesting is that some countries that might not be expected to be among the leaders in the area have produced quite extensive research, such as Brazil, Bangladesh and Iran, but also Egypt, Indonesia and Malaysia. Sweden is represented by one article.

South Korea and China's dominance also applies for filed and granted patents. In a previous study, Circular Energy Storage has reviewed filed patents in the recycling of lithium-ion batteries and found that the number is even larger than the published studies, and partly that the area is completely dominated by China, South Korea and Japan[67].

The reason for this is linked to the content of the research. Together China, South Korea and Japan account for over 90 percent of all lithium-ion battery production and are even more dominant in the production of the materials included in





the batteries as cathode, anode and electrolyte material. It is also this perspective that completely dominates the research on recycling - how to use waste batteries to produce new materials for batteries.

The research about recycling can be divided into three main categories:

- Recycling processes (from battery / production waste to separated substances or recycled components)
- Production of materials from recycled batteries
- Pre-treatment processes (sorting, dismantling and discharging)

In many cases, the categories as studied together as different recycling processes, for example, have different requirements on pre-treatment and at the same time provide different conditions for the following production of new materials from the recycled batteries.

In addition to these main categories, there is also research on how to use other substances instead of, for example, natural graphite to produce anode material and there are also several studies on how materials from lithium-ion batteries can be used in other products such as other battery types, super capacitors or materials in chemical processes [68] [69] [70].

Recycling processes

Recycling of lithium-ion batteries is often divided into three different processes: mechanical, pyrometallurgical and hydrometallurgical processes. However, in fact these are not alternative methods but rather methods used in combination with each other. In literature and studies describing the recycling of lithium-ion batteries, pyrometallurgical and hydrometallurgical processes are sometimes considered equally important. However, in the case of primary research on the actual processes, hydrometallurgical processes are in a strong majority. For example, in a compilation on general battery recycling[71] from 2004 processes from various industrial players are listed where pyrometallurgical processes are in the majority. Three different pyrometallurgical steps are also described: pyrolysis, reduction and incineration. But there is no reference to actual research that describes similar processes. In reviews that follow, pyrometallurgy does not have the same prominent role. In a compilation of Xu et al[72] from 2008, "thermal methods" are listed under the category of physical methods. The only method described is a pyrolysis method used in combination with a mechanical and a hydrometallurgical method.

In a review from 2013, Zhang et al separate treatment from pre-treatment when listing different recycling methods[73]. In this context, pyrolysis is described as a way to prepare lithium-ion batteries before they finally are recycled in either a mechanical or hydrometallurgical process. In the same study the researchers also gives an overview of available processes in which they state that several of the companies that recycle lithium-ion batteries use pyrometallurgical reduction processes originally intended for other materials, and even in cases where dedicated methods are used, the authors mean that the possibilities to recycle all substances in the batteries through pyrometallurgy are worse. A recently published compilation of lithium-ion recovery processes by Li et al[74] makes the same distinction describing pyrometallurgy only as examples of industrial processes.

Instead, most focus is on mechanical and hydrometallurgical processes. In the reviews of both Xu et al and Li et al, the processes are divided into physical and chemical:

Physical methods

- Mechanical separation - The feed material goes through a number of mechanical steps, including shredding or crushing, sieving, air, water bath and magnetic separation. The step is primarily seen as a pre-step to leaching or other chemical separation and is mainly for separating the outer casings of the batteries and, if possible, separating the coated foils of aluminum and copper.

- Dissolution - By dissolving the crushed material, or whole films with anode and cathode material, often in an NMP solution, the active material can be separated from aluminum and the copper foil. The problem according to the authors is that the process is expensive and creates a lot of waste.

- Mechanochemical Separation - A mechanical treatment that produces a chemical reaction in the material. Used primarily for LCO batteries

- Thermal treatment - An alternative, especially to dissolution, as heating the batteries to 350 degrees dissolves the binding agent between the current collectors and the active materials, which means that cathode and anode materials can be separated in the next step, either by mechanical or hydrometallurgical processing. The method is in other studies regarded as pyrometallurgical and can also, in higher temperatures (pyrolysis/calcination) be used to separate, or facilitate separation of the various active materials. Pyrolysis mentioned earlier is often described as a step with great potential as part of a chain of several processes.

Chemical methods

- Separation with acids (leaching) - A crushed material is treated in a dilute acid to achieve chemical reactions where substances form new compounds. The most common acids are H2SO4, HCl and HNO3.

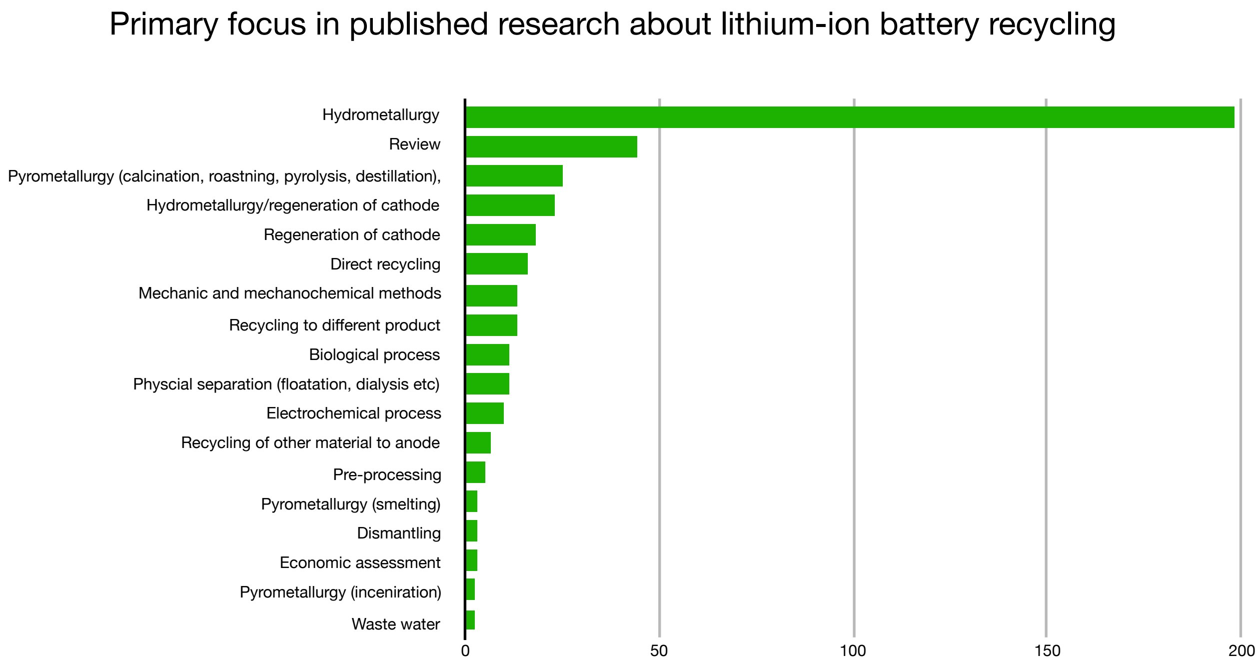
- Bio-leaching - Biological substances such as bacteria are used instead of acids, or to generate acids, to separate the various substances.

- Precipitation - Cobalt and other substances are separated in a process in which the temperature and pH value are actively changed to make the material solid and non-dissolvable.

Both of the reviews above show that all processes have been tested in several research projects, which is confirmed by this review as well. Most often, several steps are tested in different experiments, which creates an overlap between the methods. But by classifying the studies based on their main results, we can conclude that out of 312 studies we identified based on primary data, just over 70 percent refer to different types of hydrometallurgical methods, while barely 15 percent and 10 percent refer to physical and pyrometallurgical methods. Almost 4 percent of the studies are about biological extraction, which is usually also a hydrometallurgical method in which bacteria or other biological materials are used to react with the battery material.

Many studies on hydrometallurgy are based on similar principles and their main objective is to investigate different hypotheses in, for example, leaching methods with different reagents and reductants, temperature, time and various types of mechanical or other physical activation such as use of ultrasound or stirring. The most common reagents are sulfuric acid and hydrochloric acid. However, there are a large number of studies in which different types of organic acids are used, such as ascorbic acid and citric acid [75] [76] [77].

Mechanical and mechanochemical methods are most often studied as a pre-step to hydrometallurgical processes, but a small number of studies have been done on whether they can function independently. Usually, the final product is a black powder containing a mixture of both anode and cathode materials, but several research projects have developed



methods in which, by means of sieving and magnetic separation, it has been possible to separate reusable cathode material, especially for LCO and LFP batteries. More common, however, is that this is done in combination with various types of thermal treatment methods.

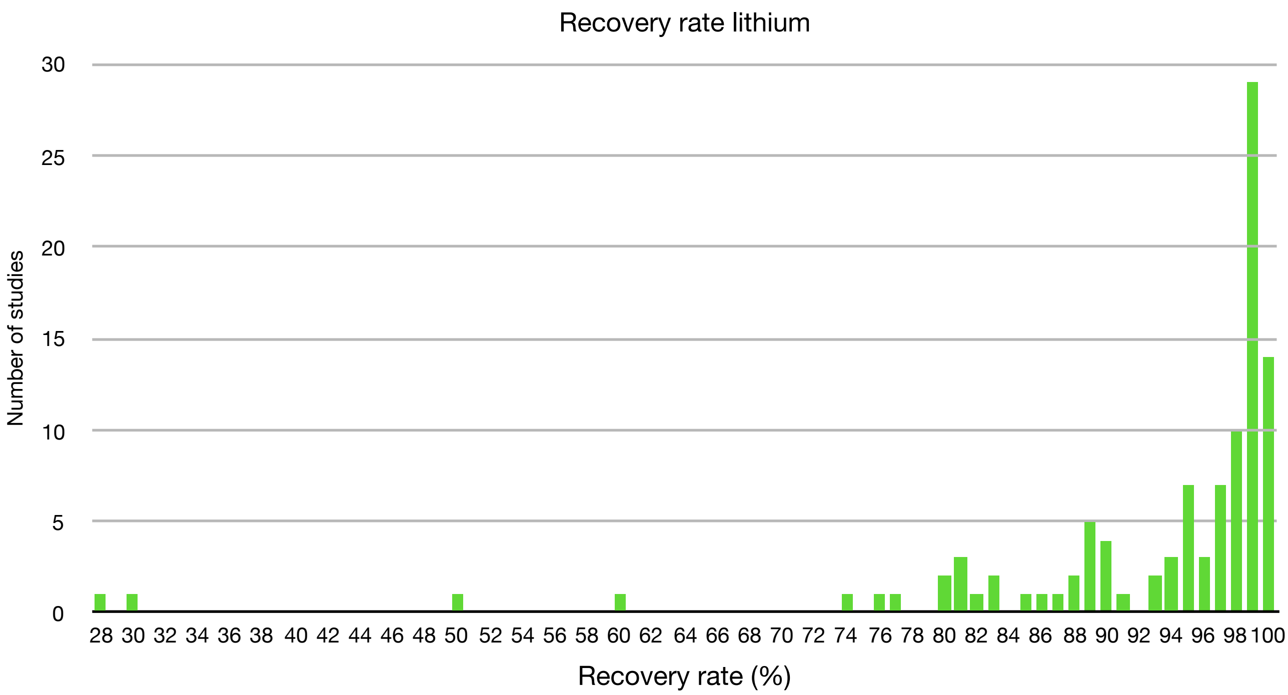
Combinations of methods for accomplishing this are sometimes called cathode-to-cathode recycling. However, the principles for cathode-to-cathode recycling are generally quite different. For LCO and LFP batteries, there are several methods where the materials are kept intact so that they after further thermal treatment in which the cathodes are re-lithiated, can be used again[78 79]. This is also called 'direct recycling'. For NCM/NMC batteries, however, it is more common for the substances to be separated first and then re-joined. One reason for this is that the input material is rarely the same as the desired new cathode material.

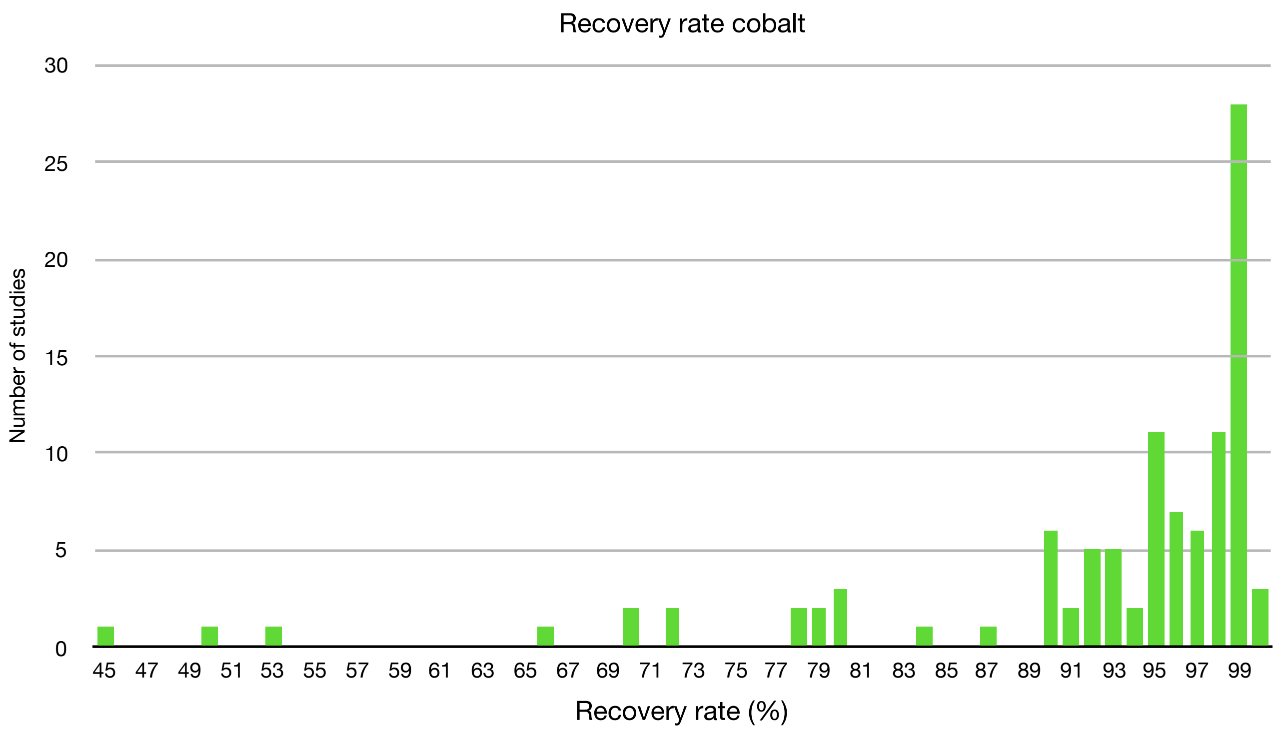
Most studies on pyrometallurgical steps relate to pyrolysis and calcination where the process is a part that also includes mechanical and/or hydrometallurgical processes[80 81 82]. Only three studies have been identified that are about smelting and one about incineration [83 84 85 86].

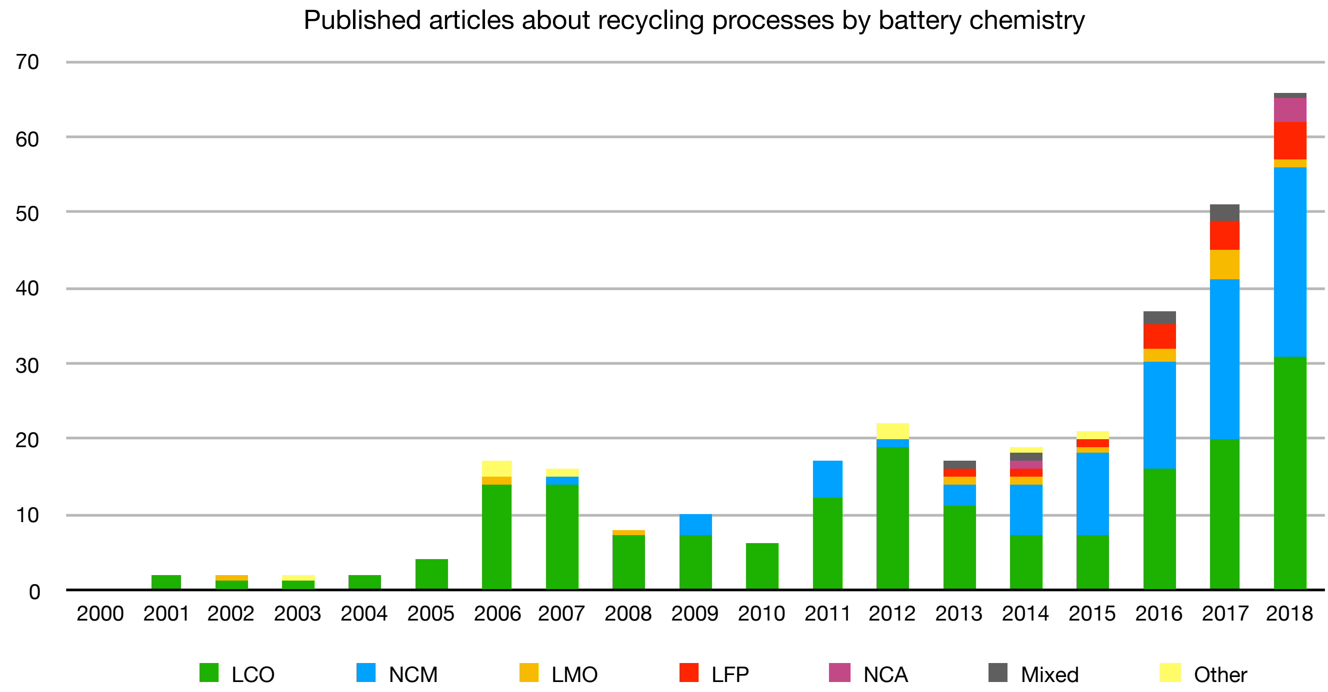
The efficiency of recycling in the various studies is usually high. Li et al lists 20 studies with an efficiency for lithium often at 100 percent while nickel, cobalt and manganese generally have an efficiency between 80 and 99 percent. This too is well in line with this overview. While a few studies have shown a recycling efficiency of 30-70 percent of different substances such as copper, aluminum and lithium, the results are usually between 90 and 100 percent. We have identified 80 studies where more than 90 percent of lithium has been recovered and 86 studies where over 90 percent of cobalt has been recovered. In addition, there are 18 and 20 studies, respectively, where more than 90 per cent of LCO and NCM cathodes have been recovered. In most cases, the purity rate (when these are stated) are high and in some cases their levels have been set as requirements for the material to be included in the recovery efficiency in the first place.

Worth mentioning is that the vast majority of the studies have been carried out in lab scale, which entails good control of the material and ability to prepare it so that it is optimal for the processes. Separation of cells has often been done by hand, which is rarely the case in an industrial process. At the same time, however, there are patents covering similar methods where the methods are based on industrial principles.

In the research reviews mentioned above, no actual evaluations are made of which process is most state-of-the-art. Nor is it described as a need for further research, but instead the authors point out that further in-depth research is required, for example in biological methods, which are considered to have great potential through its limited environmental impact.







Recycled cathode chemistries

In the same way as hydrometallurgical methods dominate the research, the same applies to which battery chemistry the methods are intended for. For natural reasons, LCO batteries have long been the most common type. These batteries still represent the largest amount of waste batteries since they have been the longest in the market. Over time, however, it becomes clear that NMC (or NCM as the cathode chemistry is called), which is the most common cathode chemistry for electric cars has been the main focus of research.

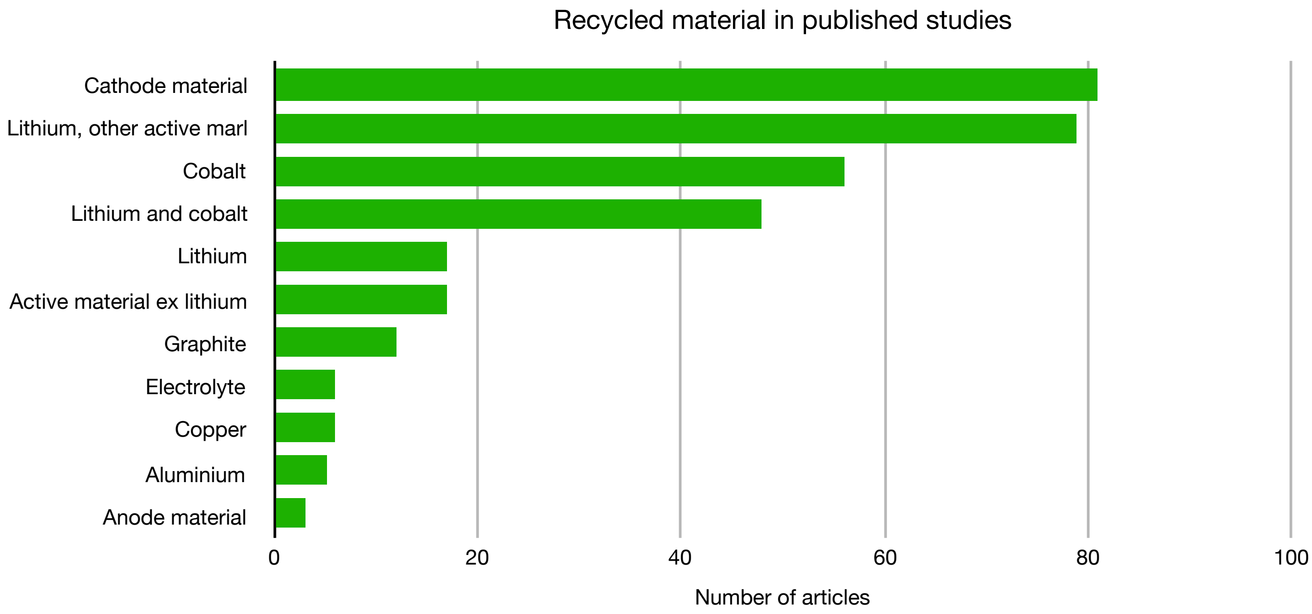
Research about recycling of LFP, LMO and NCA batteries has been significantly more sparse. This is remarkable, not least for LFP and LMO, since these chemicals are common especially in China, and they create special challenges as they do not contain cobalt and nickel, which means that the material values are relatively low. However, it is possible to see some increase in research on LFP recycling, which goes hand in hand with the industrial development in China. However, LFP is a relatively common chemistry even in Europe, especially in buses and in energy storage.

Regeneration of battery materials from waste batteries

Many of the studies on the recycling methods also include methods for creating different types of products from the recycled material. In particular this is the case in Chinese research, where it is clear that this is the ultimate goal for all recycling. It is also a natural step after hydrometallurgic and mechanical recycling methods and is probably another reason why these particular methods are so dominant.

In principle, six different ways of recycling can be distinguished:

- Battery Chemicals
- Preference to cathode material (precursor)
- Cathode Materials
- Anode material
- Electrolyte
- Materials for other applications



The distribution of the studies between these areas is relatively even over the first three categories. Recycling of anode material (graphite) and electrolyte, both of which have significantly lower values than the cathode material, has been studied much less as well as to use the material for other applications.

Li et al list 21 references distributed over three types of methods (sol-gel, thermal and co-precipitation) to create precursors. The methods differ depending on the cathode material in question. In our compilation, we have identified 81 studies with completed experiments to produce cathode materials and 78 studies focusing on lithium and other active materials in order to subsequently convert it to tradable lithium products such as lithium carbonate or lithium phosphate, or precursors. In cases in which new cathode material has been produced, the studies often are finished tests of the new regenerated cathode under real conditions, often compared with new, unused, batteries. The results are in many cases remarkably positive and in most cases the authors draw the conclusions that recycled cathode material can be used for real production of new batteries.

This areas is clearly dominated by research groups from China and South Korea, which together account for 84 percent of the studies. In addition, there are 138 studies focusing on recycling of lithium and cobalt separately (not as $LiCoO2$), lithium separately or cobalt separately. In these categories there is a larger proportion of non-Chinese studies. There are also projects where LCO batteries were used to obtain lithium and cobalt for use in NCM and NCA batteries.

Pre-treatment processes

Something often described as a problem in lithium-ion recycling is the increasing amount of different chemistry types, which makes it difficult for recyclers to correctly classify and sort the batteries. Another problem, particularly for hydrometallurgical processes, is the disintegration of the modules and cells which can be both difficult and time consuming. A third problem which is closely linked to the second, is that lithium-ion batteries are flammable and can catch fire or explode when exposed to mechanical impact. For this reason, it is quite remarkable that research on sorting, disassembly and discharge of batteries is highly unusual.

Sorting

The need for sorting and classification is mentioned as an important prerequisite for efficient recycling, but we have not found a single study that describes a method for sorting lithium-ion batteries. Rather, it is only regarded as an obstacle and an almost insurmountable problem, which is bad for reality. Three studies have been found on the classification of batteries [87] [88] [89]. This is a first step in a sorting process and thus contributes positively to the area.

Dismantling and disintegration

Dismantling of cells and packs has been studied in several contexts [90] [91] [92] [93] [94]. Above all, the research is about how to automate the process, which, not least in a European perspective, is necessary to make recycling a profitable one. The majority of the studies are about how to separate battery packs from electric cars and how this can be automated with the help of, for example, robots. Only two studies concern methods for disintegrating the components of cells in any other way than by shredding and crushing.

Nor have we found any research that specifically studies shredding, crushing or any other automated disintegration method. Several studies describe the various methods that are available in the industry, for instance to crush battery cells in an inert environment or after either being thermally treated or frozen but there is neither any analysis of the actual methods or experiments in order to evaluate them. This creates a gap between real world conditions and many of the studies of recycling processes, as these are often prepared by manual separation of anode and cathode.

Discharging of batteries

Discharge is described in many studies on treatment methods [95] [96] [97] but rarely in detail. Again, it is industrial practice that is described or method steps in lab environment. The only study that actually test different methods is made by Ojanen et al where different water solutions are tested to achieve optimal discharge[98].

Research on the environmental impact of batteries

One issue that frequently is reported in the media is the influence of lithium-ion batteries on the environment. In particular, this applies to electric car batteries, which are then often compared with cars that run on traditional fuels or with other alternative technologies such as fuel cells. What happens to the batteries in their final stages plays a potentially important role in this context, since efficient recycling and longer use of the batteries in Second life applications potentially can improve the impact of the battery, and thus also of the electric car. Likewise, an inefficient recycling would mean the opposite.

In this overview, 80 published articles or reports have been found that describe the environmental impacts of batteries themselves, or of the batteries' value chain, usually in the form of a life cycle analysis. Of these, 40 articles are studying the actual battery, while 25 articles study the actual vehicle and compare the results with traditional vehicles. In addition, there is a small number of articles that has studied the environmental impact of the battery in energy storage systems, portable electronics and aircrafts.

The life cycle analyses of batteries have been made at several different levels and with different comparisons. The earliest studies which were published around 2010 have compared lithium-ion batteries with other chemistries such as nickel cadmium, nickel metal hydride and lead acid batteries[99] [100] [101]. The data used in the studies was very limited, not least for the production phase. One of the most cited studies is an article by Zachrisson et al[102] where, on the one hand, a calculation is made of the energy requirement in the production bottom-up by starting from the actual process, and top-down, by using a battery manufacturer's electricity consumption distributed on the volume produced. In several following studies, this data is supplemented and replaced with new calculations usually from own lab studies[103] [104] [105]. Only one study has been found that uses primary data from actual production[106] where a battery from a Ford Focus, produced in the United States, is studied.

Several surveys that gather the data from previous studies have been found, among others in IVL's widely referred study from 2017[107] but also, for example, The International Council of Clean Transportation[108]. In principle, all these studies agree that the greatest impact comes from the production of the cathode in the battery, both in terms of climate impact and several other parameters. However, there are very large differences in the values between the studies. The reasons for this are several, such as different energy mix values but also completely different types of data. This is because many studies have different purposes, such as comparisons between different battery chemistries.

Most studies are so called "cradle-to-gate", i e from extraction of metals to the battery, or the vehicle, leaving the factory. This means that recycling is usually not included in the calculations, and even more rarely, reuse. Zachrisson et al, which cover transport to recycling but not the process itself, justifies this by the fact that recycling should in that case be an input value in the material and should be regarded as a separate process. The exception is a research group at Argonne National Laboratories in the United States, which in several studies has highlighted the role of recycling in life cycle analyses of lithium-ion batteries[109]. Among other things, the researchers believe that the energy consumption for extraction of eg aluminum, nickel and copper in the batteries can be reduced by 63 to 75 percent, which should be included in the life cycle analysis.

Lifecycle analyses have also been conducted that only look at the recycling process of lithium-ion batteries.[110] [111]. None of the life cycle analyses carried out on the recycling process go particularly far into the depths of the different effects of processes. Hendrickson et al[112] and Boyden et al[113] both distinguish between hydro- and pyrometallurgical processes, as does Dunn et al in his life-cycle analysis involving recycling. However, the values are not taken from real businesses in all cases. Neither of the authors have made any actual valuation of which the real risks are with the recycling operations.

Since most studies have been made cradle-to-gate or, in some cases, as far as cradle-to-wheels, the importance of reuse in most cases has not been discussed. The exceptions are two reports, one of which is the result of the EU project SASLAB[114] where a life cycle analysis is done of the actual reuse phase. The other is a study[115] in which an analysis of reused batteries in an energy storage system which was compared with a similar system using lead-acid batteries.

Only one study has been found that takes the entire user phase of a battery into account. The study that analysed how the environmental impact of a battery from a Nissan Leaf is affected by degradation[116]. It shows that the climate impact of the battery can deteriorate as much as 29 percent if degradation is taken into account. At the same time, it is one of the few studies that is not cradle-to-gate and would probably be positively affected if it had been supplemented by a study on the potential from reuse.

Research about design for recycling

In order for both recycling and reuse to be effective, as much as possible should be done to facilitate these processes already before the battery is manufactured. This includes, for example, creating modular pack designs that allow cells or modules to be easily disassembled for repair, reuse or recycling. Methods for attaching and connecting cells as well as measures such as marking or standardising materials and dismantling procedures would also be included in this.

What is therefore surprising is that not a single published article has been found in this area.

The only thing that has been found is the British project AmplifII[117] which has had the purpose of manufacturing modular battery packs that can be used in various types in vehicles and applications. The project, which has been carried out in collaboration with car manufacturers, recycling companies and recyclers, has recently been given a continuation where the same actors will deepen the cooperation.

Research in Sweden

Research in the end-of-life area for lithium-ion batteries has increased sharply in recent years, in Sweden as we'll as in other European countries. We have found 13 completed studies that are relevant to the area. In addition, at least as many projects are ongoing. The research field is dominated by Chalmers University of Technology, which participates in 12 of the 27 completed or ongoing studies in Sweden. Chalmers has also participated in five major EU projects. Luleå University of Technology has been active in three projects, while several other universities and institutes participated in individual projects. Among the companies, Stena Recycling has participated in three Swedish projects and in three European projects while Volvo and Volvo Cars participated in two projects each.

The Swedish studies and projects follow the international distribution of areas studied relatively well. Ten projects/ studies on recycling have been completed or are ongoing, seven on reuse, four on environmental impact and three on collection.

The first study relevant to the area was the previously referred life cycle analysis on lithium-ion batteries by Zachrisson et al[118] which in an article from 2010 briefly discusses the impact of recycling. The study has had a great impact internationally and is quoted by many studies around the world.

A licentiate thesis from 2012 studied the lifecycle of commercial LFP batteries in conditions that mimic them in heavy hybrid vehicles[119]. The study does not address EOL issues directly but has high relevance for, above all, reuse. The same year, a licentiate thesis on lithium-ion battery material recovery was published, where experiments were done with Cyanex 272 and Cyanex 923 to be able to recover lithium from hybrid car batteries through liquid extraction[120].

In 2016, the first major projects were completed, all of them financed by the Swedish Energy Agency,

Two projects included new research on recycling methods. In the project *Återvinning av metaller ur uttjänta Li-jon batterier (Recycling of metals from spent lithium-ion batteries)*[121] Luleå Tekniska Universitet, Chalmers Tekniska Högskola, Kungliga Tekniska Högskolan and Stena Recycling cooperated to enable recycling of more substances than today from end-of-life lithium-ion batteries by developing data for resource-efficient thermal recycling methods. In particular, aluminum, copper and lithium were studied, but recycling of metals such as nickel and cobalt was also included in the evaluation of the results. In particular, it was concluded that aluminum from the batteries could be recovered in a smelting process together with aluminum scrap. For efficient recycling of multiple metals, including lithium from lithium-ion batteries, a combination of different methods was deemed to be required.

In the project *Hydrochemical and pyrochemical recycling of metals from batteries*[122] the purpose was to develop an energy and resource efficient recycling process for NiMH batteries. Since these have several substances in common with the lithium-ion batteries, the project also has relevance for lithium-ion batteries. The goal was to develop a previously verified process from lab scale to pilot scale. The results showed, among other things, that recycling of NiMH batteries can be done more efficient by means of mechanical treatment and subsequent sieving of the different materials. The result was published in an article in the Journal of Material Cycles and Waste Management. Previously mentioned project *Efficient battery collection with consumers in focus*[123] also has high relevance in the field. However, the focus was mainly on alkaline batteries.

A project with high relevance for reuse was *Battery sensor for state estimation*[124]*.* The purpose of the project was to contribute to more efficient use of lithium-ion batteries, especially in vehicle applications, through a new way of measuring battery conditions. The project examined the reliability and function of the sensor system when tested in vehicle-type batteries. Although the results were mainly about the efficiency of the batteries in their primary applications, the results are very important when systems for reuse are designed.

In 2016, a master's thesis was also written at Luleå University of Technology, *Recovery of Lithium from Spent Lithium-ion Batteries*[125], which studied the conditions of recovering lithium from a synthetic slag from previous pyrometallurgical treatment using hydrometallurgy.

In 2017, further projects relevant to the area were completed, although not with direct focus on EOL of lithium-ion batteries. A thesis in the treatment of nickel metal hydride batteries, a project titled *Integrerad hållbarhetsanalys av morgondagens batterikoncept*[126] as well as a literature review of research in life cycle analysis for lithium-ion batteries, *The Life Cycle Energy Consumption and Greenhouse Gas Emissions from Lithium-Ion Batteries*[127]*.* The latter, which was done by IVL, received great national and international attention when it highlighted the great environmental impact the actual cell production has, which means that an electric car has produced a significant carbon footprint even before it has been used.

2018, it's clear that EOL of lithium-ion batteries becomes an increasingly important research area when four projects with very high relevance are completed.

Within recycling, the Chalmers project *Flexible and efficient (hydrometallurgical) recycling of Li-ion batteries of different chemistry*[128] studied the possibility of separating organic components from spent lithium-ion batteries by pyrolysis.

A valuable addition to research in the collection of batteries is *Sustainable collection, aftermarket and recyling of lithium-based car batteries*[129]*.* The project aims to develop a sustainable system for collecting, aftermarket and recycling lithium-based car batteries and has participants in many parts of the value chain including dismantling and insurance.

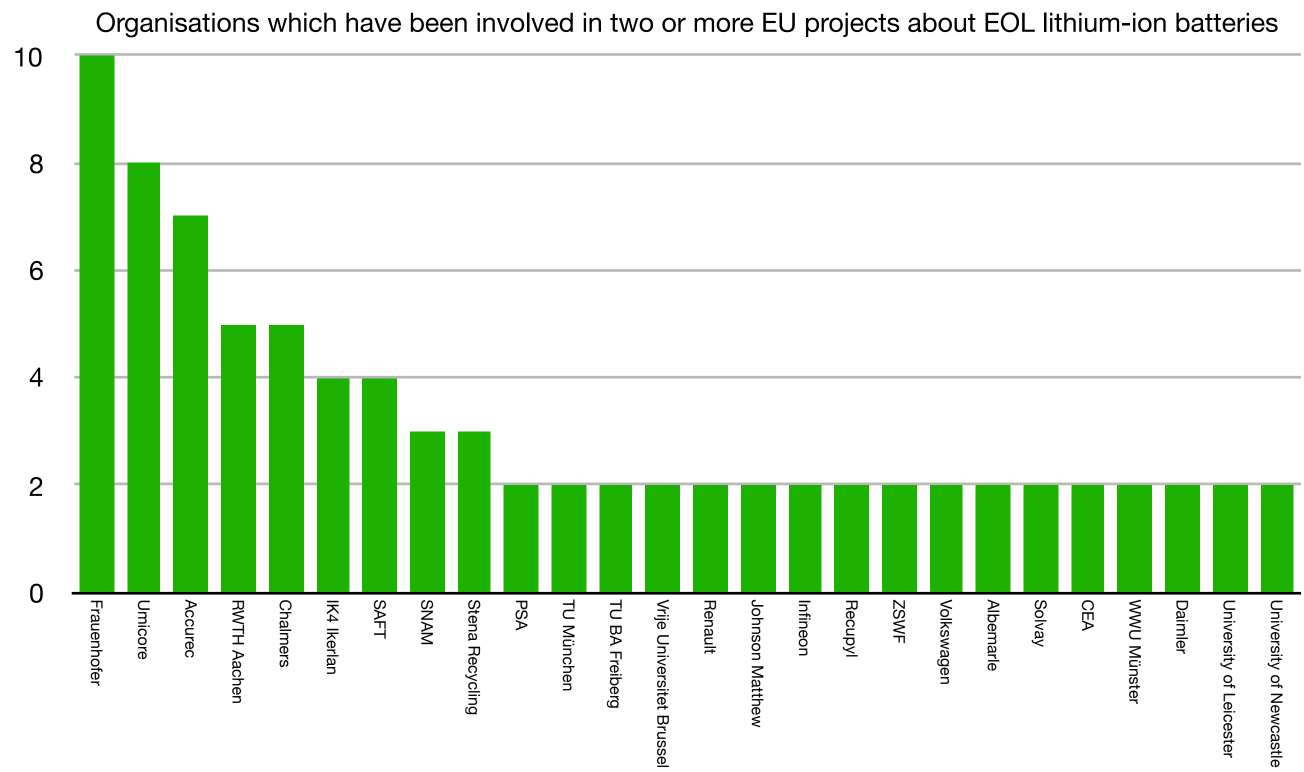
In reuse, two projects were completed during the year. The research institute Rise Viktoria conducted an stakeholder analysis in *Circular Business Models for Extended EV Battery Life*[130] where various circular business models were evaluated, including both recycling and reuse. Volvo, in collaboration with Riksbyggen and Göteborg Energi among others, carried out an installation of bus batteries that will serve as energy storage in a new tenant-owner association, Brf Viva[131].

Research on reuse and recycling in EU projects

Much of the research in Europe is done within the framework of projects funded by the Horizon 2020 research program and earlier the Seventh Framework program. Projects and individual studies have also been carried out within the framework of national research funding institutions mainly in Germany and the UK.

Since 2011, a total of 33 projects have been started where at least one work package in each has included recycling or reuse of lithium-ion batteries. Of these, nineteen have been completed or are just about to end. The various projects have a relatively large breadth with a focus on everything from recycling processes to description of the value chain. Four projects developed or tested hydrometallurgical recycling methods. As many have evaluated different types of reuse applications. Two projects evaluate pyrometallurgical methods and as many other studies they find that a combination of, for example, hydrometallurgy, pyrolysis and mechanical separation has the greatest potential.

Many of the projects are very similar. This may be due to the fact that they are often part of larger projects with the task of verifying the whole of the project, which means that the recycling part itself becomes the same irrespective of which whole entity is to be verified. For example, recycling methods for lithium sulfur batteries are evaluated in a similar way as for new optimised lithium-ion batteries. Many projects are also relatively bound by the project constellation, where, for example, projects with a recycler in the group often come to the conclusion that the desired method is exactly the one that the recycler uses in its business. It is generally also the same recycling company that occurs in the projects. Of the 33 projects that were either completed or still is ongoing, Umicore is included in eight and Accurec in seven. Then follows Stena Recycling, SNAM and Recupyl. The concentration is also evident on the academic side where Frauenhofer participates in ten projects while RWTH Aachen is involved in seven and Chalmers in five.



Of the EU projects implemented, it is above all one that stands out: Lithorec[132] (and Lithorec2). Lithorec is the only EU project that has been designed to develop and scale up a complete recycling process for electric car batteries. The method is based on a combination of thermal, mechanical and hydrometallurgical processes where the result has become a process with a very high recycling rate of most materials in several different chemistry. The project has led to a facility being set up which has been taken over by the company Dusenfeld. What also makes Lithorec stand out is the extensive academic publication, which has made the project considerably more accessible than many others. Partly, several articles have been published in various scientific journals, and a book has been published based on the project[133].

Another project that stands out is Batteries 2020[134] with a consistent goal of increasing efficiency throughout the use phase, from higher energy density in the cells to efficient reuse applications. This project is also very well published with several articles and with high activity among the project's researchers[135].

Previously mentioned AmplifII[136] is also a project where circularity is at the center with the development of modular packs that can be used in different types of vehicles and in stationary energy storage and development of recycling methods. As in the case of Lithorec, AmplifII has led to the commercialisation of several parts of the project, among other things with regard to recycling, where the company Axion tried to offer (limited) recycling of lithium-ion batteries.

A different project is ProSum[137]. The aim of this project has been to create a database for all available data on volumes, flows and treatment of electronic waste, EOL vehicles, waste batteries and mining waste. The database is available to anyone and contains very valuable information for forecasting or pure understanding of how the material flows look.

Ongoing research

Following the ongoing research within the EOL area is challenging. Mainly because it takes a long time between the actual research and the publication of it. We have therefore limited this area to an overview of known projects mainly in the EU and in the UK in the so-called Faraday program.

Of the nine ongoing EU projects that include EOL activities for lithium-ion batteries, almost half of them are similar to what's been described before, ie projects that cover all or a large part of the total value chain, and where recycling is only a part that should contribute to closing the circle. In the project Alion, for example[138] lithium aluminium batteries are developed while one work package is focused on recycling of the new chemistry. Most likely the methods used for this will not be notably different than today's methods.

However, there are a few projects that stand out.

The first is AutoBatRec 2020[139]. The project aims to identify efficient recycling methods for electric car batteries that are ecologically and economically sound and scalable. Every part of the value chain should be investigated such as a new type of collection, new transport concepts for automatic dismantling and sorting and finally separation of metals. This is one of the first projects of its kind and can be of great importance for the European end-of-life market if the results are successful.

The second is CROCODILE[140]. The purpose of the project is to demonstrate various metallurgical processes with the aim of establishing a functioning value chain for cobalt-containing materials and products such as batteries. The project includes a large number of recyclers such as Akkuser, Accurec, SNAM, Relight, Stena Recycling and Ecorecycling as well as cobalt companies such as Freeport and Glencore. With such a wide participation, the focus seems to be focused on just demonstrating existing processes in the different recycling companies with less focus on novelty. However, with the downstream players present there might be an interesting evaluation of which processes that are most economically and environmentally advantageous.

An additional project that is special is Close WEEE[141]. The purpose of the project is to increase the exchange from electronic waste where lithium-ion batteries is an important part. A new microwave technology will be used to extract substances such as lithium, cobalt and copper from batteries from mobile phones, tablets and laptops. If one can also address issues such as how to attract and collect the equipment, this project can be very important as electronics still ´ account for the largest amount of waste batteries.

Another project that brings the entire recycling value chain together is Norwegian LIBRES[142], where a large number of players from the collector (Batteriretur) to the downstream receiver such as Glencore Nikkelverk and Keliber participate. The project also includes Norsk Hydro, which has been assigned to develop a recycling process.

In the UK, several projects have been started within the framework of the Faraday Battery Challenge[143], which is a national program amounting to GBP 246 million. One of the first flagship projects was ReLiB, led by the University of Birmingham in collaboration with seven other universities and 14 industrial players. The project, which will be completed in 2021, will, among other things, produce a "triage" system for used batteries, autonomous testing of batteries, robot-based sorting, new recycling methods and development of new business models and an overview of British legislation in the area.

Within the framework of The Faraday institute, a large number of smaller projects are also carried out, such as lifetime extension of batteries (Advanced battery life extension), modular battery packs (AmplifII-2), new smart BMS (BABE), new recycling method for EV batteries (CALIBER) , re-manufacturing of batteries (R2Lib), as well as efficient value chains for re-manufacturing and recycling of EV batteries (VALUABLE).

In the United States, the Department of Energy has allocated USD 15 million for a Lithium Recycling Center[144]. The center, which will focus mainly on direct recycling of lithium-ion batteries, is led by Argonne National Laboratories along with Oak Ridge Laboratories and National Renewable Energy Laboratories. Some research groups have also been connected to the center from, among others, Michigan Technical University, the University of California San Diego and the Worcester Polytechnic Institute. The center was started in February 2019 and has funding for three years.

In Sweden, a number of major research projects are also ongoing. Two recycling projects are managed in Luleå. One, *Resource efficient recycling routes for discarded lithium ion batteries*, is led by Luleå University of Technology and aims to develop a concept that enables efficient recycling of metals such as Co, Ni, Cu and Li from waste lithium-ion batteries[145]. The focus of the study is to investigate a combination of metallurgical process steps for reprocessing the electrolyte from end-of-life lithium batteries through so-called mechanical activation. This makes it easier to separate metals in melting processes and subsequent refining steps.

The second, ReLion, is led by Swerim and will determine the conditions for a coherent recycling system for large-scale recycling of lithium-ion batteries, which should be able to handle copper, aluminum, lithium, manganese and nickel[146]. This should be done through analysis of material flows, studies of possible intermediates that can proceed to efficient processes in eg metallurgical industry, and practical tests of recycling methods. In the project description it says that it should be based on traditions in the Swedish metallurgical industry, which probably means that pyrometallurgical methods are at the core.

In the reuse area, the project "Second-use of Li-ion batteries from hybrid and electric vehicles" is underway[147]. The purpose of the project is to investigate the conditions for re-use of vehicle batteries in energy storage applications and include companies throughout the value chain, from vehicle manufacturers to energy storage developers.

A project that focus on a more limited problem but with potentially great importance is "BLACK - Battery Life and Use after collision"[148]. The project aims to verify the battery's electrical limitations in relation to mechanical shock, which can be used as a basis for future constructions of battery systems but the results should be at least as interesting for the EOL industry and for handling batteries in reuse applications when they have been in damaged vehicles.

Another niche project addressing an issue that can contribute to increased collection is REWEEEL - Automated location and disassembly of batteries from WEEE products[149]. The aim of the project is to develop an automated process for the identification and disassembly of batteries from smart phones and tablets, which can lead to an increased recovery yield.

Need for further research

What is research for?

There are, of course, several reasons to conduct research in the field of recycling and reuse of batteries. One reason is to develop new knowledge that can benefit the entire battery industry and take the area forward. Another reason, for example, for an individual company or for Sweden as a country, can be to develop its own ability to create efficient and profitable solutions and thus strengthen its own position in the value chain.

In the first case, research should focus on new areas, new technology or new methods, or verification of already made research. In the second case, the research can be seen as development of competence or technology transfer, and can be done in areas that need to be strengthened although it may not advance the research area as a whole.

The program description for the Battery Fund Program, which is Sweden's largest funding body for battery research, states that the purpose of the program is to:

- Generate knowledge, skills and technology development that forms the basis for future innovations in the field
- Achieve increased visibility for, and transfer of, research, development and new technologies between business and academia in the battery field
- Expand the area's breadth and volume to enable Swedish industry to maintain its strong position
- Indirectly contribute to increased system knowledge in the business sector around batteries and battery recycling.

It is also stated that the aim is to strengthen Sweden's position in research and development of battery technology and to develop new technical solutions and new products.

The purpose is thus double. Therefore, there is really no obstacle to developing methods in Sweden that already exist in other countries. At least not if the knowledge is lacking in Sweden. The areas that are considered to have great future potential in the EOL area, as well as within the battery area as a whole, and where Sweden lacks expertise and expertise should therefore be given priority.

What should also be important is to focus research on areas where a lack of knowledge or solutions creates limitations in the industry today both globally and in Sweden.

Likewise, it may seem unnecessary to conduct in-depth research within a specific area if the improvements that are achieved will not lead to, for example, increased recovery due to the fact that the problems are elsewhere. It is also doubtful whether there are reasons to develop methods that already on the paper have lower profitability or recovery yield than what's the case for methods and processes abroad since this probably creates poor conditions for a sustainable supply chain.

State of the art in research vs the needs in the industry

In this research summary, the focus has been mainly on three areas:

- Generation and collection of spent lithium-ion batteries
- Reuse of lithium-ion Batteries
- Recycling of lithium-ion batteries

Furthermore, the study has also covered research on the environmental impact of batteries and design for recycling and reuse.

At the overall level, it can be stated that the research is very advanced with extensive research done within recycling, relatively much within reuse and extremely little in the collection and generation.

Faced with how the market actually looks today, or what is considered to be challenges in the industry, there is a relatively large imbalance. As was noted in the introductory chapter, the biggest obstacle to increased recycling is not a lack of technical solutions. Today, there are several recyclers with efficient processes through which batteries are recycled to new battery materials – completely in accordance with what is desirable in a circular economy. What is missing, however, especially in Europe, is batteries to recycle.

This is however connected. If recyclers in other parts of the world have more efficient processes and have also access to a more lucrative market, they will always be able to pay higher prices than recyclers in Sweden. Of that reason there is a significant risk that will be impossible for recyclers in Sweden to attract sufficiently large volumes for a profitable business.

If, on the other hand, material disappears into the reuse market, effective recycling processes do not play a significant role anyway, since the value of recycled material can generally not match the value of second life products. This has done so portable batteries have left the country but, with systems for second life in place, it could contribute to make industrial batteries remain in Sweden or at least in Europe.

A general conclusion should therefore be that research that in various ways can contribute to increased volumes and increased profitability, regardless of whether it is about increased revenues or reduced costs, is of importance as it can contribute to increased competitiveness for Swedish and European companies.

Suggestion for research within generation and collection of waste batteries

This overview has shown that very little research has been produced in this area. Although several research projects have contributed to valuable knowledge of, for example, waste batteries' value chain and what drives it, it is not possible to say that there is a knowledge base to build upon. What is also noteworthy is the total lack of technical solutions, eg for increased safety or increased automation, which could both streamline and reduce the costs in the area.

Areas where increased research could play a significant role are:

- Increased system understanding - Bring understanding about how the entire EOL market is connected and how it relates to the overall battery value chain. If there is no available knowledge of when and where batteries reach EOL, where they go and what happens to them it becomes extremely difficult to prioritise measures and direct further funds to research and development. Understanding the revenue and cost structure in the entire value chain as keeping track of the the development in competing countries is of the utmost importance.

- There is also a great need to know where the thresholds exist for batteries to flow between users and collectors as smoothly as possible. What would make people submit electronic waste faster, including batteries? What would cause electronics recyclers and recycling companies to sell batteries to Swedish operators instead of abroad?

- Incentives and creative collection methods - Best practice in the collection of batteries. Modelling and testing on a smaller scale can be interesting to better understand how batteries can reach recycling faster.

- Barriers to international and intra-European trade and transport of batteries - Although most rules regarding battery recycling are created at EU level, there are still differences, especially for how batteries can be transported. This creates a lot of extra work for recyclers and often result in that best solutions in some cases cannot be chosen. The research has a role to clarify and identify obstacles and to propose relevant changes.

- Safe transports - In addition to the lack of batteries, the greatest problem for the recyclers today is the challenges that come with the storage and transport of lithium and lithium-ion batteries. Despite this, very little research has been done, which means that both legislation and commercial solutions are designed on weak grounds. More knowledge is needed about what causes fires, how batteries should be stored and packaged and how damage can be minimized. Experimental research can also contribute to new solutions and commercial products.

- Safety and working environment in the handling of lithium-ion batteries - On the same theme, there is a great need for mapping risks in the handling of lithium-ion batteries in the plants. Both to avoid accidents but also to minimize unnecessary regulation if not needed. In a business where marginal revenues are low, there is no room for expensive processes, but rather it will mean that materials are exported instead.

Suggestion for research about reuse

As stated in the chapter on reuse, a basic knowledge about using batteries in second life applications is starting to be established. However, it is yet only in its infancy and needs to be expanded. From a national perspective, there are several reasons to both broaden and deepen the knowledge in this area. First, reuse is a higher step on the waste ladder and is thus an important part of achieving several environmental goals. Secondly, a reuse market is a potential job creator where handling is required of products that were not once manufactured in Sweden but which now can employ people locally. Third, reuse is an important way to keep batteries in Sweden and Europe, which ultimately benefits recycling and a Swedish battery and battery material industry.

However, this is only provided that reuse solutions work in the way we hope it may. Here, research has an important task in critically verifying the potential in both the short and long term. The paradox of reuse is that in order for it to work as well as possible, adaptations of the products must be made already when they are designed for their first application. Taking this kind of decision, without knowing what the actual potential is about in as much as 10 years, is a challenge.

Areas where research can play a role are:

Evaluation of large-scale energy storage systems (ESS) - Most research has so far been done on very small systems. There are examples where conclusions about ageing and degradation have even been drawn at cell level and in several cases from research on an individual pack. Since an ESS earns money for its owner, a full-scale MWh facility could fulfill several purposes for eg a university or institute. So far, the academic world has the opportunity to stay ahead of the industry. There is also access to batteries in the market, it’s just about how much you are prepared to pay for them or what offer you have to individual companies. Here universities can play an important role.

Testing and Grading of Batteries - There are obvious advantages having a neutral player such as an institute or university to develop standards or openly available methods for the testing and grading of batteries. This could have great significance also for the trading of the batteries and increase the use in different types of applications.

Complete circular solutions - Once again, universities and institutes have their own vehicle fleets or work in projects with companies or public organisations that have it. Providing batteries in electric vehicles with different sensors and logs that follow the batteries during use and then using them in their own reuse systems would contribute with very useful knowledge. Even batteries outside the electric car world, such as batteries from trucks, trains, robots, bicycles etc can be used for this. What could be improved in the package for them to work better in reuse solutions? Can you combine energy storage with charging and quick battery changes (swapping)? Research that both looks for best practice and creates new solutions would provide a lot of value.

Improved battery pack monitoring and control - Development and evaluation of battery control systems (BMS) as well as different types of sensors and remote connectivity can have great significance for the capacity and life of the batteries, as well as form the basis for different services and business models.

Remanufacturing - What are the requirements for remanufacturing of batteries where they are used for the same application as before? Tests that show how it works which can give valuable information on which requirements that are needed.

Overview and evaluation of owner and business models - One of the biggest challenges with reuse is once again to get hold of batteries. As a rule, they are owned by players with little opportunity or interest in utilising the battery at EOL, but increasingly it is becoming known that the battery has a value. With too many steps in the value chain the margins are getting lower and there is a risk to end up with value chain where all players are unprofitable. Therefore, it is important to study which owner and business models that best can enable profitable reuse solutions.

Suggestions for research on recycling

Recycling is the strongest area within EOL research. Especially this applies to different types of separation methods. It is our opinion that there can be no reason to completely go out and try out new methods that cannot very likely lead to significant improvements to the methods that are already available. The pace in this area is now very high, especially in Asia, and it should be an absolute requirement from funding bodies that new proposed research in recycling should already at the time of application be compared to what's state-of-the-art.

At the same time, there is a great need to increase competence in more advanced recycling methods and not least in the manufacturing of battery materials. Therefore, this type of project should be prioritised, but they should be prioritised for just that purpose and not under the impression that the project will result in something that does not already exist.

Another major need is found within the peripheral activities of recycling such as sorting and pre-treatment, which also ties in with the needs described under the collection section on safety and transport. Everything that prevents batteries from being handled in an efficient manner is important to eliminate and here research can be very important.

Concrete needs for research in recycling are:

State of the art in re-manufacturing of cathode and anode materials - Literature studies and comparative experiments that can verify and transfer knowledge from existing research in the world in direct recycling and re-fabrication of battery materials. It is with this ability (together with local demand) that batteries can ultimately be retained in Sweden, and also attracted to Sweden from other countries, as this ensures the highest possible profitability for recycling.

Holistic evaluation of processes - There are advantages and disadvantages to all types of processes, from costs and efficiency to environmental impact and effects on transport and work environment. The methods chosen also have consequences for how a material needs to be pre-treated and sorted and which end-product that can and will be produced. What we have not found are evaluations of methods from this more holistic perspective which can both point out the most effective way for the batteries but also provide explanations as to why batteries take other paths than what both companies and legislators had expected.

Pilot projects involving recycling and remanufacturing of battery materials - Projects on a larger scale, preferably involving several institutions in order to truly become good at the entire chain from the treatment of the batteries to the production of new materials.

Methods for direct recycling - Some cathode materials such as LFP and LMO have low material values and can only be treated profitably in very efficient processes when the lithium price is high. At the same time, it is possible to retain the cathode and anode material in its original form in order to once again use it in new batteries after thermal treatment. At the same time, this requires not only technology but also an effective value chain where large amounts of the same battery type can be collected and where a need for the produced materials can be verified.

Studies and solutions for disassembly of both pack and cells - This is where the biggest costs lie for recycling of electric vehicle batteries. Finding methods that make it more effective to separate both modules and cells is therefore extremely important as it is necessary to be able to use eg hydrometallurgical processes.

Identify needs and develop solutions for sorting, classification and labelling of batteries - For many processes, the batteries must first be sorted in order to secure a high recovery efficiency. At the same time, sorting is not always necessary as the problem can be solved at an earlier stage. Therefore there is a great potential to integrate research about sorting and classification with collection, transportation and what is put on the market in the first place.

Finally – Reflections on research about EOL of lithium-ion batteries

In this study, we have identified almost 1000 different articles, project descriptions and reports. We have found that the area is divided. First of all it is divided in regards to which sub area that gets most attention where research about different recovery processes are common in most major markets while issues like design for recycling, collection and transportation have been studied much less, despite the fact that in many cases these issues are the prerequisites for the material to reach recycling at all. For the same reason, today we have overcapacity in the recycling industry on virtually every market, including China, primarily because of the lack of solutions to efficiently collect batteries .

There is also a geographical division. Where China and South Korea, which today are the leaders in recycling of lithium-ion batteries, early developed methods for converting scrap into raw materials, while other countries have basically only scratched the surface.

That this has happened is one thing. But that this development is basically untold in the Western research community is remarkable. For example, we have found reviews made in both Europe and the United States where there is no mention of Chinese research in a single reference. Although almost every year, well-written, educational literature reviews in English were published in China, which tell a completely different story than what is conveyed in the West.

When this is combined with EU projects where large amounts of money go to let recycling companies basically do the same thing over and over, or research groups that may continue researching exactly what they have previously researched in the past, this is an example of how Europe is systematically being overtaken by Asia. It has already happened in battery production. 10 years ago, the battery industry was almost entirely concentrated in Japan and South Korea. Today, more than two-thirds of the industry is found in China. Access to waste batteries and recycling technology was an important part of this development.

In order to achieve a change, we believe that it is not enough only to focus on the right strategic research areas. It also requires a different way of doing research. We believe research projects cannot just fund research staff. Larger budgets are needed to invest in equipment and materials. If research is to be at the forefront and be relevant, it must also lead and not be dependent on companies which also many times are resource constrained and lack funds to invest. This means that research is basically only done on things that have already been done before which should rather be treated as training than actual research.

We also believe that more money needs to be spent on airline tickets. Not to go around Europe for project meetings, but to China, India or Kenya where we can study the rapid development of everything from recycling to the use of energy storage. We also believe that in a small country like Sweden, it is necessary to focus the resources. The expertise that exists within the area must be clearly gathered, either physically or virtually, and it must build solid bridges internationally to keep track of what is happening in the area, and to always remain up-to-date with where the best competence can be found.

Finally researchers must be better at both using and challenging previous research. In our review we have often found a lack of comparisons and discussions about previous studies. As a reader it's easy to get the impression that many research projects are done in parallel with no connection to other researchers' work, when it instead should be done in a serial process where new results are used either to verify or to further develop methods or technologies. The beauty with science is that in practice, by using the same methods a researcher previously has been using, the outcome should always remain the same. Hence, only if there is a need to alternate the result, or the process in itself, it is necessary to repeat the effort, and it's only when we compare the results from the alternated process, volume or other circumstances with previous studies we actually learn something. However, the same thing is not true for non-scientific data such as number of recycling processes, prices, volumes and collection rates. This data is changing every day and what is factual correct one year is not necessarily correct the year after. This is especially important in a rapidly growing market where both technology and the commercial playing field is changing fast. As we in several papers have seen proof of this happening, with data that many times is more than ten years old, we believe it is extremely important that researchers, as well as all users of research, are critically reviewing references to be able to ensure the factual quality of the work they are using.

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