Chapter 9 Raw Materials and Recycling of Lithium-Ion Batteries



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9.1 Introduction

Electric vehicles (EVs) powered by lithium-ion batteries (LIBs) have quickly emerged as the most popular replacement for petrol- and diesel-powered vehicles. In the next 5–10 years, the LIB market is set to grow exponentially due to a push toward EVs by both policymakers and vehicle manufacturers [25]. Such a push will inevitably lead to an increase in demand for raw materials, which is of particular concern for critical raw materials (CRMs) such as lithium and cobalt which are of

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high economic importance [25]. Moreover, with a life span in EV of only 8–10 years, the LIB waste stream will increase considerably [39]. This is particularly important considering that, by 2025, the UK's dynamic stockpile of spent LIBs could exceed 100,000 battery packs or 42,000 tons of LIB waste [39].

Due to the economic value of the materials contained within spent LIBs and the volume of waste predicted in the coming years, the most economical and environmentally friendly option is to reuse or to recycle them. This is even more important considering that 2022 has seen the first ever increase in LIB pack prices since records began in 2010 [24]. Such increases are primarily due to rising raw material and battery component prices and the increasing inflation.

The development of recycling processes in the last decade has led to a sharp increase in the purity of materials recycled which can reduce the reliance on raw materials and alleviate some of the pressure on the natural reserves of materials such as nickel and cobalt. The most advanced recycling processes are pyrometallurgical, hydrometallurgical, and direct recycling [37]. However, even the most advanced technologies have challenges with regard to recycling efficiencies, significant environmental impacts, and safety hazards [22]. The safety hazards extend from the battery's primary use through to their final disposal, with 48% of waste fires in the UK attributed to LIBs [4]. It is worth noting that, these fires will increase significantly if the sale of EVs increases as forecasted.

This chapter starts with a brief review and analysis of the value chain of LIBs, their supply risks associated with raw materials, as well as the global impacts of using these materials, in both their original and secondary usage. This is followed by a detailed description of the three existing recycling processes for LIBs and the material yield from each of these processes, as well as a discussion on the opportunities and problems that come with these recycling processes. We briefly discuss battery recycling legislation and describe some of the safety risks associated with the transportation, processing, and recycling of LIB. The primary risks of LIB fires and how to prevent the fires are highlighted. This chapter concludes by summarizing the key findings of this work. For more details on general circular economy considerations related with batteries, including reuse and second life, the reader is referred to Chap. 2 of this book.

9.2 Battery Contents

9.2.1 Battery Families and Their Cathode Chemistries

To understand the supply and safety risks associated with the materials used in LIBs, it is important to consider the various active cathode chemistries of the numerous LIBs currently available. LIBs currently on the market use a variety of lithium metal oxides as the cathode and graphite as the anode [29].

Most existing LIBs use aluminum for the mixed-metal oxide cathode and copper for the graphite anode, with the exception of lithium titanate (Li_4Ti_5 , LTO) which uses aluminum for both [23]. The cathode materials are typically abbreviated to three letters, which then become the descriptors of the battery itself. For example, lithium cobalt oxide ($LiCO_2$) becomes LCO, which was presented in 1991 as the first major commercially available LIB technology [50]. Due to the high-cobalt content, and soaring cobalt costs, LCO batteries have become very expensive to manufacture. Problems also lie in LCO's lack of thermal stability and quickly fading capacity. Despite this, they are heavily used in mobile devices [46].

The next LIB emerged in 1996 with a cathode made of lithium manganese oxide $(LiMn_2O_4, LMO)$ [23]. Replacing cobalt in the cathode with nickel and manganese does make LMO cheaper than LCO but has resulted in a lack of cycling stability at high temperatures [46].

Most commonly used in medium- and high-range electric vehicles (EVs), due to their high energy density and low power consumption [45], is the lithium nickel manganese cobalt battery ($LiNi_xMn_yCo_1 - x - yO_2$, NMC). The NMC battery is a so-called "family" as any combination of the three metals is possible, giving rise to a variety of cathode chemistries within one family. The four chemistries which are most common are NMC-111, NMC-532, NMC-622, and NMC-811, with the numbers referring to the ratio of nickel-manganese-cobalt in the active cathode material. First commercialized in 2004, the NMC battery family boasts very good specific power, life span, cost, safety, and specific energy [31].

Similar to NMC is the lithium nickel cobalt aluminum oxide cathode ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$, NCA). NCA also has a high specific energy, power, and life span, but it is more expensive than NMC [31].

The final example is the lithium iron phosphate battery (*LiFePO*₄, LFP), widely used in medium- and low-range EVs, which has sacrificed energy density for safety, improved environmental performance, and low production costs, coming from the lack of cobalt in the cathode [45]. The market share of LFP batteries has grown substantially in recent years, from 10% of the global EV market share in 2018 to approximately 40% in 2022 [9]. One reason for this is the popularity of LFP batteries in the Chinese market, with Tesla recently announcing they will use LFP batteries in their Model 3 for the Chinese market [13]. The drive towards the electrification of public transport has also played a part in this. This is due to factors such as the absence of cobalt in the cathode reducing the production costs, the use of phosphate to increase stability, enhanced electrode stability against overcharging, and a higher tolerance to heat. All of which has lead to LFPs being commonly used in buses [44].

9.2.2 Whole Battery Pack

Most of the focus from recyclers is extracting the valuable metals such as copper, nickel, and cobalt [40] contained within the active cathode material. Despite this, the active cathode material only makes up a maximum of 35% of a LIBs' relative weight, as seen in Fig. 9.1. Pouch cells can weigh between 75 and 225 g, depending on the battery cathode chemistry.



Fig. 9.1 Relative weight percentages of LIB. (Based on Sommerville [40])

Lithium-ion cells come in three principal shapes and sizes: cylindrical, pouch, and prismatic. All three "form factors" are employed in the larger applications of LIBs including EVs and battery energy storage systems (BESS). In an EV pack, the cells are arranged in series, parallel, or mixed configurations to form a module.

Each module will also have its own electrical and thermal control components [38]. The modules are then connected in series, parallel, or a combination of the two, to form a battery pack. The modules can be mechanically locked into place or welded or glued together, which is a considerable disadvantage in their manufacturing, as it makes them particularly difficult to disassemble. The packs themselves are housed in a plastic or metal container, also containing a whole pack battery and thermal management systems.

9.3 Battery Cathode Materials and the Associated Supply Risks

A LIB's active components are an anode and a cathode, separated by an organic electrolyte, i.e., a conductive salt (LiPF_6) dissolved in an organic solvent. The anode is typically graphitic carbon, but silicon has emerged in recent years as a replacement with a significantly higher specific capacity [51]. The inactive components include a polymer separator, copper and aluminum current collectors, as well as a metal or plastic casing.

Table 9.1 Typical raw material requirements (Li, Co, Ni and Mp) for three battery		Туре	Lithium	Cobalt	Nickel	Manganese	
	LCO		0.11	0.96	0.00	0.00	
cathodes in kg/kWh [20]	NCA		0.10	0.13	0.67	0.00	
<i>6</i> []	NMC	111	0.15	0.40	0.40	0.37	
		622	0.13	0.19	0.61	0.20	
		811	0.11	0.09	0.75	0.09	

The majority of materials that are constrained by resource limitations are those contained within the cathode, as well as the electrolyte due to its lithium content [35]. The majority of LIBs on the market today have cathodes which include lithium, cobalt, nickel, and manganese due to their high energy densities. Table 9.1 shows an estimate of the amount of these metals, in kilogram required per kilowatt-hour for five popular cathode materials.

Batteries with lithium cobalt oxide (LCO) cathodes typically require approximately 0.11 kg/kWh of lithium and 0.96 kg/kWh of cobalt (Table 9.1). Nickel cobalt aluminum (NCA) batteries, however, typically require significantly less cobalt, approximately only 0.13 kg/kWh, as they contain mostly nickel at approximately 0.67 kg/kWh. Nickel manganese cobalt (NMC) batteries vary on their raw material requirements depending on which member of the battery family is being used. For example, the NMC-111 contains approximately 0.40 kg/kWh of nickel, manganese, and cobalt, whereas NMC-811 requires 0.75 kg/kWh of nickel and only 0.19 and 0.20 kg/kWh of cobalt and manganese respectively. In practice, this means a Tesla Model S, which uses a 100kWh NCA battery [25], would require 10 kg of lithium, 13 kg of cobalt, and 67 kg of nickel.

The following section describes the supply chains associated with the elements used in the manufacturing of LIBs, particularly those contained in the cathode.

9.3.1 Cobalt, Lithium, and Nickel

It is projected that, just for EV batteries and energy storage, the EU will need 18 times more lithium and 5 times more cobalt in 2030, with this increasing another three-fold by 2050, compared to the current supply to the whole EU economy [1]. This will inevitably lead to supply issues not just in the EU, but globally, and has resulted in both materials being added to the EU's Critical Raw Materials (CRMs) list [14].

The significant increase in demand for cobalt, lithium, and nickel is demonstrated in Fig. 9.2, where it is easy to see that the adoption of LIBs with cathode chemistries with very low or even no cobalt is appealing. In both scenarios modeled in Xu et al. [49], the known global reserves for cobalt could be depleted by 2050. Cobalt reserves also have the added challenge of being very geographically concentrated, partly in areas with political and social conflict [49]. The Democratic Republic of the Congo (DRC) in particular plays a dominant role in current and future cobalt supply,



Fig. 9.2 Global demand for raw materials to 2040 [20]



Fig. 9.3 Cobalt price volatility from February 2010 to August 2023 [43]

accounting for 60–75% of global mine production [2]. Cobalt mining also has the disadvantage of being reliant on the nickel and copper markets, as cobalt is primarily mined as a by-product of the two [2300], so the expansion of new cobalt mines will only occur if the nickel and copper markets are strong [2].

Due to this, cobalt markets are volatile, rising from \$31,000 per ton in 2012 to \$93,000 per ton in 2018, with another peak in 2022 [2300, 28]. The volatility is well demonstrated in Fig. 9.3. This increase resulted in a 5–64% increase in cathode material costs per technology, proving the high dependence on raw materials in the industry [46]. Moreover, the supply risk score of cobalt has risen sharply from 49 in

2007, meaning the element was uncritical, up to 60 in 2017, making it the most critical element contained within battery cathodes [46].

Cobalt plays an important role within the battery chemistries, providing high energy densities and stable batteries, and so it is unlikely that cobalt will be eliminated from LIB cathodes in the near future [2]. This benefits the recycling industry as cobalt is the main driver of the revenue produced from pyrometallurgical and hydrometallurgical recycling. With appropriate recycling facilities and further development, the industry can move away from mined cobalt and begin to use recycled cobalt from spent LIBs.

Lithium has much the same supply issues as cobalt. By 2025, it is possible that lithium demand could outgrow current production capacities [49], with one of the scenarios in Xu et al. [49] concluding that known reserves of lithium could be depleted before 2050.

Battery manufacturers are attempting to decrease their reliance on cobalt, but not lithium; this is perhaps due to lithium reserves being less concentrated in conflict areas. Despite this, in the year 2012–2013, lithium's supply risk score jumped considerably, from 52 to 57 [46]. The issue with lithium is that the industry does not appear to be trying to relieve their reliance on it at this present time. This further solidifies how efficient, cost-effective recycling is necessary to recover lithium from spent LIBs and ensures the recovered lithium is of a high enough quality to be used in future LIB manufacturing.

Lithium and cobalt also have a variety of other uses, outside of LIBs. For example, cobalt is magnetic and so when alloyed with aluminum and nickel, it can be used to produce particularly powerful magnets. Cobalt's high-temperature strength also makes it particularly important in the development of jet turbine generators. More superficially, for centuries cobalt has been used to produce blue paint. While lithium is a very light metal, it is often alloyed with others to make lightwear armor plating, and aluminum-lithium alloys are used in aircraft and high-speed trains. Interestingly, lithium carbonate can be given to people suffering with severe depression as a mood stabilizer, but the full effect of the drug on the brain is not fully understood.

Although not as critical as lithium and cobalt, nickel reserves are still a concern, with the prediction that by 2040 EVs alone could require as much nickel as the global primary nickel production in 2019 [49]. As with lithium, one scenario in Xu et al. [49] predicts known reserves for nickel to be depleted by 2050. This is mostly corroborated by Wentker et al. [46] which predicts that with the current rates of extraction, nickel reserves will be depleted in 35 years. This is concerning when it is widely accepted that the adoption of battery chemistries with high-nickel and low-cobalt content has been faster than expected and could lead to a 60-times increase in nickel demand for the EU alone from 2017 to 2060 [2].

The industry's move from high lithium content batteries just shifts the burden onto nickel reserves. This is depicted well by the projected dramatic increase in nickel demand compared to cobalt demand displayed in Fig. 9.4. Although LIBs with high-nickel chemistries have a higher energy density and therefore reach the desired range for EVs, there is some concerns over the stability of these batteries



EU Cobalt and Nickel Demand to 2050

Fig. 9.4 Cobalt and nickel demand for European EVs for high adoption of high-Ni cathodes [2]

particularly the lithium nickel oxide battery which, after two decades of intensive research, still are not commercially ready [2]. However, the revenue generated from recovered nickel is much lower than that of cobalt which, due to the move toward high-nickel low-cobalt battery chemistries, may impact economic viability of recycling, as it will depend more on the volatile price of nickel [2].

9.3.2 Manganese

The supply of manganese comes from the mining of ore and scrap, with the ore including both manganese and iron ore [42]. The majority of mined manganese comes from South Africa and Australia, with shares of 26% and 17%, respectively, with China dominating processing and consumption [42]. Helbig et al. [23] find that the supply risk score for manganese, 52 points, is particularly average for all the raw materials used in LIBs. The most notable supply risk indicators for manganese come from the static reach reserves and the substitutability. The static reach reserves are only 34 years, the third lowest out of the raw materials, and the substitutability has a score of 4 which is second lowest [23]. This means that the current easy-to-access manganese reserves will be depleted in only 34 years and manganese as a component of LIBs has very limited materials that could replace it while maintaining the battery capacity and life span. Both of these factors mean it is imperative that the purity of recycled manganese is adequate to be reused in LIBs, taking away the reliance on reserves.

9.3.3 Other Materials

Copper, steel, aluminum, and graphite are also materials found in the spent LIBs. Xu et al. [49] predict that for copper, aluminum, and graphite, all known reserves exceed demand from EV manufacturing until at least 2050. However, there is a slight concern about natural graphite, as in 2019 64% of it was produced in China, which may lead to low supply reliability through political conflicts among global powers [49]. As much as this would not be ideal, it is true that synthetic graphite has begun to dominate the LIB anode market, with a 56% share in 2018, due to both its increased performance and decreased cost [49].

However, the shift toward silicon-based anodes, as appears to be the trend, would alleviate these concerns, with 25.8% abundance of silicon in the Earth's crust [26]. Silicon-based anodes also provide good chemical stability in the electrolyte, improving safety of the battery, and the abundance of silicon in the Earth's crust reduces the overall cost.

As much as these materials are necessary to the manufacturing, and therefore the recycling, of LIBs, their lack of criticality in comparison with the other materials makes them of low concern. However, to achieve increasing recycling efficiencies according to the new regulatory framework for batteries in, for example the EU, USA, and China, it is vital to recover these fractions, no matter how small.

9.4 Lithium-Ion Battery Recycling

9.4.1 Available Recycling Processes

Due to the value of the materials contained within LIBs, it is vital that they are safely and effectively recycled. All recycling is either open-loop or closed-loop. Open-loop recycling is the most common form, in which materials recovered from the recycling process have to undergo a series of refining processes before they can be used again [37]. Closed-loop recycling, considered the best case scenario, is when the materials recovered from the recycling process are in the correct chemical form and sufficient purity levels to be reused directly in the products they were recycled from [37].

There are a growing number of recycling facilities across the globe, as depicted in Fig. 9.5. Each company achieves different recycling yields, due to having their own unique take on one of the three available recycling processes or employing a combination of two.

Three main recycling processes for spent LIBs are commonly used: pyrometallurgical, hydrometallurgical, and direct cathode recycling (which will be referred to as direct recycling). Pyro- and hydrometallurgical processes are both employed to effectively recover metals from e-waste. The recoverable materials from each of these processes are listed in Table 9.2. None of the recycling processes listed are



Fig. 9.5 Locations of recycling facilities globally, with the size of the red dot representing the recycling capacity in tons/year

Pyrometallurgical	Hydrometallurgical	Direct	
(1) Copper compounds	(1) Copper	(1) Copper	
(2) Iron compounds	(2) Steel	(2) Steel	
(3) Co^{+2} in output	(3) Aluminium	(3) Aluminium	
(4) Ni ⁺² in output	(4) Graphite	(4) Graphite	
(5) Lithium compounds	(5) Plastics	(5) Plastics	
(6) Aggregate (from slag)	(6) Lithium carbonate	(6) NMC	
	(7) Co^{+2} in output	(7) Electrolyte solvents	
	(8) Ni ⁺² in output	(8) Electrolyte salts	
	(9) Mn ⁺² in output		
	(10) Electrolyte solvents		
	(11) Electrolyte salts		

 Table 9.2
 Recoverable materials through different recycling technologies

perfect, and work is being done to improve the processes in some way. Most of the improvements are based around increasing yield or purity, reducing the use of raw materials or energy, and reducing waste [18].

Pyrometallurgical Recycling Process

Pyrometallurgical recycling is one of the most ubiquitous metal recycling technologies used today. Pyrometallurgical processes use high temperatures to extract and purify raw materials. Fig. 9.6 depicts the process flow of a generic pyrometallurgical recycling process, in which spent LIBs, either shredded or intact, are sent to a smelter which burns off electrolyte and plastics in the battery to supply heat and the gas produced through the smelting process is treated.



Fig. 9.6 Process diagram of pyrometallurgical recycling processes

Graphite/carbon and aluminum in the LIBs act as reductants for the metals and are oxidized, while cobalt, nickel, copper, and iron in the LIBs make up the matte. The rest of the materials, including oxidized aluminum and lithium, end up in the slag. It is important to note that the slag may be used as aggregate for pavement or as supplementary material for cement production and there is ongoing research into the lithium recovery process from the slag [40].

The matte undergoes an acid leaching process and then precipitation to produce iron and copper compounds. Following this, the matte can be further processed to produce cobalt and nickel compounds; the processes used are solvent extraction followed by precipitation. These compounds can also be separated fully through hydrometallurgy.

The facilities which currently utilize pyrometallurgical recycling are Accurec and Umicore [40]. In Umicore's facility in Hoboken, Belgium, only modules or packs larger than a shoebox require disassembly prior to the recycling process [40]. See Table 9.2 for a list of recoverable materials through pyrometallurgical recycling.



Fig. 9.7 Left: calcination resp. drying of spent LIB and subsequent generic hydrometallurgical recycling process. Right: the hydrometallurgical process used by TES

Hydrometallurgical Recycling Process

Figure 9.7 depicts the process flow of a generic hydrometallurgical recycling process. Hydrometallurgy uses aqueous solutions, such as acids and salts, to dissolve the metals, and then subsequent steps recover the metals from the solution.

Spent LIBs which are hydrometallurgically recycled must first be discharged and disassembled, before they are shredded. This is important as, for hydrometallurgy to be cost-effective, it is necessary to ensure that minimal extraneous material is exposed to the process [40]. In some cases, organic compounds such as the binder and solvents from the electrolyte are then burned off and carbon dioxide will be emitted. TES, a global LIB recycling company, uses shredding under inert atmospheric conditions and vacuum drying followed by condensation for this, as shown in the right image of Fig. 9.7. The organic solvent from the electrolyte will be recovered, and residual fluorine and phosphorus will be removed at the purification step of the hydrometallurgical process.

After shredding, the process is made up of several physical separation processes to separate out aluminum, copper, and steel as metal scraps, plastics, and black mass followed by a leaching process for the black mass. The final step includes solvent extraction and precipitation to produce cobalt-nickel-manganese compounds, with the potential for lithium carbonate extraction which can be used in the production of new cathode materials [11]. However, market demand for high purity materials in the correct ratios justifies further separation into individual cobalt-nickel-manganese compounds. Both Duesenfeld in Germany and Recupyl in France use this process, however only at a small scale, selling most of their black mass to the metallurgical industry [40]. Umicore performs hydrometallurgy after pyrometallurgy to further separate the compounds of transition metals [40].



Fig. 9.8 Process diagram of a generic direct recycling process

Direct Recycling

Figure 9.8 depicts the process flow of a generic direct recycling process. Direct recycling describes the process by which the battery components are recycled without breaking down their chemical structure. For this, spent LIBs must first be discharged and disassembled before they can be perforated.

To recycle the electrolyte solvent and salts, they then undergo supercritical CO_2 extraction. The rest of the LIBs can then be shredded before going through several physical separation processes, density separation, and froth flotation, which recover plastics, metals, anode material, and cathode material, respectively [11]. The final step sees the recovered cathode material relithiated, which is the process by which they restore lithium stoichiometry of the cathode by bathing it in a heated lithium solution, to produce rejuvenated cathode powder.



Fig. 9.9 Generalized recycling loop. Processes are in purple and intermediate products in blue. (Based on Sommerville et al. [40])

Direct recycling is not currently used anywhere in industry, but it is the preferred method as the active material is reused without it having to be returned to the constituent raw materials as compounds or salts [40].

9.4.2 Yield for the Different Recycling Processes

As mentioned previously, the uniqueness of each company's recycling process leads to differing material recovery percentages and purities. This section will give statistics as produced in generalized processes modeled in Everbatt [11]. It cannot be overstated how essential high material recovery percentages and purities are to the alleviation of pressure on material reserves globally. Striving for closed-loop recycling across the industry should be an ultimate goal.

A generalized recycling loop showing the potential routes in which LIB cells may be recycled is shown in Fig. 9.9, with processes in purple and intermediate products in blue. In practice, some large-scale recyclers follow the loop to the left, using a combination of pyro- and hydrometallurgical processes. Not shown in Fig. 9.9 is that some recyclers produce only a "black mass" of active material, i.e., metal oxides and carbon, which will then be sold on for pyro- or hydrometallurgical recovery [40]. Both pyrometallurgical and hydrometallurgical processes recover 98% of the cobalt from the input, and, with such a high efficiency, there should be a consistent drive to ensure spent LIBs are recycled so the cobalt can be reused, particularly due to the high volatility of the cobalt markets and low natural reserves [11].

Material recovery of lithium is not as efficient as cobalt, at only 90%, and to recover lithium using pyrometallurgical recycling, the slag must undergo a hydrometallurgical process, thus increasing recycling costs making it less attractive to recyclers [11]. This means recyclers are less likely to recover lithium, increasing the reliance on virgin materials. Luckily, like cobalt, material recovery efficiency of nickel is 98% for both pyrometallurgical and hydrometallurgical recycling [11]. Only hydrometallurgical recycling recovers manganese, but this is also at 98% efficiency [11]. Copper and steel have 90% material recovery efficiencies from all three recycling processes. Meanwhile, aluminum and graphite have 90% material recovery efficiencies from only hydrometallurgical and direct recycling [11].

9.4.3 Opportunities from Recycling

The primary advantage of LIB closed-loop recycling is that it can save raw materials. When materials such as lithium, cobalt, and nickel are so critical to the operation of LIBs but are relatively scarce, it is vital to develop recycling processes which will alleviate some of the pressure on natural reserves. It is estimated that recycling can save up to 51% of the extracted raw materials, in addition to the reduction in the use of fossil fuels and nuclear energy in both the extraction and reduction processes [8].

One benefit of a LIB compared to a primary battery is that they can be repurposed and given a second life. A LIB in an EV is classed as EOL once the warranty, usually 8–10 years, has been exceeded; however, both manufacturers and developers agree that LIBs still retain 70–80% of their initial capacity after this time [39]. Some changes to the LIB may be necessary before it can be repurposed, such as replacing damaged cells or reconfiguring the pack for non-EV use [5], but it is estimated that LIBs which are repurposed in stationary energy storage applications have a second life span of an additional 10 years before they reach their absolute EOL [39]. Another benefit of second life LIBs is both environmentally and economically valuable, as they can reduce direct energy consumption from the electricity grid [39].

The idea of a second life for LIBs from small devices such as mobile phones has been trialed in the developing world, mostly in isolated areas that are not connected to the national grid. The second life LIBs can be connected to solar panels and allowed to charge and then used to power LED-based systems [8]. This is a safe, reliable, and sustainable way to light homes, aiding in the replacement of candles and kerosene lamps. Costa et al. [8] find that a LIB that was used in the standard life of a mobile phone, 2 years, still contains 1250 cycles. This means a 12 V, 3.1A battery will be able to power a 5 W LED lamp for 4 hours every night for 3 years and the unit itself will cost only \$35 whereas kerosene lamps cost an average of \$54 per year of use [8].

Pyrometallurgical recycling has been proven to be economically feasible and conducive for large-scale operations [30]. Hydrometallurgical recycling has a number of environmental benefits, such as a low operating temperature and lower CO_2 emissions compared to the pyrometallurgical process [5]. Economically, hydrometallurgical recycling is preferable due to the increase in recoverable materials and their increased quality. The hydrometallurgical process allows most of the metals contained within EOL batteries to be recovered after extraction and separation through using strong inorganic acids to leach the metals into solvents. Inorganic acids such as hydrochloric, sulfuric, and nitric can be used to achieve 99% solubilization of lithium and cobalt [30].

9.4.4 Limitations of Recycling

The primary limitation of LIB recycling is that closed-loop recycling does not exist; the materials cannot be used in like-for-like products and instead have to be down-cycled. This is due to the waste treatments, namely, shredding, involving some degree of material intermixing and small-scale dispersion of metals into those recovered in bulk, such as aluminum and steel, reducing their quality. Although recycling cobalt from LIBs can be used in samarium-cobalt magnets for sensors and electric motors [34], the majority of recycled materials go on to be used in products with lower material quality requirements. To compensate for this, primary and secondary materials are often mixed to match material requirements in other products.

The pyrometallurgical recycling process may have generated relatively successful business models up until now, but this is likely to change. Despite the fact that the process is simple and mature and requires no sorting prior to recycling, there are a number of disadvantages [5]. Pyrometallurgical recycling requires a significant amount of energy to treat waste gases before they are safe to release into the environment [47]. This is in addition to the process itself requiring very high temperatures, making it very energy intensive [30]. Moreover, metals such as lithium, aluminum, and manganese cannot be recovered at such high temperatures, and the growing trend toward manufacturing LIBs with lower cobalt content means the revenue generated from this process will decrease. This is due to recovered cobalt generating 50–70% revenue for pyrometallurgical recycling, so lower cobalt leads to lower revenue and a lack of profit. Unfortunately, recent trends suggest that batteries with higher nickel and lower cobalt content, such as the NMC-811 batteries, are becoming more popular due to increased energy density, environmental sustainability, and reduced manufacturing cost [48].

Another disadvantage of the pyrometallurgical process is that both lithium and aluminum are entrained in the process slag and therefore require further processes to recover. The UK currently exports most of its spent LIBs to Umicore in Belgium, which uses the pyrometallurgical process; this meant lithium and aluminum were lost to the slag as it was not economically feasible to process it [47]. However, Umicore have recently employed further processing to recover lithium from the slag [5]. Although this is a positive in terms of material recovery, an additional process increases not just the total recycling cost but the environmental one too.

As with pyrometallurgical recycling, hydrometallurgical recycling does rely on cobalt recovery for the majority, in this case 40–60%, of its generated revenue. As above, the low nickel, high-cobalt content means it is not economically feasible to hydrometallurgically recycle NMC-811 batteries. However, unlike pyrometallurgical recycling, a profit is still generated per kg of NMC-532 and NMC-622 batteries when hydrometallurgically recycled. In addition to this advantage, the materials produced from hydrometallurgical processes are of a high purity while recovering most of the LIB constituents [5].

Unlike pyrometallurgical, the hydrometallurgical process does require manual deep-discharging and dismantling before recycling can begin, which requires considerable storage space, which adds to overall costs, due to an increase in labor costs, and overall complexity [5].

Hydrometallurgical recycling also produces a considerable amount of wastewater through the leaching and precipitation operations. The treatment this requires adds to both the environmental burden and overall recycling cost. Another increase in cost comes from the challenge of separating some elements in the solution, such as cobalt, nickel, manganese, iron, copper, and aluminum, due to their similar properties [5], more specifically cobalt and nickel in aqueous medium. This requires solvent extraction and consequently large amount of organic solvent with organophosphate additives to be stored and processed. Purification steps generate, e.g., $Al(OH)_3$, FeO.OH, CaF_2 , $CaPO_4$, which potentially need to be treated as hazardous waste.

Direct recycling is the newest of the modeled recycling processes and is still in its development stages for EV LIB recycling [25], which is its primary disadvantage. Another disadvantage of direct recycling is that it requires rigorous sorting before the recycling process can begin, as the exact cathode chemistry must be known prior to recycling. This is due to the inflexibility of the process: what goes in must come out [5]. This raises questions of whether the process is appropriate for an ever-changing reality; with a market already saturated by differing cathode chemistries, how can a process which can only recycle one specific chemistry at a time be sustainable? Although this is an important question, it is true that direct recycling produces the highest revenues of all the recycling processes and, due to this, it has the highest net recycling profit by a considerable margin [25]. The overarching economic benefits suggest that research into direct recycling must continue, ensuring it can be used sustainably on a commercial scale.

9.4.5 Battery Recycling Legislation

With battery recycling comes battery recycling legislation. Current legislation across all three major markets, China, the EU, and the USA, focuses on the protection of local environment and human health [32].

While the EU's original battery directive was published in 2006, the new Battery Regulation, published in 2023, will build upon and replace the 2006 EU Battery Directive, with a direct focus on the challenges that have come with rapid development of the industry [16]. Melin et al. [32] divide the new Regulation into four key elements, all of which are imperative to improving the sustainability of LIBs: The first is the Regulation aims to increase both transparency and traceability across the battery life cycle; second, it mandates carbon footprint declaration throughout the life cycle and establishing maximum thresholds, addressing climate impact of the batteries; third is an emphasis on circularity through increased collection and recycling efficiencies and mandating the use of recycled materials, particularly in batteries above 2kWh; and finally, fourth are including waste processors to the Battery Management System (BMS), verifying the battery's state of health in real time, and determining if the battery has the potential to be reused or repurposed before it is recycled.

While the EU has a number of directives to support in research and innovation across the entire battery chain, it has failed to secure key elements of the supply chain, such as raw material extraction, refining, and battery manufacturing [32]. Much is the same in the USA who, through Tesla, have been at the forefront of manufacturing but rely on global markets for refinement, production, and recycling of battery raw materials [2300].

The market is dominated by China, who occupy more than two-thirds of it [32]. This has been possible through a booming EV battery sector and strong government support in the form of subsidies and investment stimulus [32]. China also implemented the Interim Measures for the Administration of the Recycling and Utilization of Power Batteries for New Energy Vehicles from 2018 [32]. These cover minimum standards for the reclassification of batteries for second life applications, the recycling efficiency of plants treating EOL batteries, and requirements necessary to qualify for subsidies. When these measures were tightened further in 2019, they were made stricter than the regulations the EU plan to enforce a decade from now [16].

While in the USA, the Biden administration has declared the electrification of transport a top priority and produced an investment proposal of up to \$174 billion, they still lag behind both the EU and China in mandating EPR or promoting circular economy principles. The stringent measures imposed on Chinese companies, who already dominate in material refining, battery production, and mature recycling infrastructure, have allowed them to easily comply with EU regulations. This imbalance between new and mature markets has led to a variety of unintended consequences. For example, such stringent measures can lead to distorted innovation through increased compliance costs, hindering competitiveness. Moreover,

European EV manufacturers having to adhere to the new, stricter regulations will be more constrained in their options than the less regulated USA and also risk losing out to Chinese competitors due to their bigger, more mature share of the market [32].

EU regulation does aim to responsibly develop supply chains for Europe's EV industry, with recycling being at the forefront of this. However, the regulations do not require recycled material to be sourced in Europe, nor does it restrict the source of recyclables to EOL batteries [16]. This will put companies who have operated in markets such as China and South Korea, with much greater experience in battery material production, including in the use of recycled materials, in a much better position to meet these regulations. This could lead to European material producers and battery manufacturers being essentially eliminated from their own market.

One new regulation being proposed by the industry is a digital battery passport (DBP) for each battery entering the market. Digital product passports themselves are not novel; they have existed for some time to assist value chain stakeholders in achieving sustainable product management [36]. However, the concept has recently caught the attention of battery policymakers, and the European Commission is implementing DBPs [15]. The policy explicitly calls for the implementation of DBPs for industrial and EV batteries by January 2026 [16]. The EU has implemented three main EOL battery polices: maximum carbon footprint thresholds, minimum shares of recoverable materials, and DBPs. The main goal of DBPs is to enable sustainable product life cycle management and promote value-retaining processes, which in turn facilitates sustainable and circular value chains [3]. However, due to DBP development currently being pursued by nongovernmental institutions, there is a lack of clear specifications for the scope of DBPs, leading to inconsistency across the industry.

Battery legislation covers the entire life cycle of a LIB, from manufacture to initial use through to collection, processing, recycling, and disposal. The life cycle itself comes with a number of impacts, covered under environmental, social, and economic.

The steps of a LIB's life have been covered throughout this chapter; however, one important element which runs throughout the life cycle of a LIB that has not yet been addressed is the safety risks that come together with LIB's use and disposal and the social impacts these have. The way these risks fit into the life cycle of a LIB is demonstrated with the flowchart in Fig. 9.10. In the next section, we will discuss these risks, as well as addressing how it is best to reduce them.

9.5 Current Safety Concerns of End-of-Life Batteries

9.5.1 End-of-Life Management and Recycling

At present, there are very few LIB recycling facilities in the UK and Europe, despite the crucial role the industry has in the decarbonization of the planet. However, there are almost daily reports of fires caused by LIBs in both bin lorries and recycling



Fig. 9.10 Flowchart of the life cycle of a LIB

facilities [33]. In the USA, the cost is even greater, and not just financially, they have seen injuries and even deaths. This had led to material recovery facilities in the USA being increasingly reluctant to admit LIB related fires due to insurance concerns.

A report from Eunomia found that there are approximately 201 waste fires each year caused directly by LIBs [4]. While the fires do vary significantly in terms of severity and duration, it is estimated that the fires cost the UK £158 million annually [4]. The majority of these costs are found to be incurred by waste site operators; however, the cost to society, the environment, and the fire service is approximated to be £16 million. The report also breaks the waste fires into four categories, with 1 being the most severe and 4 being the least, finding that it is fires of severity level 3 which occur most frequently and therefore have the highest cost implications at over £128 million annually [4].

In the USA, the situation is even more bleak with a Fire Rover report estimating the cost of LIB waste fires to be \$1.2 billion annually [17]. Even with the report's author believing fires are underreported, there is still an alarming year-on-year increase in waste fires. Although they have not all been directly linked to LIBs, LIBs are listed as 1 of the 4 causes of the increasing number of facility fires which have resulted in 49 injuries and 2 deaths, an increase in injuries of 158% from Fire Rover's 2018 report [17].

At present, these fires are caused by small LIBs from or in small rechargeable devices such as mobile phones. However, the battery industry is ever-changing and, most importantly, ever-growing, with an onslaught of EOL LIBs from EVs and second life applications coming very soon. This will diversify the waste stream recycling facilities have to manage, and it may be that the global drive to replace fossil fuels could mean the fiery epidemic in recycling facilities is the calm before the storm [33].

LIBs can store large amounts of energy, but this energy must be released in a safe, controlled fashion. If this energy is released through abuse of the batteries, toxic, flammable, and potentially explosive gases, as well as fine particles of heavy metals, are released; this process is known as thermal runaway. This can easily be caused by crushing and penetration of the batteries, which are common practices in recycling facilities. When the batteries are crushed, the separator may be pierced, which in turn allows the anode and cathode to come into contact and a short circuit to develop. When this occurs, the heat generated causes pyrolysis of the organic solvent and other exothermic reactions which generate a mixture of gases that includes hydrogen (up to 50%), carbon dioxide, carbon monoxide, small organic molecules such as ethane and ethene, hydrofluoric and hydrochloric acid, and hydrogen cyanide. When these vent from the cells, via safety caps on cylindrical and prismatic cells, or pouch cells bursting, the gases take with them small droplets of the remaining organic solvent producing a thick, white vapor cloud [6]. Thermal runaway produces very large volumes of vapor cloud, e.g., up to c.a. 6000 L/kWh [21], and hence when this is vented, it does so at high pressure: immediate ignition of the cloud thus results in flares that can be several meters long. Delayed ignition can, and has, resulted in unconfined vapor cloud explosions (UVCE) [10], and such large volumes of vapor cloud have also led to UVCEs from small LIBs including e-scooters and e-bikes.

One common practice is the storage of road traffic collision (RTC) vehicles in vertical piles and moving them with magnetic claws and forklifts. This cannot be the case for EVs due to the risks posed by potentially damaged battery packs, which have been known to ignite hours, days, or even weeks after the initial incident [41]. Thus, it has been generally accepted that RTC EVs should be stored with a 10–20 m exclusion zone.

Although the biggest concern does lie with damaged or abused LIBs, it has been known for EVs to apparently spontaneously ignite, and, while the cause is still unclear, BMS failure, defects in design and/or manufacture, or some contamination during manufacturing are the most common postulations [7].

The lack of understanding of the risks and hazards of LIBs is concerning; while there seems to be some in governments, the public appears to be wholly unaware: an example is the incidents of fires due to members of the public assembling spare e-bike batteries bought online and doing so by soldering. Companies are currently able to sell secondhand EV battery packs, even batteries that are damaged or missing their BMS, to the public, with no safety warning. While this may not be illegal at present, it is wildly irresponsible. Without the appreciation for the complexity of the protection systems or the hazards of abused batteries, the risk of personal and municipal harm cannot be understated [7]. It is evident that abuse of batteries, such as rapid charging, does destabilize them and can reduce the onset of thermal runaway to room temperature [27], and damaged EV batteries have an increased risk of explosion. LIBs are safe and stable to use under specified limits of temperature and charge, but where does liability lie when misuse leads to injury or death?

9.5.2 Reducing Waste Fires

The most obvious way to reduce waste fires caused by LIBs is to ensure the batteries do not end up in the residual and mixed recycling waste streams in the first place. There are two easy to implement, short term measures which may help with this. Firstly, separate kerbside battery and small waste electronic and electrical equipment (WEEE) collection from households, and, secondly, increase the number of retail collection points for batteries and small WEEE [4].

However, the responsibility cannot be placed only on the consumer; instead, there must be supporting policy mechanisms that would fund systemic change in the collection of spent LIBs. The most effective way to do this is to financially incentivize or deter consumers from incorrectly disposing of batteries, alongside an improved collection system. These policies may include the banning of batteries from residual and mixed recycling waste streams, fining those who do not comply; enforcing enhanced extended producer responsibility (EPR) for batteries and small WEEE to pay for and coordinating, improving collection, and reflecting the cost of fires; creating a deposit return scheme (DRS), or other incentivization method, to encourage consumers to return spent LIBs and small WEEE for recycling; and introducing fee modulation within WEEE EPR system that allows producers to pay lower fees for design features that facilitate easier battery removal by consumers [4].

Moreover, there is evidently a problem in the WEEE management stream, with the increase in fires being an indicator of the need for change. The GRINNER project aims to rectify this. Funded through the European Union's (EU) Horizon Europe program, the GRINNER project will focus on the reduction of fires caused by LIBs in the WEEE management chain [12]. The plan is to develop an AI-powered battery detection system utilizing data from X-ray detectors and pick-and-place robots. The system will use X-ray detectors to analyze X-ray data and detect waste containing batteries that can then be removed by a vision-based pick-and-place robot [12]. The robots will be commercially available and easily incorporated into existing WEEE and other recycling environments, to extract batteries and other e-waste safely and effectively before they are damaged by machines that crush and consolidate waste.

Batteries themselves do contain several methods to improve their thermal stability. For example, each battery contains a BMS which controls and prevents conditions which could lead to failure, such as overcharging and overdischarging [19]. The BMS also operates the battery for the best application performance and a long life span. In addition to BMS, all batteries contain a thermal management system (TMS) which maintains the optimum operating temperature of between 20 and 40 °C and keeps temperature changes between the modules to a minimum [19]. As a whole, the battery compartment is also built with fire prevention in mind, being designed to survive structurally and containing a vent to let out any pressure buildup [19].

9.6 Conclusions

It is evident that the EV revolution is well underway, with a vast number of raw material requirements and battery waste being created.

With such a diverse product market, there is a great amount of choice for battery manufacturers, from the low cost LFP batteries to the high capacity NCA batteries. The diversity of this market does not come without its problems, with many of the materials used to produce the battery cathodes coming with considerable material criticality issues, particularly lithium and cobalt. While the market does appear to be moving away from batteries with a high-cobalt content, the use of lithium is here to stay, due to its stabilizing properties. Moreover, this move away from cobalt has led to the development of batteries with a higher nickel content, simply shifting the burden onto nickel reserves.

Made up from a variety of toxic and rare materials, stockpiling and landfilling spent LIBs is not an option, so the waste streams generated from them need sustainable, cost-effective, high-yield management systems, such as recycling. The recycling market has already undergone change: pyrometallurgical recycling began as the most popular recycling method; however, there is no lithium recovery under this method. This has led to the growing popularity of hydrometallurgical recycling which produces high material yields, for low environmental cost. As direct recycling is still in its relative infancy, it is not yet available for commercial use; however, this method of recycling is the industry's best chance at achieving closed-loop recycling.

There are some very clear advantages of recycling LIBs, with the main one being the alleviation of pressure on reserves of the raw materials. It is important, however, to consider second life applications of LIBs that have reached their EOL in their first application. For example, LIBs in EVs reach their EOL in an EV after only 8–10 years, but they retain 70–80% capacity. These batteries can go on to have a second life in stationary energy storage, storing renewable energies such as solar to be released in times of high demand, alleviating pressure on the national grid. Smaller LIBs from small electronic devices can also be used in such a way to power individual electronic devices.

As closed-loop recycling, the method by which recycled products are used in likefor-like products, has not yet been achieved, the recycled products must be downcycled. Mostly, the recovered materials are mixed with other raw materials to contain the correct mix to be used in products with lower material quality requirements. This is only one of the limitations of LIB recycling; the others include the hazardous waste produced from the recycling processes and the fact that LIB recycling is still not economical, with government grants needed to fill the gap.

Safety in recycling plants has already proven to be a real concern, and that is without the addition of a very large number of very large EV batteries entering the waste stream. To date, fires caused by LIBs have cost the UK and US economy billions and will continue to do so without sufficient policy intervention which prevents spent LIBs from finding their way into conventional waste streams. Due to the high chance of thermal runaway, spent LIBs cannot be handled in the way

lead-acid batteries are; long gone are the days of scrap vehicles being stored in piles 20 meters high.

This chapter has presented the cathode chemistries and the supply risks that come with the most important raw materials for each cathode. There is a discussion to move away from high-cobalt chemistries, the impacts on nickel reserves, as well as the low substitutability of other materials such as manganese. The different recycling processes, with a simple explanation of each, accompanied by a flow diagram were presented. The yield of each process was discussed followed by the opportunities and problems we are presented with when recycling LIBs, namely, the environmental benefits and down-cycling of products. This chapter covers battery recycling legislation, including DBPs and how they aim to aid in the adoption of a circular economy. We finish with an overview of the safety issues which come throughout the life cycle of a LIB, with a particular focus on how the LIB waste stream is incredibly high risk.

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