

**Challenges and recent developments in supply and value chains of electric vehicle batteries:
A sustainability perspective**

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1. Lithium-ion batteries and electric vehicle revolution

Lithium-ion batteries (LIBs) were initially used as a power component in different types of portable electronic devices, from mobile phones, power banks, tablets and laptop computers to cameras and camcorders. Nowadays, LIBs are incorporated into ever widening application areas (e.g. electro mobility, grid energy storage, etc.) due to some of their unique advantages such as high energy density, high reliability, reduced memory effect, long service life, etc. (Hua et al., 2020, Mossali et al., 2020, Paul A. Christensen, 2021). These had led to an extraordinary increase in the capacity and mass of batteries placed on the market within the last decade. Accordingly, the uptake of LIBs increased more than 7 times, from 29.6 GWh in 2010 to more than 217 GWh in 2019 (Fig.1) (Melin, 2021, Melin et al., 2021). The total capacity of LIBs that will be placed on the market is expected to reach more than 2500 GWh by the end of 2030 (Melin, 2021).

LIBs started to be used in electric and hybrid vehicle market from 2010, reducing the share of nickel metal hydride in the market (Melin, 2018). The records also show (Fig. 1a) that LIB application in EVs surpassed the others and dominated the LIB market with 51% of the market share. The application of LIBs in light and heavy duty continues to growth in the current decade and expected to reach a remarkable share as of 77% in 2030 (Melin, 2021). According to the records, a large proportion of LIBs were placed on the Chinese market in the last decade (with the share of 49% in 2019), followed by Europe and the US (Fig.1b), which show the main LIB markets.

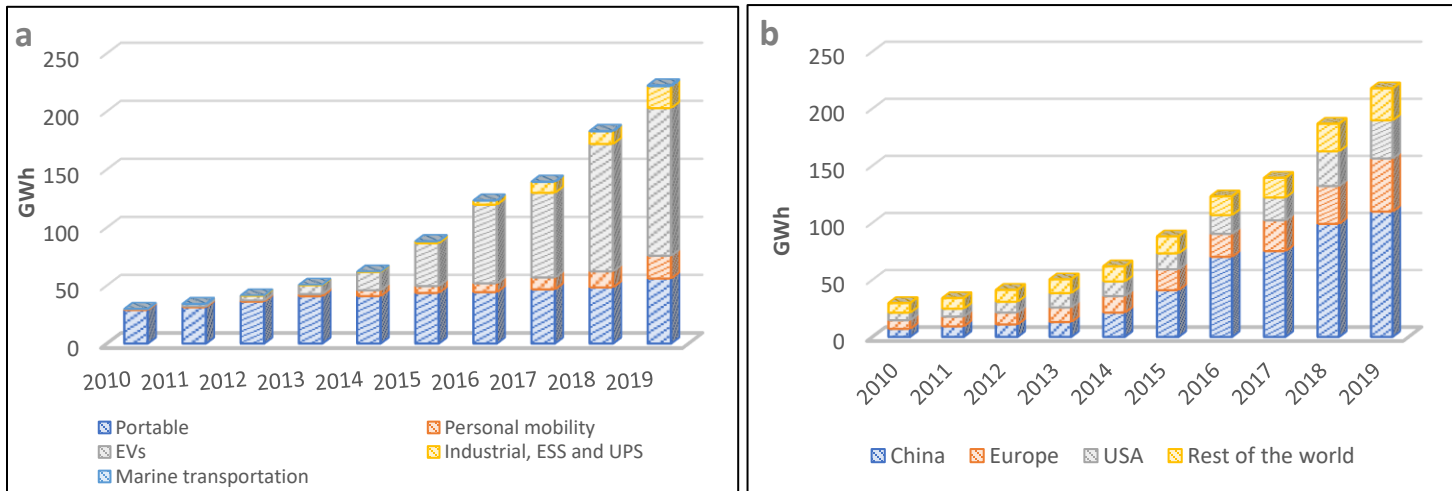


Figure 1. LIBs placed on the market between 2010-2019, a) based on different applications, and b) based on market geographical segments (Melin et al., 2021)

With the ever-increasing awareness over the adverse impacts of global warming, industries such as transportation have been considered as the main concern of various carbon reduction strategies and targets. The main impetus behind the electrification of transport is decarbonization of this sector (Crabtree, 2019) and is now considered as one of the main elements of governments plans around the globe to meet net zero targets. The advantages of LIBs as a power component for vehicles as well as higher efficiency of EVs compared to internal combustion engines is creating a revolution in the sector (Harper et al., 2019b, Karmaker et al., 2018, Carvalho et al., 2015).

During the last decade, the EV stock has drastically increased from a few thousands to 11.3 million EVs (Fig. 2a). According to the projections by International Energy Agency (IEA, 2021), the recent huge increase in EV stock is still nothing compared to what awaits, which shows an electric revolution is happening (Fig. 2b). This means that there would be probably more than 142 million EVs if existing government polices met; or there might be more than 227 million EVs if fully compatible scenarios with the climate goals of the Paris Agreement (so called ‘Sustainable Development Scenario’) are followed.

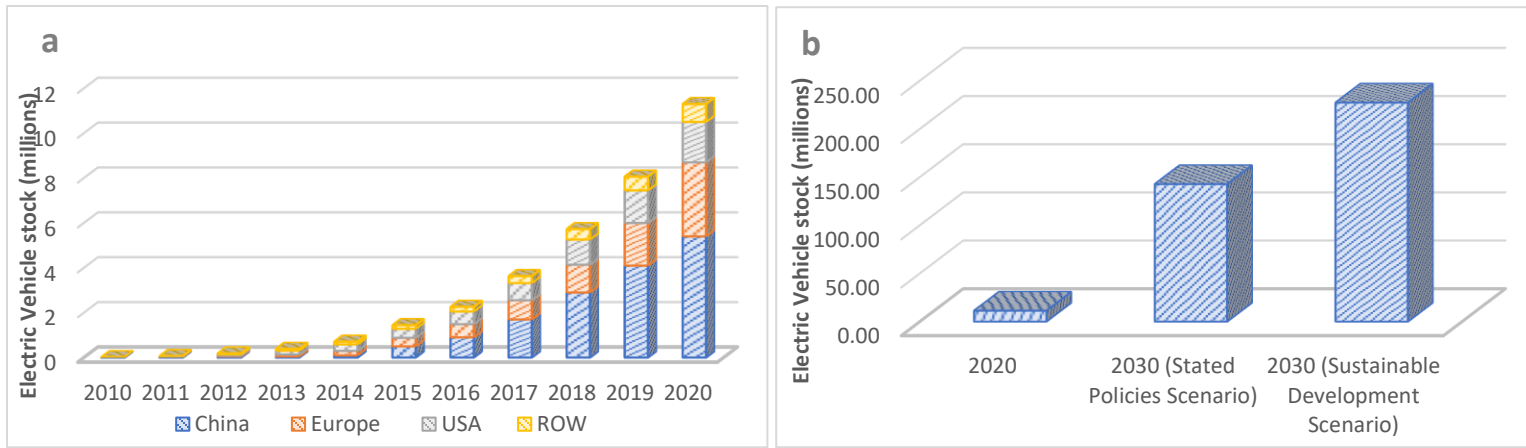


Figure 2. EV stock* a) between 2010-2020, and b) in 2030 projected by International Energy Agency (IEA, 2021).

ROW includes Australia, Brazil, Canada, Chile, India, Japan, Korea, Malaysia, Mexico, New Zealand, South Africa and Thailand; **Europe includes:** EU27+ Iceland, Norway, Switzerland, and United Kingdom.

*including passenger light-duty vehicles, light-commercial vehicles, busses and trucks; and it considers battery electric and plug-in hybrid electric vehicles (not fuel cell electric vehicles).

Despite the fact that LIB market will continue to grow in at least the same pace over this decade, there are some questions about the sustainability of their supply and value chains. These have further raised arguments for the reliability of LIBs as the leading technology in the long-term. In fact, the prevalence of LIBs and the benefits they present could have some unintended sustainability challenges, which need further investigation. Thus, the goal of this special issue was to address some challenges for the sustainable supply and values chains of EV batteries and call for additional investigation into the challenges and opportunities created by expected growth of EV adoption and end-of-life EV batteries. The special issue focused on the following, but was not limited to (Rajaeifar et al., 2020):

- Environmental, economic, or social impacts of EV batteries over their entire life cycle
- Resources and energy flows in the supply and value chains of EV batteries
- Dynamic modelling of the material supply chain of EV batteries
- Novel theories and solutions for end-of-life EV batteries such as collection, sorting, reverse logistics, and recovery
- Sustainability assessment of second life EV batteries for different applications
- Novel recycling techniques for end-of-life EV batteries
- Innovative management practices, business models, or policy frameworks for second life and recycling of end-of-life EV batteries
- Regulatory framework and legislation for managing end-of-life EV batteries

The next chapters focus on the supply and value chain challenges for electric vehicle batteries as well as the development made by this special issue.

2. LIBs supply and value chain challenges

2.1. Supply and demand of the battery materials

Many advanced and emerging technologies depends on a particular set of materials (or they so called ‘materials dependant’) and LIBs are not exemption. The production of LIBs involves the supply chain of various elements (Fig.3), such as nickel, cobalt, aluminium, magnesium and copper, etc. but in different severity (Fig.4 and 5). According to the list of critical raw materials (CRMs) for the EU, lithium, cobalt, magnesium, and natural graphite are among the critical metals and all used in LIBs (European Commission, 2021). A similar list published by U.S. Department of the Interior also includes the same elements (Gaines et al., 2018).

Among the mentioned elements, there are concerns over the criticality of lithium, cobalt, and graphite but from different perspectives. In fact, the high supply risk for these materials could lead to supply shortages and price volatility (Mayyas et al., 2019). In case of lithium, concerns over its scarcity have found less significant compared to cobalt (Gaines et al., 2018, Narins, 2017), however, the challenge is whether the mining industry would be able to build up lithium production in short-term to supply the lithium required for the widespread adoption of LIBs in EVs (Olivetti et al., 2017) and grid energy storage (Kamran et al., 2021). There is an ongoing discussion in the literature on whether and under which time frame, lithium availability is likely to demonstrate a serious impediment to the EV adoption (Kamran et al., 2021).

On the other hand, producing one ton of lithium leads to the depletion of huge volume of minerals which requires excessive amount of energy and water, and contributes to considerable environmental impacts (Meshram et al., 2014, Katwala, 2018). The huge amount of water required for lithium extraction affects the farmers in some regions and could create competition between ‘water for food’ and ‘water for mineral extraction’ (and subsequently contradicts with climate change mitigation strategies), which could finally lead to water import in major producer countries (Harper et al., 2019a).

Cobalt has been the centre of criticality concerns which the concern over its critically is mainly around the political instability of its main producer rather than economics (Olivetti et al., 2017). Most of cobalt mining happens in DRC (around 70% of global cobalt production (USGS, 2020)), a country with some political unrest records which could affect the cobalt supply and its price (Mayyas et al., 2019). Figure 6 shows the cobalt production by different countries around the world.

Moreover, there are some complex social issues with copper-cobalt mining in DRC such as child labour at some small-scale mines (Faber et al., 2017). Compared to Cobalt production from copper, which is mainly located in DRC, the geological concentration of cobalt from nickel mines is not of a concern. However, the production of cobalt as a by-product of nickel depends on the demand for nickel (Olivetti et al., 2017).

Another challenge in the supply chain of cobalt is that more than half of the cobalt and copper mining companies in DRC-that produce half of the country’s output as well- are now Chinese-owned (Farchy and Warren, 2018). These companies will give priority in cobalt supply to Chinese LIB manufacturers, if any cobalt supply interruption occurs (Mayyas et al., 2019). Moreover, another issue in the supply chain of cobalt is the concentration of cobalt refining in China which made it the biggest supplier of the refined cobalt (Farchy and Warren, 2018).

It should be noted that concerns over the natural graphite supply is also mainly around the concentration of both production and reserves in China (Mayyas et al., 2019). These seem to be only short-term concerns

due to the abundance of graphite around the globe and recent exploration and development in some other countries (Olivetti et al., 2017).

Overall, it should be highlighted that differences in the assumed concentration of minerals, reserve estimates, and the uncertain nature of future extraction projects led to variability in the supply estimation of the required materials for batteries (Olivetti et al., 2017). Another supply chain challenge is the uncertainty in the battery material demand estimations. In fact, estimation of the quantity of material that will be required for the emerging applications of LIBs is complicated and uncertain. The primary market for LIBs was consumer electronics, while EVs, personal mobility and stationary energy storage are the emerging markets for LIBs that are increasingly demanding more batteries (Fig.1). Among these markets, the consumer electronics is mature and therefore the estimations could be made with relatively higher reliability. In contrast, the demand for EVs, personal mobility and stationary energy storage are highly speculative. More specifically, battery demand projections for such markets differ significantly due to variability in the considered battery chemistry, battery material intensity, EV use and penetration estimations, and other battery application assumptions (Olivetti et al., 2017).

1																	18	
H 1.01																	He 4.00	
2												13	14	15	16	17	18	
Li 6.94	Be 9.01												B 10.81	C 12.01	N 14.01	O 16.00	F 19.00	Ne 20.18
11	12												13	14	15	16	17	18
Na 22.99	Mg 24.31												Al 26.98	Si 28.09	P 30.97	S 32.07	Cl 35.45	Ar 39.95
19	20	3	4	5	6	7	8	9	10	11	12	30	31	32	33	34	35	36
K 39.10	Ca 40.08	Sc 44.96	Ti 47.87	V 50.94	Cr 51.99	Mn 54.94	Fe 55.85	Co 58.93	Ni 58.69	Cu 63.55	Zn 65.38	Ga 69.72	Ge 72.63	As 74.92	Se 78.97	Br 79.90	Kr 83.80	
37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	
Rb 85.47	Sr 87.62	Y 88.91	Zr 91.22	Nb 92.91	Mo 95.95	Tc 98.91	Ru 101.07	Rh 102.91	Pd 106.42	Ag 107.87	Cd 112.41	In 114.82	Sn 118.71	Sb 121.76	Te 127.6	I 126.90	Xe 131.29	
55	56	57-71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	
Cs 132.91	Ba 137.33		Hf 178.49	Ta 180.95	W 183.84	Re 186.21	Os 190.23	Ir 192.22	Pt 195.09	Au 196.97	Hg 200.59	Tl 204.38	Pb 207.2	Bi 208.98	Po [208.98]	At 209.99	Rn 222.02	
87	88	89-103	104	105	106	107	108	109	110	111	112	113	114	115	116	117	118	
Fr 223.02	Ra 226.03		Rf [261]	Db [262]	Sg [266]	Bh [264]	Hs [269]	Mt [278]	Ds [281]	Rg [280]	Cn [285]	Nh [286]	Fl [289]	Mc [289]	Lv [293]	Ts [294]	Og [294]	
57	58	59	60	61	62	63	64	65	66	67	68	69	70	71				
La 138.91	Ce 140.12	Pr 140.91	Nd 144.24	Pm 144.91	Sm 150.36	Eu 151.96	Gd 157.25	Tb 158.93	Dy 162.50	Ho 164.93	Er 167.26	Tm 168.93	Yb 173.06	Lu 174.97				
89	90	91	92	93	94	95	96	97	98	99	100	101	102	103				
Ac 227.03	Th 232.04	Pa 231.04	U 238.03	Np 237.05	Pu 244.06	Am 243.06	Cm 247.07	Bk 247.07	Cf 251.08	Es [254]	Fm 257.10	Md 258.1	No 259.10	Lr [262]				

Figure 3. General elements used in LIBs (B and Cl could be used in the electrolyte profile as LiBF_4 , or LiClO_4 , Si is used in some anode architectures, Ti is used in Lithium Titanate anodes).

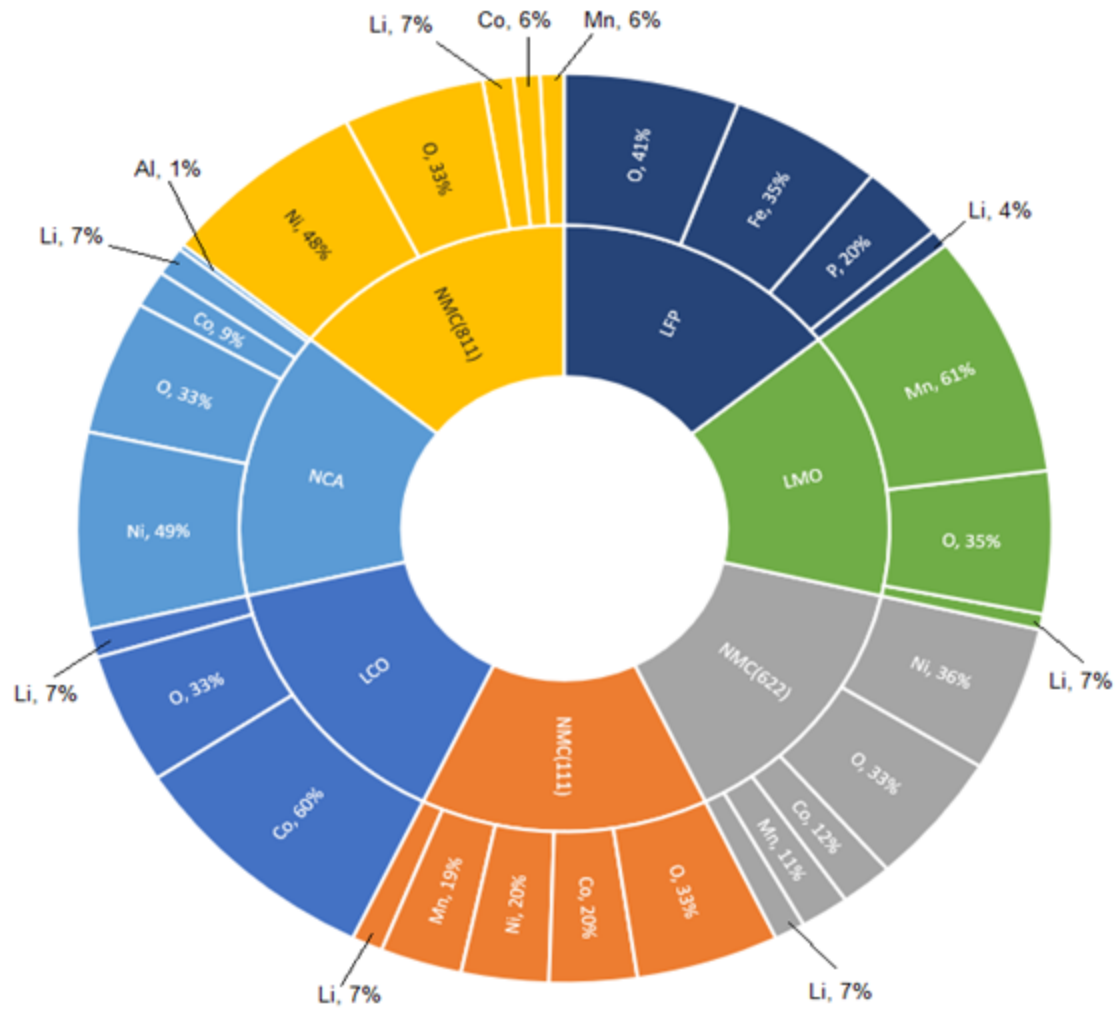


Figure 4. Different elements used (by mass percentage) in different cathode materials adopted from EverBatt Model (Dai et al., 2019b)-

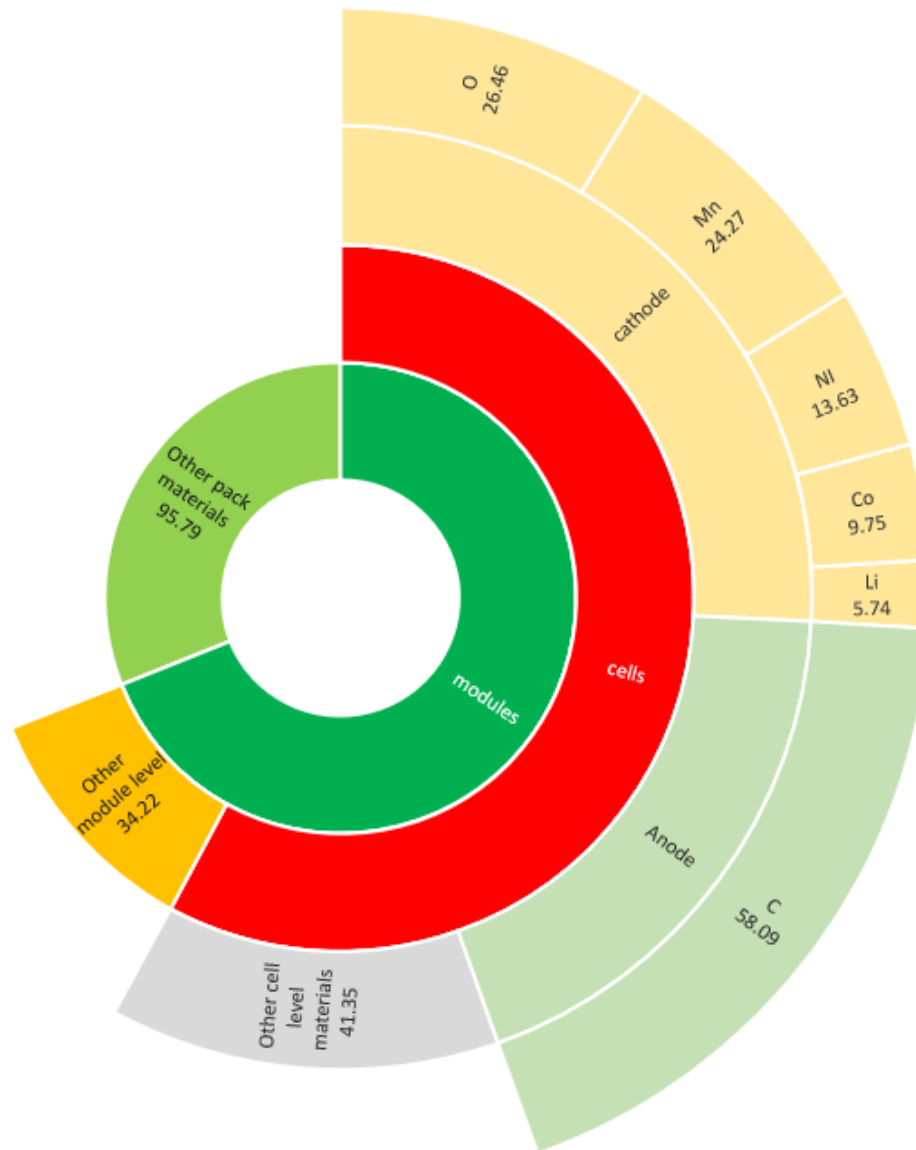


Figure 5. Bills of materials for an EV (Nissan Leaf Tekna 2018) battery with an NMC-532 cathode chemistry (numbers show kg weight).

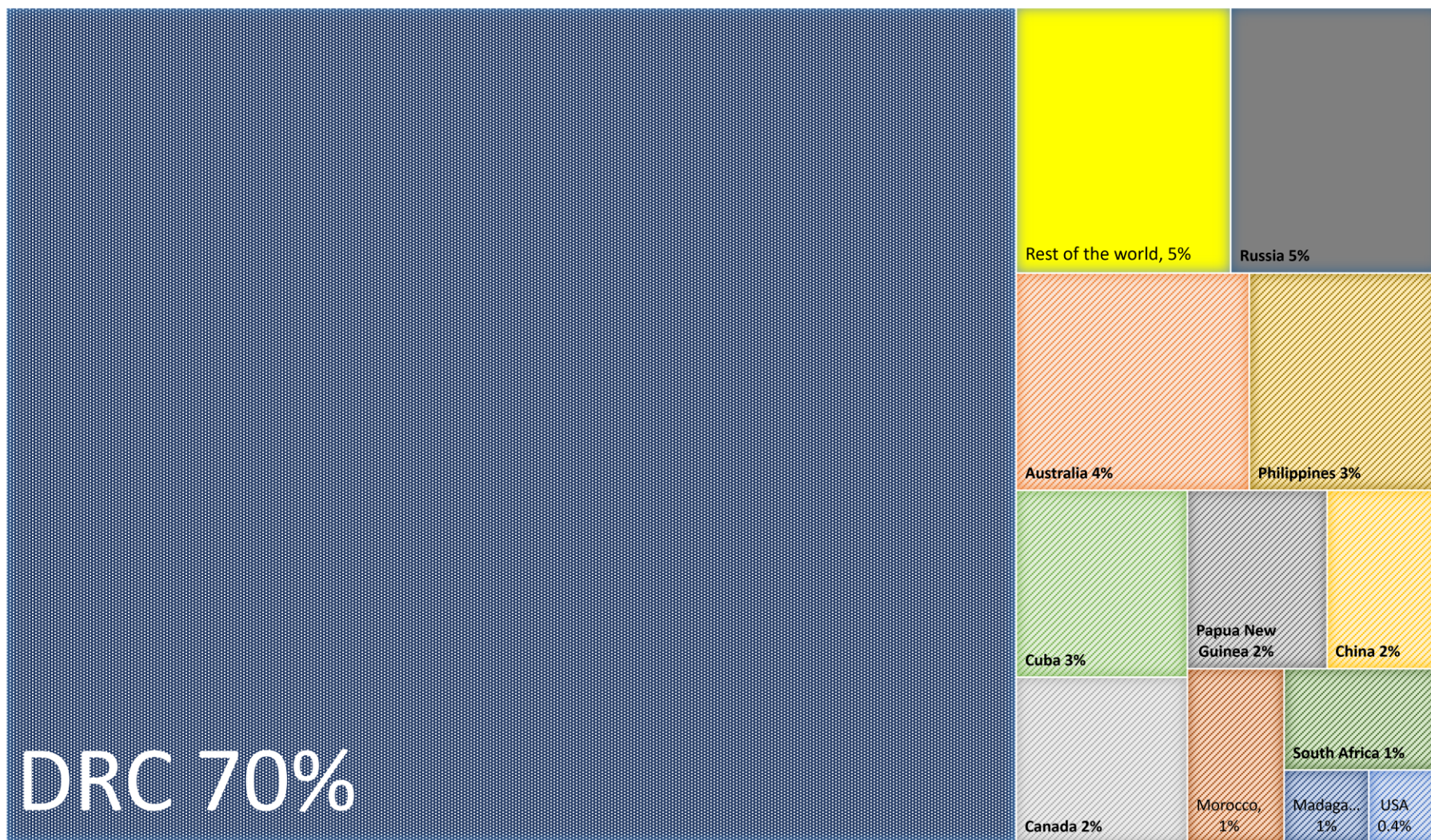


Figure 6. Contribution of different countries from global cobalt production (estimated values for 2020 from (USGS, 2020)).

As the production of LIBs ramps up to meet the growing demand in the mobility and grid energy storage applications, the environmental impacts of mining and upgrading battery materials need more attention (Mayyas et al., 2019). A challenge is that the required data for environmental impact assessment of mining and upgrading activities of battery materials are not available for the majority of mines (or production sites) along different regions. This means that for some supply chains there is a significant lack of data. Some widely used databases for life cycle assessment studies provide the life cycle inventory from only a few mines while considering only one or two conversion routes for minerals. For example, there are many routes for the creation of NiSO but the available inventories do not differentiate them. Overall, apart from some efforts performed in assessing the environmental impacts of mining and upgrading LIB materials, there is still a lack of enough research on comprehensive environmental impact assessment in these areas.

With the upcoming EU regulation on documenting carbon footprint for batteries over 2 kWh capacity (European Union, 2020), all the stakeholders in battery supply chain including mining and refining companies would need to provide the carbon footprint of their processes and might need to optimize their activities to reduce their carbon footprint compared to their opponents in the market. However, there is a risk that companies with a high market share do not show enough willingness to optimize their processes quickly.

It should also be highlighted that upstream activities of battery material production as well as cell manufacturing consume a great deal of energy and therefore the energy supply as well as the consequent environmental impacts of such processes depends on the electricity mix and heat sources (Kelly et al., 2020). On the other hand, for some battery materials, the required energy for mining, or other upgrading processes (e.g. beneficiation, primary extraction, and refining) is mainly provided by different fossil resources among them are natural gas, coal, diesel, and electricity (from the grids with considerable fossil shares). This has to change toward using renewable energy sources to power the mining and upgrading activities in order to make the LIBs more beneficial for the environment, however, costs, materials needed and environmental impacts of using renewable energies for such applications need to be justified.

2.2. Manufacturing

Cost of battery manufacturing is a key manufacturing challenge which impede widespread adoption of LIBs for commercial use in transportation and grid storage applications. In fact, apart from further improvement in energy storage efficiency (in terms of both volume and weight) needed for LIBs (Masias et al., 2021), battery manufacturing cost needs to be reduced to increase the penetration of EVs and make them more competitive with the internal combustion engines. Moreover, reducing these manufacturing costs are critical for a clean energy future and widespread use of renewable energies in electricity generation (i.e. grid storage applications). A Bloomberg NEF report has shown that the average cost of LIBs (pack prices) in the market fall by 89% within the last decade, reached to \$137/kWh in 2020 (compared to \$1,191/kWh in 2010)(BloombergNEF, 2020). It should be noted that about 26% (\$35/kWh) of the pack prices in 2020 belongs to the pack portion (pack level materials, i.e. excluding cells) of the battery while the rest belongs to the cell portion. Although pack prices of less than \$100/kWh was reported for batteries in e-buses in China in 2020, the average market price is expected to reach close to \$100/kWh by 2023 (BloombergNEF, 2020) and further down to \$58-73/kWh by 2030 (BloombergNEF, 2020, IHS Markit, 2020).

There are some cost reduction approaches suggested in the literature, e.g. material selection and innovation, improvement in manufacturing process, pack and cell design improvements, water-based processing, use of solid state batteries (SSBs), etc. but they come with their own challenges (Daniel, 2015, Masias et al., 2021). The diversity of approaches to reduce the battery manufacturing cost make predictions for 2030 uncertain, i.e. depends on the adopted approaches the pack prices could be lower/higher, or the costs could be reduced sooner/later than 2030. Also, raw material supply interruption or price increase could make a challenge for battery manufacturers to reduce the cost of manufacturing in line with the expected numbers.

As a manufacturing challenge, reducing the time for electrolyte wetting, formation, and aging of LIBs is of a critical importance. These processes can take a few days (for wetting and formation) to 1-2 weeks (for aging), contribute to a significant portion of capital expenses and take up one fourth of floor space (Wood III et al., 2019). There are also a number of technical challenges faced by battery manufacturing, reported in the literature (Kwade et al., 2018, Liu et al., 2021).

As a cost and environmental impact reduction opportunity, closing the value chain loop for LIBs through recycling could be a promising solution (Masias et al., 2021). In order to achieve an efficient closed-loop supply chain for LIBs, battery packs end up the end-of-life stage need to be recycled easily, with lower complexities and costs, i.e. they should be designed for disassembly and recycling. Nevertheless, there is no battery pack design standardization at present that lead to easy and efficient disassembly and recycling of packs, modules and cells; and there might not be any in the near future (Arora and Kapoor, 2018). In

fact, the current designs of LIBs have made recycling of them complex, for example, different cell types could affect the efforts needed for the following recycling, e.g. more efforts are needed to recycle cylindrical cells through direct recycling why it is much effortless to recycle pouch or prismatic cells using the same treatment technology. The challenge with design for recycling is to provide an optimized design that consistently works with the end-of-life treatments and lead an effective recycling.

The other issue with the manufacturing is the high amount of energy needed for this stage and its consequent environmental impacts. Recent life cycle assessment (LCA) reports show that manufacturing stage is one of the main contributors to the energy consumption and GHG emissions in the cradle to gate life cycle of LIBs (Ciez and Whitacre, 2019, Dai et al., 2019a, Kelly et al., 2020) and could generate higher GHG emissions compared to an internal combustion engine (ICE) on a cradle to gate basis (Kim et al., 2016). Since the energy consumption of some equipment such as dry room mainly depend on the facility throughput, the energy intensity (and consequently carbon intensity) of the battery assembly is throughput-independent (Dunn et al., 2015). Apart from energy consumption optimization or possible technology advancement that could help energy consumption to stand at a lower level, the carbon intensity of the energy source used for manufacturing is a critical parameter. It is obvious that energy mixes characterized by more fossil fuel could increase the GHG emissions of LIBs life cycle.

Overall, apart from recent research progresses made, LIB manufacturing still lags behind and not much progress has been made (Liu et al., 2021). Battery manufacturing needs to meet the rising demand, strict quality requirements, cost and competition pressure while sustainably produce batteries and generate low waste. Simultaneously, manufacturing must meet market desire for high energy and power densities, safety and longevity which materialize drive longer trips without having to recharge EVs.

2.2. Use

The use phase of EVs does not produce tailpipe emissions, however, there are hidden impacts related to fuelling LIBs. In fact, the environmental impacts of the use phase are shifted to the electricity generation at power plants, that could produce electricity from a different range of sources, from renewables to fossil ones. Although the continued reliance of electricity generation on fossil fuels is not the major obstacle to widespread adoption of EVs, it can diminish the cost-effectiveness and environmental benefits of EV adoption as a climate change mitigation strategy. On the other hand, the increasing trend in the substitution of internal combustion engine vehicles with electric ones would lead to an increased demand for electricity and in case of a national fleet, it could be huge amount of electricity needed. Therefore, the power

generation industry must be able to meet the increased demand to accommodate EVs without straining the electricity grid.

Despite the recent progresses made in increasing the energy capacity of LIBs that significantly improved the range of most electric vehicle models in just a few years, a limited driving range or ‘range anxiety’ still present a challenge to many customers. However, this is not the only technology limitation. In fact, the time taken to charge an EV battery is another concern for many drivers that pose a challenge for EV adoption. There are also some challenges related to charging infrastructure such as patchy charging infrastructure, charging fees, etc. but it seems more related to the sustainability of EVs as a whole rather than their batteries.

2.3. End-of-Life

The rapid adoption of EVs would lead to staggering number of end-of-life LIBs in the near future. Projections show that end-of-life LIB stocks would reach to ca.1.6 million tonnes in 2030 (from ca.0.3 million tonnes in 2020), and that EVs will make up half of the stream (Melin, 2021). Generally, three main pathways could be considered for the end-of-life batteries as disposal, reuse (including remanufacturing and repurpose) and recycling (Mrozik et al., 2021). Disposal, which generally means sending the end-of-life batteries to landfills, is the least preferred option as it leads to material loss, increase environmental risk and health hazards, and is a waste of economic opportunity. Moreover, stockpiling (or even worse, landfilling) and shipping end-of-life batteries abroad as a part of end-of-life vehicle wholesale were found to be less preferable options (Harper et al., 2019b). Therefore, remanufacturing, repurpose and recycling are the proposed pathways to extend the service and benefits of EV batteries. However, adopting these pathways is not a simple and straightforward task and there are significant environmental, economic, technical and policy challenges that needs to be addressed.

The challenges with the end-of-life LIBs start from their removal (from the cars), collection and transportation of them (as parts of the LIBs reverse logistic). Handling end-of-life EV and removing their batteries require specific knowledge, trained personnel and specialized tools (Elwert et al., 2018), which is still lacking in the current practises used in the industry. Health and safety risks caused by handling batteries at this stage also is of concern that could threaten untrained personnel’s life (Harper et al., 2019b). Overall, cost, safety, and time issues are the main concerns with the dismantling batteries from EVs which needs more attention.

Lack of efficient and cost-effective collection system that comprehensively work along different battery chemistries/types and throughout different regions in a country is an impediment to commercialization of reuse and recycling. This is specifically important for low cobalt batteries in which the economic benefit

from the recycling is lower. Transportation costs and harmonization of battery shipment regulations around the world are also the other challenges that needs to be addressed. The other reverse logistics challenges could be finding the optimal locations for collection, reuse, and recycling of LIBs as well as optimizing the material flows in the reverse logistic network. There are also safety concerns regarding the stockpiling of end-of-life LIBs for re-manufacturing, repurposing, and recycling. A list of fire incidents in LIB treatment/collection facilities could be found in (Paul A. Christensen, 2021). The following chapters address some reverse logistic challenges related to reuse and recycling of LIBs.

2.3.1. Reuse

Reuse of EV batteries encompasses remanufacturing and repurpose of them. Remanufacturing is an opportunity to refurbish retired LIBs and use them again in automotive applications after replacing faulty and degraded cells with qualified ones. Despite the benefits that remanufacturing could offer, there is a lack of large-scale remanufacturing of end-of-life EV batteries at the present due to substantial differences between battery designs and chemistries among original equipment manufacturers (OEMs)(Hua et al., 2020).

Repurpose is the re-use of end-of-life batteries in less demanding applications such as uninterruptible power supply (UPS), energy storage systems (ESS), electric forklifts, electric scooters, etc. Cascaded reuse of LIBs in a hierarchy of applications is a suggested way to optimize the environmental impacts from material use in the life cycle of batteries and could be considered as a favourable option prior to recycling (Harper et al., 2019a, Ahmadi et al., 2017). However, repurposing end-of-life batteries in second-life applications may not be economic for all markets (Gaines, 2018). A recent review reported that the OEMs or ESS integrators had a very tiny contribution to the research on the battery second life and thus most of the economic assessments are based on estimations which shows an uncertainty in the calculated achievable revenues for certain stationary applications (Martinez-Laserna et al., 2018). Therefore, there is a lack of techno-economically viable business models that could provide meaningful and reliable economic results. Moreover, the review also showed that there is a lack of enough research and conclusive results on the analysis of second life battery ageing performance as a crucial factor that affects technical and economic viability of second life batteries (Martinez-Laserna et al., 2018). Furthermore, accurate measurement of state-of-health (SOH) and remaining useful life (RUL) of end-of-life batteries is still a challenge, which creates inconsistencies in assessing batteries due to lack of standards (Haram et al., 2021).

Overall, the major concerns with the second life are the economic viability and the market potential (Martinez-Laserna et al., 2018, Haram et al., 2021). In fact, with the anticipated decreasing trend in the

price of LIBs, old batteries may lose their competitiveness in the market especially if labour costs should be added (Melin, 2021). Despite the availability of some second life battery technologies for commercial applications, the market structure to demonstrate the benefits of the second life batteries for such applications is not clear (Abdel-Monem et al., 2017). Another challenge is that, due to some safety risks, specific knowledge, qualified personnel and specialized equipment are required for dismantling and inspection of retired EV batteries, refurbishment and reassembly, as well as cell replacement (Standridge et al., 2014, Hua et al., 2020).

2.3.2. Recycling

The main impetus for recycling of LIBs is to divert materials from landfill, reduce the costs of LIB life cycle, and return strategic and critical materials into the market to avoid their shortage, initial mining and production steps. Nevertheless, recycling of LIBs is still in its infancy and need rapid development in various aspects, to be able to quench the abovementioned desires.

There are different recycling approaches to treat LIBs. Pyrometallurgical, hydrometallurgical (including bio-leaching), and direct recycling are the main recycling processes that could be employed for material extraction from end-of-life LIBs (Lander et al., 2021). These processes could generally benefit from some pre-treatment processes such as discharging, disassembly, crushing, screening and separation (Hua et al., 2020). Therefore, there could be a combination of different main and pre-treatment process to effectively deal with end-of-life LIBs. However, none of these combinations is the economically, technically and environmentally ideal method, each one has its own advantages and downside (Gaines, 2018). Moreover, variability in battery pack designs and chemistries (i.e. different pack architectures, cell designs and chemistries), the ever-evolving design of them, and the fact that the batteries were not actually designed for recycling, made the LIB recycling a complicated and challenging task.

Discharging batteries is a common practice in most of designed recycling processes in order to recover the unused energy of the batteries and make them safer for further handling and treatment (Sommerville et al., 2021). Despite the progress made in using different techniques for discharging batteries such as salt-saturated solutions (Nie et al., 2015, Shaw-Stewart et al., 2019), Ohmic (Krüger et al., 2014), etc. there are still challenges such as long process time, risk of thermal runaway, and potential safety risks that need to be addressed. For example, using brine solutions to discharge batteries would increase the safety risks for damaged or highly-charged cells, and that challenge is more focused by the literature at the pack-level not cell-level (Sommerville et al., 2020).

Another challenge with the discharging stage is the optimum level of discharge which varies according to depth of discharge (DOD) and cell chemistries (Harper et al., 2019b). More specifically, over-discharging cells could result in the dissolution of copper into the electrolyte and lead to contamination of the reclaimed materials in the material extraction stage (most importantly cathode materials) (Sommerville et al., 2020). Any possible increase in the voltage could increase the risk of short-circuiting and thermal runaway (Guo et al., 2016).

Disassembly is also a common practise in most of recycling processes that could be performed at different levels, i.e. module and cell levels. The main challenge with the recycling lies in the fact that there is no widely accepted standard for battery pack, module, or cell designs within the battery manufacturing sector (Arora and Kapoor, 2018). This has led to a substantial difference in the physical configurations of battery pack components (i.e. pack, module or cell) which presents a challenge for disassembling of the batteries due to different approach required for disassembling the different batteries available at the end-of-life market. Moreover, the current disassembly processes involve human participation (Soh et al., 2014, Sommerville et al., 2021) which could be used in combination with some automation. The efficiency of this process could be improved significantly with a rapid harmonization in the standardization for battery configuration and structure in the design stage. However, the main issue is that lack of an optimized design for recycling, which lead to more efforts required for dismantling batteries either by humans or machines (Harper et al., 2019b), and consequently makes disassembly a complicated task. Robotic disassembly as the cutting-edge technology and a promising solution that could reduce the time, safety risks and cost of the recycling also faces some challenges. Generating algorithms and software that can control cheap hardware to behave in a flexible and intelligent way in order to process complex disassembly problems and handle uncertainty remains a major challenge in this area (Harper et al., 2019b). Moreover, there are some technical challenges with the use of cloud modelling and cloud computing in robotic disassembly (cloud robotics) that needs to be addressed (Wan et al., 2016, Yan et al., 2017). It should be noted that overcoming these challenges would also benefit the reuse stage.

Pyrometallurgical processes has their own pros and cons reported in the literature. The main challenges with pyrometallurgical process are the high capital cost, high amount of energy consumption (and GHG emissions), economic reliability on recovery of specific metals, as well as large amount of batteries needed for the operation (large scale operation)(Rajaeifar et al., 2021, Zeng et al., 2014). The trend in the LIB market in reducing cobalt content will significantly affect the economy of pyrometallurgical recycling and presents a challenge for recyclers. Removing some battery materials such as aluminium and copper by the means of a pre-treatment (e.g. shredding before sending the material to the furnace) also add an extra cost to the process and could increase energy consumption by the smelter (Gaines, 2018). Thus, finding an

appropriate and effective pre-treatment as well as alternative source of energy are still among the challenges for such processes (Rajaeifar et al., 2021). Moreover, in order to achieve complete recyclability of LIBs, developing complementary treatment processes is needed to aid pyrometallurgical recycling, however, these processes need to present low cost and energy demand.

Hydrometallurgical process seems to be the dominant technology in the mid-term. However, this treatment method have to deal with issues of process complexity, the neutralization costs, large volume of solvent consumption, long processing time, and materials cross-contamination hazards (Harper et al., 2019a, Hua et al., 2020). Bioleaching process could offer lower costs and emissions, however this recovery method has to be scaled up from the low technology readiness levels (TRLs) and for that needs to overcome some challenges such as long microorganism cultivation (incubation) time, unclear economies of scale, as well as easy contamination (Hua et al., 2020, Qi et al., 2020). Although they are considered as bioprocesses, it does not mean that they have no impact on the environment and therefore this aspect has to be addressed.

As an emerging concept, direct recycling could offer promising advantages and provide a closed-loop recycling of LIBs from EVs through providing high quality battery materials that need the least remanufacturing efforts due to retaining their structure. Low cost and energy demand as well as the potential for significantly reduce the life cycle emissions are among the other advantages of this approach. Nevertheless, direct recycling processes are unable to handle a mixed stream of various battery materials and are successful when single battery chemistry or compatible chemistries are used. Therefore, and considering the lack of design for recycling standards, a challenge is that specialized direct recycling processes is needed for different battery chemistries and types. Alternatively, the input battery materials could be separated based on cathode type, otherwise the value of the reclaimed materials would significantly decrease (Gaines, 2018). However, designing efficient and economical separation technology is a challenging task that needs more investigation. Another challenge is that direct recycling is very sensitive to the cathode contamination by other metals (e.g. aluminium) that could reduce the electrochemical performance. Such low-quality materials will be less favourable in the market (Harper et al., 2019a). In this regard, processes that could offer no or less component contamination are preferable. Overall, this technology still needs improvements to be scaled up from laboratory scale to higher TRLs (Hua et al., 2020). Table 1 also shows some challenges facing the recycling of some battery components.

Table 1. Challenges facing the recycling of some battery components (Harper et al., 2019a, Hua et al., 2020, Nirmale et al., 2017, Gaines, 2018).

Component	Activity or process	Challenge
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binder	Removal through the common pyrolysis or dissolution methods	create some health challenges, e.g. pyrolysis of the PVDF binder releases HF, dissolution using NMP as a solvent creates the risk of toxic emissions
binder	Undesirable reaction between PVDF binder reaction and electrode material in pre-treatment	Lack of enough investigation in the literature
binder	Removal of binders from the black mass to release the metal oxides and graphite	Finding quick and low energy processes that could be employed on a commercial scale
binder	Substitution of fluorinated binders (in order to make recycling easier and more effective)	Finding the appropriate binders such as water-based, cellulose- and lignin-based binders that could be employed on a commercial scale
electrolyte	Electrolyte recovery	Lack of large-scale application of the recent developed methods. Other challenges: lithium salts recovery, recycling cost and purification
graphite	Recovery	Lack of enough investigation in the literature
SEI	Removal	development of process that could effectively remove SEI
separators	Recovery	Lack of processes for effective recovering of separators
aluminum foils/copper foil	Separation	Remaining small copper /aluminum foil pieces in the black mass after segregating the foils make serious concerns in case of reuse of the recovered cathode or anode materials

Overall, recycling processes must be cost-effective and flexible enough to deal with old/current battery chemistries and designs as well as emerging composition of LIBs. In line with that, there is a necessity of harmonization in standards for battery structure and configuration and materialize design for recycling. A big economic challenge, however, is that with the gradual removal of cobalt from the cathode chemistries, the economic impetus for recycling of these batteries reduces since the advantage from the recycling of lower value materials cannot compete with cobalt. In order to improve the economic viability and environmental balance of recycling which is now mainly dependent on cobalt content, and also to maintain a long-term recycling industry, low cost, flexible and more efficient recycling processes are urgently needed (Mayyas et al., 2019, Harper et al., 2019a). Also, despite the promising advantages of closed-loop recycling, there could be a difference in chemistry of the recovered cathode and anode materials and the required cathode and anode chemistries in future battery designs. Moreover, there are questions about the economies of scale of recycling processes in the future expecting a dramatic increase in the volumes of end-of-life batteries (Wang et al., 2016).

Recycling processes has their own environmental impacts and might pollute environment. Thus, recycling processes must be thoroughly considered from the environmental aspect. At the moment most of research done in assessing the environmental impacts are focused on GHG emissions, neglecting the other emissions

and impact categories. Also, the life cycle inventory data for different commercial processes are not widely available. The challenge is that in the lack of commercial data, environmental impact assessment of recycling processes needs to be considered at high TRLs (while recycling technologies are still infant) and for future conditions (prospective environmental impact assessment). These issues are often neglected in the literature.

3. Recent developments and future research directions

The current chapter focuses on the collected papers in this VSI, providing a review of the published papers and discussing their main findings. Various recent studies focused on demand projections of EVs and related resources needed in various countries. Castro et al. (2021) provided projections for Brazilian EV market by 2030. The authors assessed the effect of the vehicle fleet expansion on the demand flows of active materials using an MFA approach, and evaluated how different waste management strategies could respond to the increased material flow for batteries. It was concluded that in 2030, there would be a major demand of up to 180 thousand tons in Li, Co, Ni, Mn and graphite as a result of 1.8 million new EVs entering the Brazilian market. The surprising fact is that the current domestic production of the country for Li and Co could not supply the required materials for the estimated number of EV in 2030, which means that either an increase in the production or import rates must be followed. The Brazilian fleet expansion in the future is staggering enough to affect the global mineral flow and contribute to the future mineral supply disruptions. There will be also more than 340 thousand units of retired EV batteries in the same year which need adoption of proper management strategies to help avoiding supply disruption of minerals through recycling, repurposing, or remanufacturing.

Yao et al. (2021) projected the nickel demand from nickel-bearing batteries and the potential for nickel recycling from end-of-life EVs in China in 2030. Projections showed that the number of Electric and hybrid vehicles that will be placed on the market in China in 2030 could reach up to 2300, 3800, and 9800 thousand units under different penetration rates (i.e. lower growth rate, standard growth rate, and higher growth rate, respectively). Findings also illustrated that the nickel demand for manufacturing nickel-bearing batteries can reach 140 thousand million tons in 2030 while nickel recycling could supply between 19.5-32.3% of the total demand for manufacturing such batteries (depends on the growth rate considered) and thus should be considered as a vital strategy. The assumption for nickel recycling potential however considers complete recycling of end-of-life vehicles which needs proper collection and waste management strategies to be adopted. Fallah et al. (2021) provided a long-term projection of the number of EV fleets and end-of-life battery stock in the Republic of Ireland. The study addressed the uncertainties in the end-of-life battery stock estimation by detecting different influencing factors and modelling the outcomes under different

scenarios. Moreover, the impact of government policies on the evolution of vehicle technologies in Ireland along with other factors such as supply-side growth and cost parity in the automobile industry were also investigated. Results showed that adding an end-of-life value to the retired batteries could increase the sale of EVs over the subsequent years. Moreover, adopting more environmentally friendly policies could significantly change the trend of EV adoption in Ireland. In fact, the authors found the government intervention as the most critical element for achieving a fully electrified transportation system and recommended continual policy support by the government in order to achieve a faster transition to an EV fleet. In this context, future research can focus on development of such studies focused on developing countries, especially highly populated countries and top mineral producers, in order to account for their contribution on future supply/demand patterns (Castro et al., 2021). Besides, future research works should be focused on developing more applicable prediction models and collecting more actual data on EV stock and EoL EVs (Yao et al., 2021).

Hu et al. (2021) studied the international EV-LIB trade and proposed a trade network risk transmission model to study the hidden systemic risks in international EV-LIB trade. More specifically, a complex Network Analysis tool was utilized to reveal trade interactions from a systematic, dynamic, and global perspective. Additionally, the systemic hidden supply risk in the EV-LiB trade was investigated. It was revealed that although their trade network has heterogenous and diverse structure, but such diversity would not work well for China and Germany. It was also highlighted that although the supply disruption risk of the US and Germany is considerable, but it is negligible compared to China and South Korea's supply disruptions. In the latter scenario, 277 countries will be affected negatively. Finally, it was recommended to automakers to establish localized LIB manufacturing in conjunction with the EV assembly plants to avoid such disruptions. The authors recommended that a simultaneous consideration of supply and price volatilities should be studied, as the future work.

Many researchers have provided some empirical evidence for EV battery regulation and adoption challenges. Albertsen et al. (2021) reviewed the circular business models (CBMs) and circular economy (CE) strategies in the literature. The authors scrutinized the type of implemented CBM by European original equipment manufacturers (OEMs) as well as their driving factors. It was found that greener products made from the second life applications may not be appealing for all customer segments. They have to be either cheaper or provide same quality and warranties. In another empirical study, Azadnia et al. (2021) discussed various barriers to develop end-of-life management of LIBs from EVs through reverse logistics activities in European context. They concluded that 'market and social', and 'policy and regulations' categories are the two most influencing barriers to the implementation of LIBs from EVs reverse logistics. This result was complemented by Kumar et al. (2021) where they identified the challenges in sustainable supply chains of

EV batteries. It was found that ‘availability of the charging stations’ and ‘renewable energy production’ are two of the most prominent challenges in a developing country like India. Future research in this area calls for more empirical studies in identifying risks, challenges and gaps in business models and regulation directives for end-of-life management of EV batteries. Furthermore, comparison of such models among developing and developed economies will provide more in-depth insights.

Transportation and energy sectors are expected to benefit from LIBs towards achieving their net zero targets in the next few decades. This might however create a material demand conflict between different applications and will put a much higher demand on raw metals for batteries compared to the current demand in the market. In their paper focusing on the nation-wide demand for use and recycling of critical LIB metals (i.e., Li, Co, Ni and Mn) for EVs and grid storage in the UK, Kamran et al. (2021) found that over the next three decades, end-of life EV battery recycling is a promising solution to this that may be very effective at significantly reducing demand for these virgin metals, to the point where their net demand in 2050 could be equal to or lower than at present, despite a projected shift to 100% light-duty EVs within the same time frame. Additionally, their research indicated that the adoption of shared mobility schemes would be capable of reducing the demand even further, temporarily creating an oversupply of secondary metals after 2040. The authors also identified a knowledge gap in consequential environmental analyses of scenarios that co-evolving energy and transport sectors. One element to perform such analyses is the availability of primary inventory data. Crenna et al. (2021) identified a critical knowledge gap in the availability of production-scale inventory data for lithium-ion batteries, and proceeded to fill that gap by proposing a modular approach and developing a set of updated “cradle-to-gate” life-cycle inventories for the predominant battery chemistries in the current EV market, namely: NMC111, NMC811 and NCA. The modular approach employed by the study could be also used as common framework to model different types and generations of LIBs. The study also discussed the sensitivity of the results to a range of modelling options, implying the relevance of transparency when making choices in compiling the LCI.

End-of-life repurposing of EV batteries for second-life applications is a complementary strategy to recycling, which concurs to the reduction of the overall environmental impacts of the LIB supply chain. To accurately quantify the benefits arising from second-life applications, however, careful attention must be paid to the chosen allocation method to allocate the impacts between first and second use. Wilson et al. (2021) developed a novel physical allocation method based on a range of critical performance criteria, and applied it to a second-life case study in Australia. Results showed that battery repurposing can achieve significant carbon emission benefits, but that there are clear break-even points in terms of remaining storage capacity and module retention rate at the end of the first life. Still on the topic of end-of-life repurposing for second life applications, Schulz-Mönnhoff et al. (2021) turned their attention to how to correctly

take into account the combination of different battery storage applications in multi-use second life scenarios. A novel life cycle assessment framework was proposed, and its application to a German case study was tested. Results showed that carbon emission benefits for multi-use scenarios can be 10-22% lower than in single-use applications.

A further aspect of the end-of-life management of EV batteries that needs to be carefully addressed is that of their collection and transportation. Slattery et al. (2021) conducted a literature review on how such processes are handled in various regions of the world, and found that 70% of the reviewed studies indicated collection and transportation as lingering challenges or obstacles to battery end-of-life management. Also, in terms of the relative shares of the environmental burdens of recycling, these preliminary but necessary steps tend to account for between 1% and 3.5% of the total life cycle GHG emissions. It is recommended that future research focus on locating optimal places for battery collection and storage, considering both existing and projected infrastructure while examining local environmental, cost as well as social impacts. Furthermore, there is a necessity of further research in some safety and regulatory aspects as well as cost considerations for storing batteries (Slattery et al., 2021).

In order to achieve sustainable EV battery use, higher levels of circularity of battery systems are needed. A qualitative assessment of 44 commercial LIB recycling companies showed that most of the developed recycling process among the industry aimed at recovering valuable metals and sometimes graphite, while less effort put on reclamation of other battery materials e.g. plastics, lithium salts, solvents, and phosphorus (Sommerville et al., 2021). In case of plastics, although many companies separate them from the battery waste stream, their reclamation is not a widespread practise, i.e. it is not clear if companies send plastics for further recycling, disposal or any other treatment processes. Since it is anticipated that the quantity of high value metals drops in the future battery design, there is a need to focus on efficient recycling of more components as possible (Sommerville et al., 2021).

The review of the current commercial LIB recycling processes by Sommerville et al. (2021) showed that manual disassembly is the dominant practice for dismantling batteries in the industry. Manual disassembly is time consuming, costly and may bring safety risks which necessitates the inclusion of automation in the disassembly process. Glöser-Chahoud et al. (2021) highlighted the importance of a systematic industrial disassembling system as a key enabler of circular economy solutions for end-of-life EV batteries. The study developed a qualitative framework supported by quantitative figures from lifecycle and economic assessments as well as stochastic simulation models to demonstrate the contribution of industrial disassembling processes to different end-of-life LIB utilization pathways including both recycling and second-life concepts. The findings of the study highlighted that in line with achieving a long-term

sustainable electromobility, end-of-life treatment of LIB needs to be further improved, particularly by establishing a well-functioning systematic and in the best case automated industrial disassembling to enable the economic feasibility of optimum utilization for each component (entire battery systems, modules or cells). However, battery design as a major challenge for automated disassembly must improve to enable efficient closed-loop supply chains (CLSCs) for LIBs including both second-life concepts through repurposing or reconditioning and subsequent efficient recycling.

Closing the recycling loop for end-of-life LIBs through direct recycling, i.e. a recycling approach by which the active cathode materials are reused in the new batteries with minimal treatment; is always preferable compared to general closed-loop or open-loop recycling where recovered metal salts needs to be further processed to be used in battery manufacturing (Sommerville et al., 2021). In fact, direct recovery of cathode materials could offer great economic and environmental advantages. As a promising approach toward direct recycling of cathodes with PVDF binders, Ji et al. (2021) proposed an environmentally friendly and efficient process to separate and recover cathode materials by investigating binary eutectic systems of three inorganic lithium compounds, i.e. LiCl, LiNO₃ and LiOH. The highest peel-off efficiency was found in LiOH- LiNO₃ (LHN) system (as 98.3%), which helps the decomposition of binder and capture release of hydrogen fluoride (HF) which is a toxic air pollution if released (Mrozik et al., 2021). Such a treatment does not significantly change the crystal structure chemical composition and morphology of the recovered materials while it improves lithium content, compensating the loss of lithium owing to charging/discharging. The proposed approach reveals new potential for sustainable direct recycling of end-of-life LIBs (Ji et al., 2021). However, since purity level of the material waste streams is important to achieve direct recycling, still more sophisticated and independent processes is needed to take off the cell packaging and the cell components.

The qualitative assessment performed by Sommerville et al. (2021) also showed that shredding and separation is the most used treatment process prior to chemical extraction processes such as hydrometallurgical recycling. This has also reported by Thompson et al. (2021) in which they referred to shredding as a commonly employed preliminary step in the recycling of end-of-life LIBs due to its several advantages, e.g. reducing the size of battery material, promoting reaction, facilitating passivation, etc. Nevertheless, shredding could lower the purity of the recovered materials leading to a reduction in economic profitability of the process (Thompson et al., 2021). Thompson et al. (2021) performed a comparative techno-economic research among ten different hydrometallurgical approaches to LIB recycling to compare the use of shredding vs. disassembly. The results showed that cell disassembly could bring more cost saving (in the range of 40-80%) compared to shredding (<20%). However, there are some barriers to quick, efficient and hassle-free dismantling that originates from complex cell design with

numerous cells and modules in a battery pack, therefore simplifying the arrangement of cells as well as considering design for recycling are of great importance (Thompson et al., 2021).

In order to drive the circular economy of EV batteries forward, policies with more holistic reuse and recycling options are needed (Sommerville et al., 2021). The mentioned policies should target higher energy efficiency for the reclamation processes, and closed-loop recycling that benefits from green reagents if hydrometallurgy is used. Currently, hydrometallurgy and pyro-metallurgy are mainly engaged in recycling processes for material extraction from end-of-life LIBs, that generate a great amount of GHG emissions (Rajaeifar et al., 2021) and use synthetic chemicals that may risk human health and ecosystem quality. Bio-hydrometallurgical recycling approach, as an environmentally friendly pathway, uses microorganisms and biotechnology to leach and recover target metals at a lower cost (compared to pyro- or conventional hydrometallurgy) and greater feasibility of implementation in large scale without generating hazardous or toxic products. Bioleaching processes-which are based on acidolysis, redoxolysis or complexolysis mechanisms, and involve either contact or non-contact procedures- are affected by various factors, e.g. type of microorganism, pre-treatment, substrates, pulp density, pH and temperature, etc. To further enhance bioleaching efficiency of metals from LIBs, catalysts may be introduced and organic acid production may be optimized. The pregnant leaching solution (PLS) also needs proper recovery of the target metals to make the whole technology economically viable. Biosorption, bioprecipitation and bioelectrochemical systems are the most promising approaches to recover metals from PLS, each has its own advantages and drawbacks. All these different processes and mechanisms, as well as the influencing factors to bioleaching were reviewed by Sethurajan and Gaydardzhiev (2021). Overall, recovery of metals from LIBs using biotechnological methods is still in its infancy and requires further research specifically for improving efficiency and selectivity (Sethurajan and Gaydardzhiev, 2021).

There are also some recent development in LIB material reclamation processes which presented by some published papers in this VSI. Lin et al. (2021) developed a novel hydrometallurgical leaching process using Organic Aqua Regia (OAR) that was made from mixture of pyridine (Pr) and SOCl_2 (at varied molar ratio)- to recover lithium and cobalt from end-of-life LCO batteries. The leaching efficiency and kinetics of the leaching process under ultrasonication were studied by optimizing leaching process and performing sensitivity analysis of various factors in leaching, where leaching efficiency was compared with other acids and activation energy was determined. The results showed a leaching efficiency of 99% for Lithium and 94% for Cobalt, showing a promising performance compared to nitric and citric acids as two common acid leachants. Moreover, the proposed process requires less electricity, water, and time. In addition, global warming potential (GWP) of the process was considered and compared with that of the other acids, and the results showed less GWP for OAR (Lin et al., 2021). In another study, Fu et al. (2021) presented an

innovative pre-treatment approach for recycling of organic binders from end-of-life LIBs. Instead of using high temperature to separate organic binders from cathode materials (that leads to hazardous fluorinated gas generation), the invented pre-treatment approach employs supercritical carbon dioxide (SC CO₂) and dimethyl sulfoxide as co-solvent to remove polyvinylidene fluoride (PVDF) and liberate cathode material from aluminum substrate in end-of-life LIBs. The optimum conditions were obtained at 343K, 80 bar and 13 min, giving a high efficiency (i.e. 98.5 wt% pure PVDF dissolution), energy-saving and environmental-friendly features (Fu et al., 2021). dos Santos et al. (2021) proposed a hydrometallurgical process-called RecycLib that employs less-cost reagents with reduced environmental impact to recover valuable active materials in LCO and NMC cells in cylindrical and prismatic cell geometries. The proposed treatment is capable of recovering over 90 % of cathodic metallic oxide from LCO and 80% of that from NMC cells. The RecycLib technology showed a great potential for environmental improvement and economic performance enhancement.

Overall, in order to establish sustainable circular economy of LIBs, constant desire for green, economic and efficient recycling processes (that recover the most materials they can) is ever required. More collaboration between academia and the battery recycling industry is needed in order to establish vigorous circular economy strategies that supported by environmentally friendly, flexible and cost-efficient recycling processes.

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