Title
Lithium-ion battery material circularity: material availability, recycling economics, and the waste hierarchy

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Lithium-ion battery material circularity: material availability, recycling economics, and the waste hierarchy

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Abstract

Electric vehicles (EVs) are a critical element of strategies for decreasing global greenhouse gas emissions. EVs are powered by lithium-ion batteries, which are material-intensive and require mining and processing that result in environmental and social impacts. Several governments, including the European Union, the state of California, and the Republic of China, have recognized that reusing, repurposing, and then recycling the battery at its end-of-life, also known as the waste hierarchy, is necessary to mitigate the externalities of the transition to EVs. Policy focused on the lithium-ion battery end-of-life have focused on recycling, and particularly on recovering cathode materials instead of the anodes and other materials, due to their comparably higher environmental, economic, and social impacts, as well as the geographical concentration of production. Prior studies have evaluated these impacts but have not considered the influence cathode chemistry and technological development may have on material circularity, resulting in a difference of material composition between batteries reaching their end-of-life and batteries currently being manufactured.

For policies intended to create a circular EV battery industry to be effective, the quantity of materials reaching their end-of-life, and the environmental and social tradeoffs between end-of-life solutions, must be determined. This research uses methods from industrial ecology, including material flow analysis and life cycle analysis, to address a gap in lithium-ion battery policy; in particular, the design and evaluation of policies that consider the rapid evolution of lithium-ion battery technologies which will result in decreased use of cobalt and an increase in energy density. Material flow analysis is used to calculate spatially and temporally resolved battery material flows and propose a method for calculating feasible recycled content standards, accounting for cathode chemistry mix, EV sales, and lifespan. Technoeconomic assessment is *used to evaluate the cost of recycling, considering location, cathode chemistry, and transportation mode. Lastly, life cycle analysis is used to evaluate the material life cycle impacts of directly recycling high cobalt batteries in comparison to extending the lifespan through reuse.*

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Summary of abbreviations

Al: Aluminum

Co: Cobalt

Cu: Copper

CO_2e: Carbon dioxide equivalent

EPA: Environmental protection agency

EPR: Extended producer responsibility

EV: Electric Vehicle

EU: European Union

Fe: Iron

GHG: Greenhouse gas

Gr: Graphite

GWP: Global warming potential

IWG: Interagency Working Group

LCA: Life cycle assessment

LCO: Lithium-cobalt-oxide

LFP: Lithium-iron-phosphate

LIB: Lithium-ion battery

LMO: Lithium manganese oxide

Mn: Manganese

MFA: Material glow analysis

NCA: Nickel-cobalt-aluminum

Ni: Nickel

NMC: Nickle-manganese-cobalt

NO_x: Nitrogen oxide

PHEV: Plug-in hybrid electric vehicle

RoW: Rest of world

Si: Silicon

SoH: State of health

SO_X: Sulfur dioxide

US: United States

1. Introduction

1.1. Motivation

The transportation sector is a large contributor to global greenhouse gas emissions (GHGs), representing about 15% of greenhouse gas emissions (GHG) (Skea et al., 2022). Electrifying this sector is a key GHG mitigation strategy that can greatly contribute to achieving international climate goals (Skea et al., 2022). Research using life cycle assessment (LCA) has shown EVs result in lower life cycle impacts than internal combustion engines, placing them as an important determinant in decarbonizing transportation (Ambrose et al., 2020; Pero et al., 2018). Lithium-ion batteries (LIBs) are a crucial enabling technology, powering the traction motor for electric vehicles (EVs).

While EVs have the potential to significantly reduce GHG emissions and local air pollutants, their production is not without harm. The technology's promise has led scholars to further analyze the issues associated with LIBs, including the life cycle impacts and the potential for material reserves to be able to meet forecasted demand (Ambrose & Kendall, 2016; Klimenko et al., 2021; Sovacool, 2019).

The supply chain is a hot spot in the life cycle impacts of LIBs due to the environmental and social externalities of ore extraction and processing sites (Ambrose & Kendall, 2016; Bauer et al., 2015). In addition, the majority of LIB materials are categorized as *critical* due to their economic importance, difficulty to substitute, and susceptibility to supply disruption (Olivetti et al., 2015; U.S. Department of the Interior, 2021). Forecasts demonstrate the current reserves of cobalt and nickel will not be able to support EV demand up until the turn of the century, and the use of recycled materials may be necessary (Klimenko et al., 2021; van den Brink et al., 2020).

Efforts to reduce these associated impacts and address foreseeable barriers to EV uptake, include research on:

- Battery technology, including material substitution and increased energy density
- Reusing and repurposing batteries at the end of their use in an EV
- Material recycling and circularity

Innovation of battery technology has the potential to decrease the materials demanded and substitute those that are particularly harmful. This is currently occurring through the decreased use of cobalt in high-capacity lithium-ion batteries which use a nickel-manganese-cobalt cathode. In addition, large impact reductions are possible through the recycling and reuse of LIBs after their an EV (Richa, Babbitt, & Gaustad, 2017). use in an EV (Richa, Babbitt, & Gaustad, 2017). The continued use of products through reuse, and the use of recovered materials in manufacturing, is conceived of as the circular economy. Contradictory to the typical linear model, at the end of life, the EV is not waste, but an asset and essential input to future manufacturing (Ellen Macarthur Foundation, 2013).

Material circularity is understood as a key tool in creating a sustainable battery ecosystem and decoupling industry growth from extraction. It has the potential to solve two issues: material availability and material extraction impacts. In addition, the repurposing of batteries for a stationary application, prior to recycling, has the benefit of extending the lifespan and using the battery to support renewable energy development (International Energy Agency, 2020).

This work is motivated by the potential to decrease LIB impacts through the circular economy model. The research approach reflects the perspectives and methods of industrial ecology, applying LCA and material flow analysis (MFA) to calculate the circularity potential of LIB materials, propose circularity policy, and assess the preferable end-of-life pathways for EV batteries. In addition, techno-economic modelling is used to analyze the economics of recycling pathways.

1.2. Research contributions

The goal of this research is to provide a technically sound analysis that will help guide policymakers and regulators. The research questions were inspired by conversations with industry professionals and government agency representatives who were seeking to better understand how to mitigate the impacts and risks of EV deployment.

This research contributes in the following ways:

- Provides an overview and perspective of current LIB policy
- Estimates material demand and circularity potential for key LIB materials at a global and regional scale
- Assesses the impact of cathode chemistry change on material circularity
- Proposes recycled content standard for the US and the use of MFA in their estimation
- Estimates the economics of recycling mixed cathode chemistry streams retired in the US
- Assess the economic and environmental impact of recycling batteries retired from the US in China, versus domestically
- Evaluates if high cobalt batteries should be reused when more material efficient batteries are manufactured. Thus, assessing the impact of technological development on the waste hierarchy.

These contributions are part of 4 sections:

Section 4 is a perspective piece and overview of LIB policy. Emphasis is on US policy at both a state and federal level.

Section 5 uses MFA to calculate the theoretical maximum of potential LIB material circularity at the global and regional level (US, Europe, China, RoW). Material circularity is estimated under varying conditions for dominant cathode chemistry and LIB lifespan.

Section 6 then narrows the scope of material circularity research view to the US. The tightened geographic focus mirrors the regional management of batteries. The MFA developed in section 5 is extended, increasing complexity to better represent practical constraints on a circular battery economy. This enhanced MFA is then used to calculate achievable recycled content standards for the US. The economics and life cycle environmental impacts of recycling are assessed to provide greater insights into recycling benefits and possible areas of cost and emissions abatement. This section is inspired by the recycled content standards proposed by the European Union and the debate over proposing these standards by the California Lithium-ion Battery Recycling Advisory Group (European Commission, 2020; Kendall et al., 2022).

Section 7 assesses the influence of technological development on the waste hierarchy. As policy development is underway, and the LIB repurposing and recycling industry expands, crucial questions about the environmentally preferable path for batteries has been an aspect of vibrant discussion. This research uses LCA to review the relative environmental benefits or disbenefits to directly recycling high-cobalt LIB chemistries for material recovery and use in the manufacturing of new batteries with higher cobalt content.

This interdisciplinary research required the understanding of: 1) the technical aspects of lithium-ion batteries and their end-of-life processing; 2) the criticality and availability of materials used;

3) the social and environmental implications along the supply chain and life cycle; 4) possible routes of impact reduction; and 5) LIB policy development. Sections 2 through 4 provides this overview, before diving into the quantitative research in Sections 5 through 7.

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2. Lithium-ion battery technology, materials, and end-of-life processing

2.1. Lithium-ion battery technology

The lithium-ion battery (LIB) is a favorable storage technology due to its high energy density, efficiency, and cycle life (Miao et al., 2019). The battery is made up of an anode, a cathode, two current collectors, a polymer separator, and electrolyte. The battery stores electricity through a chemical reaction which causes lithium-ions to flow between the anode and cathode, passing through the electrolyte and the separator. When the battery is discharging, the lithium-ions flow from the anode to the cathode. The chemical imbalance then allows the electrons to flow through the positive current collector, to the device receiving the electricity, and back to the battery through the negative current collector (Miao et al., 2019).

The current collectors are typically made from aluminum and copper, the anode is typically graphite, and the cathode has multiple variations containing critical materials which will be elaborated on in Section 2.1.1. The impact of cathode and anode material substitutions on environmental impacts and circularity potential are a point of research and discussion throughout the dissertation.

2.1.1. Cathode chemistry

The cathode chemistry is an area constantly under development due to its influence on the battery cost, safety, energy density, lifespan, environmental impact, and supply risk susceptibility (Miao et al., 2019).

Over the past two decades there have been eight prominent cathode chemistries (Table 2.1). The chemistry containing the highest amount of cobalt is lithium cobalt oxide (LCO), which is popular in electronics but was only used briefly in the Tesla Roadster and Smart Fortwo electric drive, but then was quickly replaced with less expensive and less environmentally harmful chemistries. Nickle-manganese-cobalt followed, referred to as 'NMC111' due to equal ratios of the nickel, manganese, and cobalt. This chemistry has been developed over time to contain less cobalt with names that continue the nomenclature: NMC523, NMC622, and most recently NMC811. The lower cobalt NMC chemistries are more energy dense and lighter. The chemistry nickel-cobalt-aluminum (NCA) contains slightly more cobalt than the NMC811 and is produced by Panasonic, although Tesla is the only EV manufacturer to use the chemistry.

Lithium-iron-phosphate (LFP) is much less expensive to produce but has lower energy density (Miao et al., 2019). Historically it has not been as prevalent in EVs, and mostly used in China, with numbers dwindling since 2013 (EV Volumes, 2020). This trend is expected to reverse because of the announcement by Tesla that they are using LFP in over half of their Model 3 line (Lambert, 2022; C. Xu et al., 2020). Lastly, lithium-manganese-oxide (LMO) was popular while combined with NMC due to its high internal resistance, enabling fast charging. This chemistry is now rarely used because it has lower capacity and lifespan than comparable chemistries (Miao et al., 2019).

The use of cobalt in LIB cathodes has been of great concern due to its environmental impacts, cost, human rights impacts, and susceptibility to supply chain disruption. As previously discussed, industry and researchers have been developing chemistries that use less cobalt (e.g. NMC622 and NMC811), and companies are switching to chemistries that use no cobalt (e.g. LFP), although it is still in use due to its high specific energy that enables compact LIBs to have a high discharge rate (Miao et al., 2019). Since cobalt is the most expensive material in the manufacturing process, it is also the most lucrative to recover from recycling. Several researchers have stated this trend towards low cobalt chemistries, or away from cobalt completely, will make an already struggling recycling industry uncompetitive, a topic which is explored in Section 6 of this dissertation (Harper et al., 2019).

Table 2.1: The eight prominent cathode chemistries used in lithium-ion batteries for automotive applications.

Acronym	*Stoichiometry*	*Materials*
LCO	$LiCoO2$	Lithium Cobalt Oxide
NMC 111	$LiNi0.33Mn0.33Co0.33O2$	Nickel Manganese Cobalt
NMC 523	$LiNi0.5Co0.3Mn0.2O2$	Nickel Manganese Cobalt
NMC 622	$LiNi0.6Mn0.2Co0.2O2$	Nickel Manganese Cobalt
NMC 811	$LiNi0.8Mn0.1Co0.1O2$	Nickel Manganese Cobalt
NCA	$LiNi0.84Co0.12Al0.04O2$	Nickel Cobalt Aluminum
LFP	$LiFePO4$	Lithium Iron Phosphate
LMO	$LiMn2O4$	Lithium-ion Manganese Oxide

2.1.2. Emerging technologies

Technology innovation has been focused on increasing energy density to lessen the use of critical materials. Recent research has shown there may be potential for LIBs to increase density with the use of silicon, instead of graphite, in the anode (Baasner et al., 2020). Silicon anodes are not yet market-ready due to volume change during charging and discharging, which results in cracking, capacity loss, and a potential safety risk from thermal runaways (Baasner et al., 2020). While silicon anodes enable higher density, research has demonstrated some solutions to accommodating for the silicon expansion, such as nanostructured anodes, results in higher impacts and similar density to typical graphite anode battery (Wu & Kong, 2018). Research on other approaches, such as the columnar silicon thin film anode, is underway to grasp the benefits of silicon (Piwko et al., 2017).

Solid-state batteries have an even greater potential to change the market. These batteries use a solid electrolyte of either polymer or an inorganic solid, such as glass, resulting in 20% more energy density. In addition, a solid electrolyte enables the use of lithium-metal as an anode, which could increase the capacity by 70% (Watanabe et al., 2019). Unfortunately, these batteries are still in the development stage with significant hurdles to overcome, for example the inability for electrons to easily pass through the solid electrolyte and the buildup of lithium dendrites, resulting in capacity fade (Janek & Zeier, 2016). QuantumScape is a startup which claims they have had success with their solid-state battery, having partnered with automotive companies, they are aiming for production within the next two years (QuantumScape, 2022).

If this technology does succeed, it has the potential to abate, but not get rid of, the environmental burdens of LIBs (Kallitsis et al., 2020; Lastoskie & Dai, 2015). An LCA comparing solid-state batteries with those using liquid electrolyte found the environmental burden was lower in all categories assessed due to 1) the increase in capacity, requiring less material to attain the desired range and decreased weight of the battery, and 2) decreased energy demand in the manufacturing stage (Lastoskie & Dai, 2015).

2.2. Material impacts and availability

While EVs result in lower life cycle GHG emissions than the traditional internal combustion engine vehicle, the high material usage raises concerns over their resulting impacts (Ellingsen et al., 2014) and the availability of resources to meet upcoming demand (Klimenko et al., 2021).

It is vital to assess these concerns in order to understand areas of impact mitigation and potential risks to expanding LIB manufacturing. This section explores the impacts, availability, and producers of the following materials: cobalt, nickel, lithium, and copper. Table 2.2 reports the top producers, along with life cycle energy use, SO_X, and water use of LIB materials.

Table 2.2: Geography of production and environmental indicators of LIB Materials. Data about top producers is from USGS (National Minerals Information Center & USGS, 2021). LCA impacts are per-kg of battery material (Dai et al., 2019)**.**

Material	*Top Producers (2019)*	*Energy Use (MJ/kg)*	*SO_X (g/kg)*	*Water use (L/kg)*
Cobalt	Congo (71%), Russia (6%), Australia (5%)	55.96	25.35	72.37
Nickel	Indonesia (30%), Philippines (16%), Russia (10%)	27.07	244.18	27.56

Lithium	Australia (55%), Chile (23%), China (10%)	16.23	2.38	2.85
Manganese	South Africa (29%), Australia (17%), Gabon (13%)	3.06	7.14	0.65
Copper	Chile (28%), Peru (12%), Congo (7%)	44.47	145.59	13.4
Aluminum	China (56%), India (6%), Russia (6%)	121.6	26.62	228.57

2.2.1. Cobalt

Cobalt production is geographically concentrated, with about 70% produced from mines in the Democratic Republic of the Congo (DRC). Global demand for cobalt is expected to drastically increase as a result of increased battery demand (Helbig et al., 2018). Research shows this increase is unlikely to be met with virgin material; Klimenko et al. (2021) estimates that at the current development rate of EVs, and the current expansion of mining and recycling, demand will surpass currently known global cobalt reserves by 2060. Their research also demonstrates the necessity of a circular economy: if recycling increases along with mine expansion, demand will be met with 55% of reserves in 2100.

The mining of cobalt within the DRC is a bleak reality. An estimated 20% is mined using artisanal practices, some of which result in human rights abuses, including child labor (Amnesty International & Afrewatch, 2016). The workers are exposed to landslides, heavy metals through inhalation, and contaminated water (Amnesty International & Afrewatch, 2016; Tsurukawa et al., 2011). These human rights abuses and poor conditions are a continuation of the cobalt mines grueling history of colonial extraction. The mines were started under Belgian colonization, where they profited off the Congolese mines for 70 years (1912 to 1989), which includes a 29-year period after declaring Congolese independence in 1960 (Tsurukawa et al., 2011).

A new period began with the Sino-Congolese Cooperation Agreement that was finalized in 2008, under which Chinese state-owned companies planned to invest 6 billion USD into the DRCs infrastructure. The International Monetary Fund objected to the original deal amount of 9 billion USD stating the debt load could be detrimental to the DRC. This led to the loan decrease, although the tonnage of materials to the Chinese was not reduced in tandem. Experts argue this is not a win-win deal and may result in the Congolese being yet again exploited by a foreign entity (Ross, 2015).

2.2.2. Nickel

Nickel has recently been deemed a critical material by the US due to the increased demand associated with the growth of LIBs. Class I nickel is required for the NMC cathode production, a high purity nickel representing about half of global supplies. Class II nickel has been in higher demand over the last decade, reducing costs and representing the majority of processing. The increased demand of Class I has resulted in rising costs, although this price increase is not expected to be enough to incentivize the capacity expansion needed to meet forecasted demand (Campagnol et al., 2017).

Indonesia (30%) and the Philippines (16%) are the greatest producers of Nickel. These countries are biologically diverse and included in the 17 mega-diverse countries by the World Conservation Monitoring Centre of the United Nations Environment Program (Iberdrola, 2022). The mining process is extremely harmful to the natural habitat, especially without restoration. In

Indonesia the mining strips the top layer of soil, exposing a subsoil (below 30 meters) that is low in nutrients and unable to support plant growth. The native plants are unable to regrow after the mining is completed, although the land can be brought back to life through soil rehabilitation (van der Ent et al., 2013).

2.2.3. Lithium

While Chile has historically been the largest producer of lithium, Australia surpassed its production in 2014, a year before LIBs became the largest end-user (Ambrose & Kendall, 2020). Since 2015, all lithium exporting countries have increased their production (United States Geological Survey, 2020), and this trend is expected to continue rising with EV sales. Plans of production in the California Salton Sea are underway, earning the region the name “Lithium Valley”. The lithium will be extracted from geothermal brines, a new and low environmental impact technology that has not be demonstrated at industry scale. The large production is expected to meet all of the US demand for lithium, and approximately 50% of global demand. Global demand for lithium is expected to only reach about 50% of lithium supplies, and a shortfall is not forecasted (Klimenko et al., 2021).

Despite the comparably low impacts of lithium that is shown in Table 2.2, the mining practices currently underway, including from hard rock and the evaporation from brines, does not come without damages. One of the largest production sites is located in Salar de Atacama, Chile where water is considered a nonrenewable resource due to immense scarcity. The area is also home to indigenous peoples who are in constant conflict with the industry over water rights (W. Liu & Agusdinata, 2020; Schlosser, 2020). The mining has been found to result in water shortages in local indigenous communities by both a social life cycle assessment (Egbue, 2012) and an interdependency study (W. Liu & Agusdinata, 2020). The three companies that run the operations in this region have been accused of bribery, tax evasion, and price-fixing, yet permits for expanded production continue to be granted (Schlosser, 2020).

2.2.4. Copper

Copper demand is increasing with the expansion of renewable energy, including LIBs, solar photovoltaic panels, and wind turbines. Historical models for copper demand have predicted a shortfall, due to an underestimation of reserves expansion and an overestimation of demand (Hunt et al., 2021). A recent model has forecasted that about 90% of identified resources will be extracted if we are to reach the 2° Celsius climate goal, although this will drop to about 50% of resources if the predicted reserve expansion is met (Seck et al., 2020).

The impacts from copper production overlap with several of the materials discussed. Chile is the largest supplier of copper, and similarly to the case of lithium, there has been conflict over water resources due to the industry's large consumption (Aitken et al., 2016). In addition, copper and cobalt are co-products of the mining process in the DRC and thus result in similar human rights abuses. The processing and refining stage of copper sulfide ores result in high amounts of SO_X emissions, similar to nickel (Table 2.2), although it is reported that some processing plants in the DRC decrease these emissions by capturing the SO_2 to produce sulfuric acid for the hydrometallurgical process (Dai et al., 2019).

2.3. Lithium-ion battery end-of-life processing

When batteries reach the end of their life they can be reused, repurposed, remanufactured, and then recycled. This incremental process is referred to as the waste hierarchy and follows the circular economy model (Richa, Babbitt, & Gaustad, 2017). The recovered materials from recycling can be used in the manufacturing of LIBs, thus displacing virgin material. The final step in the waste hierarchy, recovering the materials through recycling is essential to decreasing impacts and ensuring a large enough supply for future LIB demand.

2.3.1. Reuse, repurposing, and remanufacturing

The reuse, refurbishing, and repurposing of a battery after the use an EV has environmental benefits (Richa, Babbitt, & Gaustad, 2017). It prolongs the use of a product which has already been manufactured, and when repurposed to support renewable generation, it provides carbon abatement via increased supplies of low-carbon electricity generation (Bobba et al., 2018; Casals et al., 2017; Cicconi et al., 2012; Cusenza et al., 2019; Faria et al., 2014; Genikomsakis et al., 2014; Richa, Babbitt, & Gaustad, 2017; Richa et al., 2015; Sathre et al., 2015).

- Reuse: The battery is removed from the EV and then the pack, module, or cell is placed into a new vehicle.
- Refurbished: The battery is removed from the EV, repaired, and then reused.
- Repurposed: The battery is removed from the EV and repurposed to then be used in a stationary storage application (e.g. load leveling, arbitrage, or transmission congestion).

When retired, the battery typically has between 80% (Yang et al., 2018) and 60% (Hall, 2021) capacity left, and while this lower capacity may not be ideal of a car owner, there are many applications which don't require as high of energy density. Repurposing LIBs for these purposes is occurring at industry scale. For example, B2U, a LIB repurposing company, has the largest second life battery energy storage system, at 17 MWhs. This facility is located in California and supports solar generation. Repurposers typically received LIBs through a contract with OEMs and have reported difficulty in precuring batteries due to currently low retirement rates (Hall, 2021). This is expected to become less of an issue while more EVs retire over the future decade.

2.3.2. Recycling

Currently, the recycling rate of EV batteries is unknown, due to the lack of tracking and reporting of battery disposition. While the statistics are unavailable, it is known that EV batteries are entering the recycling stream. Companies such as Redwood Materials, Licycle, and Lithion report their operations are currently recycling batteries from EVs and consumer electronics (Carney, 2021; Lithion Recycling, 2019). There is some conjecture that the majority of LIBs from EVs are being recycled and reused. This assumption stems from recycling and reuse companies stating the inhibiting factor for scaling is the current low supply. The scale of retirement will greatly increase as the first large wave of EV batteries retire in the coming years.

There are three types of lithium-ion battery recycling processes. Two of which occur at industrial scale (hydrometallurgical and pyrometallurgical) and one which is still in the development phase (direct cathode recovery). Each process uses a different method for metal recovery, results in different yields, and produces different final products.

Pyrometallurgical processing has been common in the recycling of electronics for metals recovery. Prior to pyrometallurgical treatment, batteries can be mechanically treated by sorting and crushing, and then subjected to temperatures of 150 to 500 C to remove electrolyte and organic solvent. The pyrometallurgical process consists of heating the LIB to temperatures of 1400 to 1700 C to create a Co–Ni–Cu–Fe alloy of the recovered materials and a slag of the unrecovered materials, including lithium. The alloy produced is a mixture of metals, but can be run through an additional hydrometallurgical process to recover the constituent target materials of cobalt, nickel, and copper (Assefi et al., 2020).

The hydrometallurgical recycling process also requires pre-treatment, which typically consists of discharging, dismantling, or mechanical crushing, and sorting the following from the rest of the materials: active cathode, anode, electrolyte, copper foils, and aluminum foils. Next, the cathode active materials are separated from the aluminum foil by a dissolution process using organic solvents, the binder is removed, and the electrolyte is recovered. The hydrometallurgical process then begins by leaching with inorganic or organic acids to create a solvent containing the materials. The materials cobalt, nickel, manganese, and lithium are then recovered from the solution using solvent extraction, chemical precipitation and/or electrochemical deposition (Yao et al., 2018).

The direct recycling method similarly begins with discharging, mechanical separation, separating of cathode active materials from the aluminum foils, binder removal, and electrolyte recovery. At this point, the direct recycling process differs from hydrometallurgical by recovering the full cathode and performing a relithiation process. The use (cycling) of batteries decreases the lithium within the cathode by up to 60% via reactions with the electrolyte or isolation in the anode. Relithiation processes are still in the research and development stage and several different methods are being researched including thermal, hydrothermal, redox mediator, ionothermal, and electrothermal processes. In addition, Argonne National Lab's ReCell center is currently researching the possibility of upcycling cathodes to different stoichiometry, for example taking an NMC111 cathode and upcycling the cathode to an NMC811 cathode (Gaines et al., 2021).

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3. Methodological foundation

The industrial ecology methods of material flow analysis (MFA) and life cycle assessment (LCA) are used in this research to better inform LIB policy. The waste hierarchy and circular economy frameworks are also incorporated into the work. These methodological foundations are described further in this section.

3.1. Material flow analysis

MFA is the calculation of material stocks and flows through a system. It is a tool which can be used to calculate social metabolism and the boundaries of natural resource use. In turn, policies can be used to control flows and manage natural resources before reaching ecological disaster or the depletion of materials (Hendriks et al., 2000).

MFA has been used to assess the potential for material reserves to meet forecasted EV demand, as well as the need for recycling and repurposing of LIBs in China (Song et al., 2019), Europe (Baars et al., 2021), and globally (Richa et al., 2014). Richa et al. (2014) conducted the first global LIB MFA and analyzed all LIB cathode materials, concluding that future MFAs should include more refined forecasts of EV sales, battery technologies, and lifespans. Some recent studies have developed MFAs that reflect Richa et al.'s recommendations, embedding forecasts of market and technology development, but they do so at a regional rather than global scale (Baars et al., 2021; Song et al., 2019).

3.2. Life cycle assessment

Environmental life cycle assessment (LCA) is the study of the environmental impacts throughout the life cycle of the product, and throughout the supply chains that support the product system (Guinee et al., 2014). The processes include material extraction, manufacturing, transportation, use, recycling, and disposal. The tool can be used to inform policy and decision-making in support of environmental management and sustainable development. Information provided by LCAs can be used to compare similar products and identify the less environmentally harmful option. It can also be used to identify a high polluting hot spot in a product system where reductions may be most effectively targeted (Muralikrishna & Manickam, 2017).

The theory of LCA was originally used to assess the efficiency of energy sources. These analyses assessed the consumption of materials onsite for energy production and then expanded offsite and up the supply chain. The Resource Environmental Profile Analysis in the 1960s expanded beyond energy accounting to assessing the environmental impacts of products life cycle. A few of the products assessed include packaging materials, appliances, automobiles, and housing. Prior to 1990, the studies went by various names until the term "life cycle assessment" was coined (Horne et al., 2009).

The International Organization of Standards (ISO) 14040: 2006 is the most widely cited sources for the principles and framework for LCA (International Organization for Standardization (ISO), 2006; Muralikrishna & Manickam, 2017). There are four distinct stages within the framework:

1. **Goal and Scope:** Defining the product life cycle and the aspects of the life cycle that will be included in the analysis.
2. **Inventory analysis:** Description of the material and energy flows within the product system. The inventories for analysis are determined and gathered.
3. **Impact assessment:** The inventories are translated to the impact categories based on the characterization factors chosen. An example of an impact category is Global Warming Potential, which is characterized by CO_2eq. Normalization and weighting of the impact categories is a voluntary approach which can be used for comparison.

4. **Interpretation:** This step is based on critical review of the impacts, as well as the inventory and scope of analysis chosen.

LCA has been frequently applied to assess the impacts of EVs, LIBs, recycling processes, and repurposing. Lastoskie and Dai (2015) used LCA to demonstrate that the manufacturing of batteries with nickel and cobalt results in high human toxicity, particulate matter formation, freshwater eutrophication, and mineral depletion. They came to this conclusion by comparing the impacts of cathode chemistries, a similar approach to Ambrose and Kendall (2016)

LCA has also been used to identify a route of impact reduction through battery recycling and reuse (Richa, Babbitt, & Gaustad, 2017). An LCA completed by Richa et al. (2017) found environmental benefits were mitigated through reuse, repurposing, and recycling. This study also found that while recycling is a less burdensome process than mining and processing, the impacts differ depending on the recycling process; hydrometallurgy requires 25% less cumulative energy demand than pyrometallurgy.

3.3. Circular economy and the waste hierarchy

Industrial ecology has long posited that our modern, industrialized economies operate on the basis of linear flows of energy, materials and waste, and that a system of linear flows cannot be sustained. The idea of a circular economy from the is to transform linear flows of resources and waste into circular flows; in other words, a transition from the linear cradle to grave model to a circular cradle-to-cradle. In a circular economy, instead of disposal, materials are recycled and recovered to then be used in then use again in manufacturing. This circularity of resources reduces environmental and social impacts associated with extraction and disposal.

The ideal of circularity has become more prevalent in literature and policy as climate change impacts and resource security becomes a global issue. Circularity principles are the drivers of recycling materials used in everyday products, such as plastics, with the aim to decrease the environmental impacts and increase material efficiency of society (Corona et al., 2019). In addition, renewable energy technologies such as solar photovoltaic panels, wind turbines, and LIBs, contain critical materials. Securing a supply of these critical materials is essential to clean energy development and energy security. Their recovery through recycling can decrease environmental impacts and provide a local supply of materials, mitigating dependencies on international supply chains subject to geopolitical and market uncertainty (Richa, Babbitt, & Gaustad, 2017; The White House, 2021)

The waste hierarchy is a key feature of circular economy frameworks, such as those developed by the European Union (EU) and United States Environmental Protection Agency (US EPA). The hierarchy provides an order of preference for managing waste: prevent, reduce, reuse, recycle, recover, and finally dispose (Figure 3.1) (Environmental Protection Agency, 2021; European Commission, n.d.).

The European Commission has proposed battery policy focused on creating a robust and connected recycling and manufacturing battery economy in Europe. One feature of the proposed battery policy is recognizing the importance of connecting waste management processes, namely recycling, with manufacturing infrastructure that can use recovered materials. Simply recycling batteries and recovering materials without attendant manufacturing infrastructure does not fundamentally address dependency on international supply chains for battery production. While battery circularity policy has not been proposed or adopted in the US, the Biden Administration has recently conducted several studies including a report on the lithium-ion battery supply chain

(The White House, 2021) and has also dedicated funds in the 2021 Infrastructure, Investment, and Jobs Act towards battery recycling and reuse research and development (Infrastructure Investment and Jobs Act, 2021).

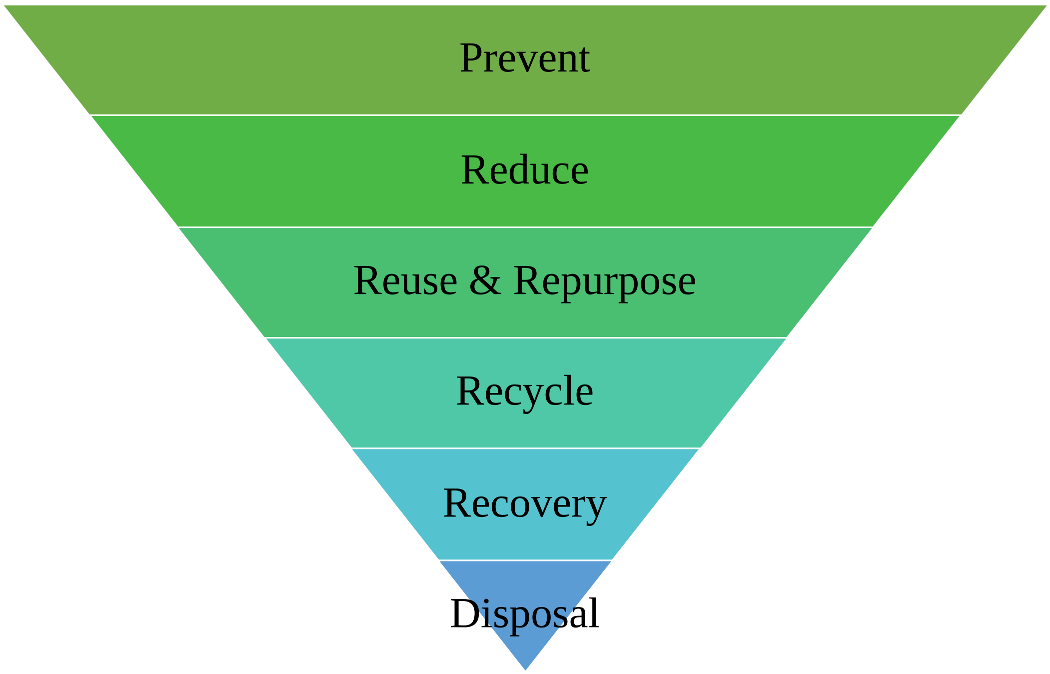

Figure 3.1: The waste hierarchy

3.4. References

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4. Lithium-ion battery policy, current happenings, and perspective

4.1. Scope and Purpose

This section reviews global lithium-ion battery policy with an emphasis on the US approach. The information provided informs the discussion sections in the quantitative research sections.

4.2. Lithium-ion battery policy

The LIB sphere is ripe with innovation, policy development, and media attention. The technology provides a path to transport electrification, which is seemingly achievable with the coordination of the public and private sector. However, hurdles remain for widespread electrification via LIBs including resource security, corruption and harm in the supply chain, and the need for end-of-life management infrastructure and policy that anticipate the enormous flow of future retired batteries (International Energy Agency, 2020). These issues are not new to the transportation sector; internal combustion engine vehicles also have a complex international supply chain for materials, but more importantly for the petroleum, that are subject to disruption. In addition, the industry which deals with the vehicle end-of-life processing is robust and has handled technology changes over time. In addition, the motivation for transport electrification is to mitigate climate concerns, therefore it is morally consistent, and in the best interest of the movement, to lower the upstream and life cycle impacts of the replacement technology.

Governments around the world are developing LIB policy, including the Republic of China (China), the European Union (EU), and the United States (US). China has used investments and regulations to increase their manufacturing capacity, while the EU has proposed strict manufacturing and circularity requirements (Melin et al., 2021).

The US has yet to pass LIB regulations, but federal action includes research and development funding, indicating a market-based approach. To date, the US has invested in research and development of domestic recycling and reuse, in addition to efforts to increase domestic mining capacity. These efforts have emphasized the need for energy security and decreased LIB impacts. While environmental and ethical sourcing concerns have been expressed, policy proposals do not consist of regulations addressing these issues. This indicates the batteries sold in the US may not be held to the high standard that is required to create a sustainable and ethical LIB supply chain.

The aim of increased battery recycling, repurposing, and energy security can be seen as the commonality between global LIB policy. Although, as briefly demonstrated, the policy approach and environmental and social standards vary between the regions, suggesting the future markets and their impacts will also likely vary.

This section will first provide an overview of the Chinese and EU approach to LIB regulation and industry development, and then review, compare, and evaluate the US policy approach.

4.2.1. China

China is the leader in LIB material refining, manufacturing, and recycling. Their approach has been to secure and develop a supply chain, including recycling infrastructure for material recovery (Holslag, 2021). The Chinese government has direct control over the industry and has provided subsidies and investments for LIB development. This high involvement of the government has been a catalyst for fast growth resulting in majority control over the global LIB industry.

LIB circularity policies in China began with the 2017 Promotion Plan for Extended Producer Responsibility Systems. This policy requires producers to be responsible for recycling LIBs. To-date, additional policies have been enacted including battery labeling, design for recycling, and traceability. In conjunction with circularity requirements, funding for recycling infrastructure and pilot plants has been dispersed (Li et al., 2021).

In order to secure a critical material supply chain, China has acquired contracts for material extraction from many countries in Africa. They have engaged in what has been referred to as "infrastructure for mineral" deals, i.e. the development of infrastructure such as roads, schools, hospitals and factories, in exchange for mineral rights (Gulley et al., 2018; Rapanyane, 2022). The approach of seeking dominance in the minerals market is reported to have been spurred by fear of resource insecurity. Deals made with the Democratic Republic of Congo has led to China owning 20-30% of global cobalt supplies and 35-50% of global cobalt refining and production (Holslag, 2021). This accrual of a crucial LIB material supply chain is now seen as a threat to other regions' energy security.

The deals made to acquire minerals from Africa have been criticized as being inequitable and contributing to the resource curse of the mineral owning country. The resource curse describes a country that is rich in resources but unable to fully benefit from their wealth and provide the appropriate public welfare to their people. Economies experiencing this condition heavily rely on the extraction of resources without a diversified income stream. The root of this curse in post-colonial countries stems from the colonial model of extraction, which did not include investments in infrastructure, education, or expansion to other markets. In addition, the instability caused by colonialism has, in many cases, led to corruption within the current day governments. Injustice and harm caused by this model is continued by the further extraction by many actors (Rapanyane, 2022).

4.2.2. European Union

The EU is taking a different approach than China to the LIB market. While both regions emphasize the need to recycle batteries, the EU is also focused on mitigating impacts of production. Although, the EU has not invested as heavily in increasing their market share of virgin materials production such as China has.

Current policy development builds on the 2006 Battery Act, which includes extended producer responsibility (EPR) and required collection of retired batteries. This 2006 Act failed to address challenges related to LIBs, deficits that are clearly outlined in a 2019 study by the European Commission (European Commission, 2019). They have since proposed replacing the Battery Act with policy attempting to decouple growth from resource use (European Commission, 2020). This policy is unofficially called the "EU Sustainable Battery Policy" and is not only focused on the battery end-of-life, but also on creating a low impact and circular supply chain. The regulation incentivizes sustainable manufacturing and recycling within the EU, while also setting strict requirements regulating upstream impacts, manufacturing, reuse, refurbishing, repurposing, and recycling at the end-of-life (European Commission, 2020; Melin et al., 2021).

- *Upstream requirements* for the supply chain include mandated third-party due diligence in line with the OECD Due Diligence Guidance. This guidance lays out routes to avoid impacting communities, impacting the environment, and contributing to human rights violations and bribery. These requirements are criticized as having several shortcomings which undermine the effectiveness of the policy (Amnesty International, 2022). Material tracing is part of the Battery Passport, an online system which would track many aspects of the battery life cycle, including where the materials are sourced.

- GHG constraints are applied to *manufacturing and material processing*, including the emissions from recycling and recovering materials. Batteries are required to be manufactured with portions of recycled lithium, manganese, and cobalt. These recycled content rates increase over time. In addition, batteries are required to be designed with disassembly in mind. Design for disassembly increases efficiency and safety for those dealing with retired EVs.
- Once a battery has reached the end of its life, there are several requirements which will enable the *reuse, refurbishing, and repurposing* of the battery. These include 1) the ability to transfer extended producer responsibility to the repurposer, and 2) requiring access to battery information. The access to information will remove expensive barriers at the battery's end-of-life. The policy will require a label, including the chemistry of the battery, a QR code with additional information, and the development of a battery passport which provides the ability to access state-of-health information after the battery is removed from the vehicle.
- *End-of-life* requirements include a required collection rate, recycling rate, material recovery rate, and reporting.

In summary, the EU Sustainable Battery Policy includes strict regulations for sourcing, manufacturing, and recycling. The regulation aims to ensure clean and equitable development of the industry. Concern has been expressed that the EU policy overregulates fast-changing technology, which could result in pushing up the cost of EVs, and depressing EV uptake. In addition, the regulation does not require recycling or sourcing of reclaimed materials from the EU. Considering China has a much more robust LIB recycling market, they are well positioned to fill this need, and it is feared the EU will not develop a competitive market of their own, but rather just be a primary customer for secondary materials (Melin et al., 2021).

4.2.3. United States, Federal

The US has taken a less cohesive approach than the EU and China, with various agencies at the state and federal level working to address this issue. Currently, there are no circularity or due diligence requirements in place that are specific to LIBs or their critical materials, and only a handful of US states have implemented landfill bans (International Energy Agency, 2020; Melin et al., 2021). California, and more recently Washington State, are in the process of developing their own policy, although none has yet to be enacted (*Assembly Bill No. 2832*, 2018).

Development of domestic supply chains and recycling infrastructure

Federally, action is focused on funding the research and development of a recycling and reuse industry, while also taking steps to increase national production of critical materials. Three prominent research and development funding sources include:

- The 2022 Infrastructure Bill, which dedicates funds towards securing a domestic supply chain of critical materials, recycling, and reuse, while also initiating the creation of a Task Force focused on developing an extended producer responsibility (EPR) battery recycling framework (Infrastructure Investment and Jobs Act, 2021).
- The US Department of Energy Vehicle Technologies Office's funding of ReCell Center, which is dedicated to advancing battery recycling (Gaines et al., 2021).
- The Department of Energy's award to National Labs and Universities for research focused on securing a domestic supply chain of critical materials (Department of Energy, 2021).

Presidential actions focused on LIBs include a recent report by the Biden Administration which outlines the supply risk of LIB materials and potential solutions. Solutions in this report include: increased mining, national supply chain development, and recycling (The White House, 2021). Following this report, in March of 2022, President Biden signed the Defense Production Act (DPA) Section 303. This Act is intended to bolster domestic production of critical materials required for large capacity batteries (The White House, 2022).

DPA enables the public and private sector to work together to provide essential goods for National Defense. Powers of DPA include the ability of the federal government to 1) require businesses to prioritize government orders, 2) provide presidential loans to businesses for increased production, 3) install equipment in government buildings, and 4) if authorized, the ability for companies to coordinate together, which would usually violate anti-trust laws (Lawson & Rhee, 2020; Siripurapu, 2021).

The Presidential announcement of DPA states that mining should be done "… with strong environmental, sustainability, safety, labor, Tribal consultation, and impacted community engagement standards…" (The White House, 2022). These guidelines are not a requirement, and without set regulations, building up domestic supply chains holds priority. This act has been criticized by public interest groups because DPA provides the ability to expedite and use government resources for supply chain development, without additional policy and requirements for sustainability and equity in the mining process. Mining standards are beginning to be addressed through the development of Interagency Working Group (IWG) on Federal hardrock mining laws, regulations, and permitting. Topics to be covered by the IWG include, but are not limited to, environmental impacts of mining, engagement with Native Tribes and local communities, and assessing the relevancy of the General Mining Law of 1872 (United States Department of the Interior, 2022).

Review of the General Mining Law of 1872 has been requested by advocates and lawmakers because of its low requirements of industry and little environmental and community protections (Higginbotham, 2021). Under this law, hardrock mining approval is required on federal land in 19 of the 50 US states if companies can prove it to be economically viable. In addition, industries are not required to provide royalties to the US for the materials extracted (Disbrow-Monz, 2022).

Mining in the US has devastated Native American communities and created Superfund sites which the US Environmental Production Agency is tasked with cleaning up using taxpayer money. Without strict mining regulations, sustainability and equity is likely not a priority in mine development, and therefore the historical process of extractive industries that undermine communities and cause environmental destruction has the potential to continue (Woody et al., 2010).

The Mining Law of 1872 has recently been under review by legislatures; the Senate Committee on Energy and Natural Resources held a hearing on the law, and two bills requiring stricter mining standards in the US have been introduced in the 2022 legislative session (H.R. 7580 and S. 4083). H.R. 7580 would make two prominent changes: 1) it would restrict mining from sacred sites, critical habitats, areas of critical environmental concern, National Conservations Systems, and areas designated National Wild and Scenic Rivers System; and 2) require royalties be paid from the materials mined. While there have been bills introduced in the past, H.R. 7580 has received more attention this legislative session due to federal focus on the clean energy transition and the recent Presidential determination of DPA (H. R. 7580, 2022).

The Inflation Reduction Act of 2022 will also impact critical material supply chains. Eligibility for the EV tax credit included in the Act hinges on 1) a portion of the constituent materials (mined, processed, or recycled) be sourced from the US or free trade agreement (FTA) countries, 2) a percentage of the battery components manufactured or assembled in North America, 3) final assembly of the battery occur in North America, and 4) the battery or critical materials cannot have been extracted or processed by a foreign entity of concern, including China (*Inflation Reduction Act,* 2022). Due to the majority of mining, processing, and manufacturing of materials and components ocuring outside of North America and FTA countries, and specifically the supply chain dominance of China, these requirements are not likely to be met in the near-term by automakers (except potentially by Tesla). While the requirements may incentivize national supply chain development, there is potential for it to inflate material costs and negatively impact EV uptake. The increased costs are likely due to the near-term start date of requirements, including critical material sourcing beginning at 40% in 2023, and the ban on materials sources from China beginning in 2024. This does not provide adequate lead time for mine or processing infrastructure development, which has a timeline of four to 12 years (The White House, 2021).

Previous to this Act, several House and Senate Bills were introduced (i.e. S.1918 — 117th Congress) which also aimed at incentivizing a recycling industry and building national supply chains, although none passed (*Battery and Critical Mineral Recycling Act of 2021,* 2021).

Overall, Federal LIB policy has focused on the development of a material supply chain and a recycling industry, but does not include any additional recycling, circularity, environmental, or social regulations for battery manufacturing, recycling, or reuse. This is vastly different than the EU proposal of regulations aimed at decoupling material extraction and emissions from battery production and end-of-life processing. The US approach follows more closely to the Chinese goal of bolstering resource security and domestic processing with high dependency on investments to stimulate the recycling and reuse market. Federally, the US has indicated an EPR program may be considered by recently dedicating funds to convening a task force, which must deliver EPR recommendations within a year. In addition to the work of this task force, there is the potential of stricter federal mining requirements emerge from the IWG (Infrastructure Investment and Jobs Act, 2021).

Global supply chain impacts

Even with increased domestic mining, the US will likely need to continue imports of critical materials (The White House, 2021). As demonstrated in section 2.2, there are high environmental and social impacts caused by mining globally, and for batteries to be produced conflict free, an approach to mitigate harm of imported supplies is necessary.

Currently, US laws do prohibit practices that are seen in the global supply chain, such as the human rights violation of child labor and the act of bribery. Section 307 of the Tariff Act of 1930 (19 U.S.C. §1307) prohibits the importing of goods made with slavery or child labor. Despite reputable research and reporting of illegal activity throughout supply chains (Amnesty International & Afrewatch, 2016; Sovacool, 2021), this law is rarely used by the US to block imports (Congressional Resource Services, 2022).

Some of these critical materials are also associated with bribing of foreign government officials (Sovacool, 2019), a practice restricted by the US Foreign Corrupt Practices Act of 1977. The US has recently cracked down on Glencore, the largest producer of cobalt (Resource Matters, 2021); in March of 2022 they pled guilty to corruption charges resulting in a fee of 1.1 billion US dollars (Helman, 2022). Glencore is a cobalt supplier for many companies, including BMW, CATL, and Volkswagen, to name a few. US policy requires reporting of supply chains according to the Dodd-Frank Act 2010, section 1502, and most of the companies sourcing from Glencore state they have an internal process for assessing supply chain due diligence. Despite these processes, the bribery claims against Glencore that have been made for years did not deter the companies from continuing to source from Glencore (Resource Matters, 2021).

The lawsuit against Glencore is hopefully a signal that the US will begin pursuing companies found to implement corrupt tactics to secure resources. Unfortunately, current conditions and historical precedence suggest that framework in place today is insufficient for deterrence and continues to allow corrupt practices.

4.2.4. California

California,, the leader in EV and climate policy in the US, has been a large supporter of the development of lithium extraction near the Salton Sea and is preparing to enact additional regulations at the state level to manage end-of-life EV LIBs. Currently, there are no federal requirements for recycling or reusing LIBs. Assembly Bill 2832 (AB 2832) required the creation of an Advisory Group tasked with developing EV recycling policy for California (*Assembly Bill No. 2832*, 2018). The Advisory Group was made up of members from the automotive and battery industry (6), waste management industry (5), public interest organizations (3), and government agency (5). In March of 2022, a final report was released to the California Legislature which provides policy recommendations by the group. These recommendations are based on a final vote, which the government agency representatives recused themselves from. Thus, the automotive and battery industry represents 40% of voting members, waste management industry represents 33%, and public interest organizations represent 20% (Kendall et al., 2022).

The report focused on defining a responsible party at the battery end-of-life, and recommends the legislature require automakers to ensure the batteries are recycled, referred to as EPR. This is a large step towards mandating recycling of LIBs in the US and it is important to note that a version of EPR was supported by the automotive industry members. This was a policy position that evolved over the course of the two years and is likely the result of several factors.

First, the market for materials and the recycling industry is rapidly developing (Carney, 2021; Li-Cycle, 2020; PR Newswire, 2022). The recycling sector announcements of planned facility capacity, successful recovering of battery grade materials, and apparent economic success suggests a thriving future for the industry. These market signals suggest that in the future, batteries may be an asset instead of a burden to those responsible for the batteries at their end-of-life. Another positive market signal for LIB end-of-life is the increase of virgin material prices, resulting in a higher breakeven price for recycling (LME, 2022).

Automakers' commitment to recycling has been further demonstrated with new partnerships; Ford and Volvo have recently partnered with Redwood Materials, an LIB recycler. They are jointly funding a program which will collect and recycle any EV LIBs that are retired within California. This 'learn by doing' approach will hopefully educate actors on the location of batteries after retirement, the types of batteries retiring, and optimal reverse logistics pathways (Korosec, 2022).

An EPR program can be structured in various fashions, and it is important to note that the version of EPR which was proposed by industry, and received the most support by the Advisory Group, places the automobile manufacturer as the party responsible for ensuring the battery is recycled, unless the EV or battery has been acquired by an auto dismantler (or other user such as a repurposer), at which point it becomes the responsibility of the auto dismantler or other user. It appears this was favored by industry because they are operating under the assumption that if a dismantler has acquired an LIB and then does not want it, then there is not an economic value to the battery.

While an EPR is beneficial to ensuring batteries are recycled, this caveat may lead to orphaned batteries due to the inability to identify which dismantler owns or removed the battery. If this policy was to be adopted, there would likely need to be a fund and government program to cover the recycling of the orphaned batteries.

In addition to EPR, the following policies gained majority support from the Advisory Group:

- *Increased data transparency* through labeling, a QR code, and access to state of health metrics
- Incentives and reduced regulatory burden to *support the recycling and reuse industry*
- *R*esearch, training, and reduced regulatory burden for the *transportation and reverse logistics of retired batteries*

These policies address crucial efficiency, safety, and cost barriers of recycling, and have the potential to greatly increase recycling rates.

Additional policies, categorized as circular economy principles, did not receive majority support by the Advisory Group. These policies include recycled content standards, recycling efficiency thresholds, design for recycling, third-party recycling verification, and required metric reporting. Supporting members of these policies mostly consisted of the public advocacy organizations. The lack of support from other members stemmed from the desire to avoid blocking industry development, the potential to increase EV costs, and the fear of setting bad standards and thresholds due to a lack of information. This echoes the federal approach of holding off from the command-and-control requirements and focusing more on market incentives and a form of EPR.

4.3. Challenges of the US approach

The exclusion of supply chain and circularity requirements in US policy, both federally and at the state level, has the potential to lead to a US LIB industry which is unsustainable and unethically sourced. While federal policy states the intention of enhancing material security in a sustainable and ethical way, the policies lack binding requirements. The expedition of US mining without requirements is therefore concerning because it has the potential to continue damaging extraction and refining practices. In addition to ethical development of mining in the US, the lack of focus on ethical supply chains from globally sourced materials is an area with needed policy and focus.

Regulations for recycling, circularity, and supply chain emissions are also lacking from the US approach. While the AB 2832 report proposes implementing recycling regulations, the missing circularity and environmental requirements has the potential to result in lower recycling rates and a higher impact supply chain than other regions. Design for recycling, recycled content standards, collection rates, recycling efficiency, and GHG emission requirements, are a few of the regulations that the AB 2832 Advisory Group did not have majority vote for. In addition, the Advisory did not propose policy which restricts the exportation of used EVs or used batteries. This lack of regulation could result in the exportation and dumping of waste and the loss of potentially recoverable critical materials.

4.4. Next steps

The forecasted rapid demand in EV battery materials reflects a hopeful future where transportation is electrified, and climate change goals are reached. This future requires negotiating the complexity of necessary and urgent transport decarbonization, and the needed reduction of the enabling technology's supply chain impacts. So how can climate goals be met while also mitigating the impacts of material usage?

Solutions require a multifaceted approach of both recycling and mineral extraction. A swath of policies is needed which specifically address the following priorities: a sustainable and ethical supply chain; sustainable manufacturing and recycling; domestic supply chain development; efficient and safe material circularity; and recycling of LIBs at the end-of-life. Possible policies are listed in Table 4.1.

Table 4.1: Possible lithium-ion battery regulations which could increase the sustainability, equity, and circularity of the industry.

Goal	Policies to enable achieving goal
A sustainable and ethical LIB supply chain	• Increased due diligence requirements with government follow through for bad actors (i.e., the OECD Due Diligence Guidance) • Repeal of the 1872 Mining Act and replacement with policy that protects native lands, prioritizes environmental protection, and requires the payment of royalties • Free, Prior, and Informed Consent (FPIC) by affected communities prior to mining • Material and supply chain tracing (i.e., the battery passport)
Sustainable manufacturing and recycling of LIBs	• GHG emission standards for recycling and manufacturing • Recycled content standards • Recycling efficiency standards

Domestic LIB supply chain development	• Regional recycling or sourcing requirements
Efficient and safe LIB material circularity	• Design for recycling and disassembly • Access to battery state of health • Battery labeling
Recycling of LIBs at end-of-life	• Extended producer responsibility

4.5. Conclusion

EV expansion is faced with various material related issues that will need to be met by a multifaceted approach. As this section lays out, different policy approaches have been used by governments around the world to secure resources, increase circularity, and reduce impacts. The US is beginning to actively address these issues through policy, which has been focused on increasing domestic mining, manufacturing, and recycling. While industry development is vital to EV expansion and meeting climate goals, it is essential to ensure, through policy, that the materials are sourced ethically, recovered materials are being used in LIB manufacturing, the batteries are recycled, and this circular supply chain has low environmental and social impacts.

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5. Circularity Potential of Lithium-ion Battery Materials in Electric Vehicles

5.1. Scope and purpose

To effectively prepare for the management of batteries at their end-of-life, the kWhs of EVs retiring, the material of batteries, and the timing of retirement, must be forecasted. This analysis demonstrates the circularity potential of materials, as well as creates a model that can be further developed to advise policy in Section 6. The model is used to estimate the global retirement and demand of pack-level lithium-ion battery materials for EVs until 2040 and the percentage of potential circularity, parsing results by the following regions: US, Europe, China, and the RoW. Scenario analysis is used to demonstrate the impact of changing cathode chemistries, various EV sales forecasts, and the lifespan of second-life batteries on the circularity potential of LIB materials.

This section is adapted from the following publication:

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5.2. Introduction

EVs are still a small part of the on-road vehicle fleet, although global EV sales have increased drastically, from around seven thousand in 2010 to more than two million in 2019. China constitutes the largest EV market, totaling 53% of sales in 2019, followed by the European

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Union (EU) at 26%; the United States (US) at 14%; and the rest of the world (RoW) at 7% (EV Volumes, 2020). Modern EVs are powered by large format lithium-ion batteries (LIBs), a key enabling technology for EVs, as well as stationary energy storage applications (Malhotra et al., 2016).

Circular economy strategies have the potential to reduce demand for primary material (Gaines, 2018; Mathieux et al., 2017; Richa, Babbitt, & Gaustad, 2017), and their capacity to do so hinges on the demand, use, and retirement patterns of EV batteries over time, as well as evolutions in LIB chemistry.

Cathode chemistries are defined and differentiated by the transition metals that are combined with lithium in the cathode. Today, the most common EV LIB cathode chemistries are lithium nickel manganese cobalt oxide (NMC 111, NMC 523, NMC 622, and NMC 811); lithium nickel cobalt aluminum oxide (NCA); lithium cobalt oxide (LCO); lithium iron phosphate (LFP); and lithium manganese oxide (LMO). Recent developments in cathode chemistry have focused on increasing density and reducing material costs, specifically by transitioning to cathodes with lower cobalt content (Schmuch et al., 2018). Cobalt has historically been the most widespread LIB transition metal (Zeng et al., 2019), but it is the most expensive and has a high human and environmental cost (Amnesty International & Afrewatch, 2016). As a result, battery producers have innovated chemistries with other transition metals, and low-cobalt cathodes are expected to increase their market share in the future (Bloomberg New Energy Finance (BNEF), 2019). Not only has cobalt use decreased, but in part enabled by cost reductions, battery size has been increasing in new EVs to deliver longer driving range and higher performance (Ambrose et al., 2020). These two trends are the primary determinants of cathode material requirements for past, present, and future EVs.

Electric vehicle production is also expected to impact future demand for graphite, which is the most common anode material. Like other aspects of battery design, anode technology is developing at a rapid pace; for example, silicon-based anodes are an alternative that may improve volumetric energy density. This analysis is limited to changing cathode materials and does not consider the effect of developments in anode chemistry, but this is an important topic for future research.

5.3. Literature review

A number of previous studies have examined LIBs or related material systems using MFA to assess resource security, infrastructure needs for end-of-life management, circularity, or environmental impacts. Many MFAs have focused on flows of lithium used in LIBs (Ambrose & Kendall, 2020; Lu et al., 2017; Sun et al., 2017; Ziemann et al., 2012, 2018), while others include additional cathode materials such as cobalt and nickel (Asari & Sakai, 2013; Chang et al., 2009; Pehlken et al., 2017; Richa et al., 2014; Song et al., 2019). Criticality has been used as a justification for focusing only on certain materials (E. A. Olivetti et al., 2017; Pehlken et al., 2017; Song et al., 2019; Zeng et al., 2015; Ziemann et al., 2012), and in particular lithium and cobalt. While supply risk and economics are important motivators for circularity, they do not include other important factors such as the social and environmental impacts of production and refining of materials (Crowson, 2011; Graedel et al., 2012, 2015; Huijing, 2018; Olivetti et al., 2015). For example, nickel is not considered critical, but its production has severe local air pollution effects (Dai et al., 2019). Capturing these broader impacts is a rationale for examining all cathode materials, regardless of their criticality.

Among the more comprehensive studies, Richa et al. (2014) conducted the first global MFA analyzing all LIB cathode materials and concluded that future MFAs should include more refined forecasts of EV sales, battery technologies, and lifespans. Some recent studies have developed MFAs that reflect Richa et al.'s recommendations, embedding forecasts of market and technology development, but they do so at regional rather than global scales. For example, Song et al. (2019) conducted an MFA of all cathode material in China using one cathode chemistry forecast with NMC, NCA, and LFP as the dominant chemistries. Baars et al. (2021) focused on the European Union including three cathode chemistry scenarios that alternate between chemistry variations of NMC 622, NMC 811, NCA, and an alternative chemistry without nickel or cobalt. Both studies concluded that recycling can only provide a fraction of regional material demand and that current recycling infrastructure must be scaled to support this retired supply.

5.4. Gap in literature

This study responds to the gaps identified in previous work, using prospective MFA to examine material demand and retirement of cobalt, lithium, nickel, manganese, aluminum, copper, and graphite at the pack-level, for the global EV market, but at a regional scale (China, US, Europe, and RoW). It then estimates the impact of changing cathode materials on potential circularity through 2040. The MFA includes forecasts of EV sales; estimates of expected in-use lifetime; and evolving battery chemistries, all modeled by region and dynamically over time.

Given the uncertainty of forecasts for markets and technologies, we use an ensemble of scenarios to explore future EV sales and cathode chemistry market shares. Model results estimate the amount of material demanded and retired in each region on a yearly basis and provide insights into the potential for recovered material to meet future regional demand (i.e., circularity).

Although the material scope is pack-level, this study focuses on the cathode materials cobalt, lithium, nickel, and manganese because they are of greatest concern for recycling due to their high cost, environmentally intensive production, and material criticality. The material flow results for the other metals are not presented but have been included in Figure A of Appendix A.

In addition, the GWhs of retired battery capacity available for reuse in second-life applications are reported for each region, which is a possible alternative to recycling directly after use in an EV. Due to the likely prospect of second-use applications, the impact that a prolonged in-use lifetime will have on global circularity is also demonstrated.

5.5. Materials and Methods

The MFA framework developed in this study models demand for new cathode material over time, LIB lifetime in an EV (and thus the timing of future retirements), and the quantity of recoverable material based on idealized recycling rates. Among these models, determining future cathode material demand for EV batteries is the most complicated. It requires modeling and estimation of (i) future EV battery sales, (ii) the capacity of batteries in sold vehicles, and (iii) the cathode material composition. The MFA is built on country specific EV and PHEV sales and manufacturing data, national and regional projections for future sales, regional market shares of different cathode chemistries, and trends in cathode chemistry adoption. Future EV sales and changes to cathode chemistry are examined through scenario analysis (scenarios S1 through S4 for EV sales, and scenarios C1 through C6 for cathode chemistry changes).

5.5.1. Modeling Framework

The MFA model examines material demand, in-use stocks, and retirement using an annual time step. Figure 5.1 describes the key components of the demand model and illustrates the scenario ensemble, which generates a factorial of 96 potential results for each cathode material ® considered in any given year (*t*), as a function of the region (*r*), the EV sales scenario (S1-S4), and the cathode chemistry mix scenario (C1-C6).

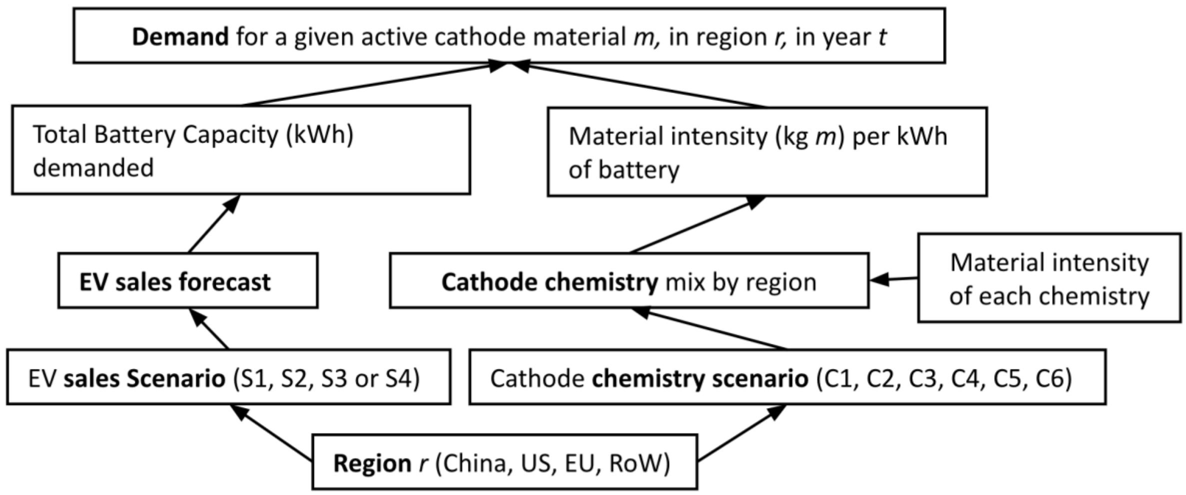

Figure 5.1: LIB Cathode MFA Demand Model

The retirement model generates an estimate of recoverable material in metric tons for each cathode material *c* (Figure 5.2a), and total retired capacity in GWh (Figure 5.2b) in each region for any year t.

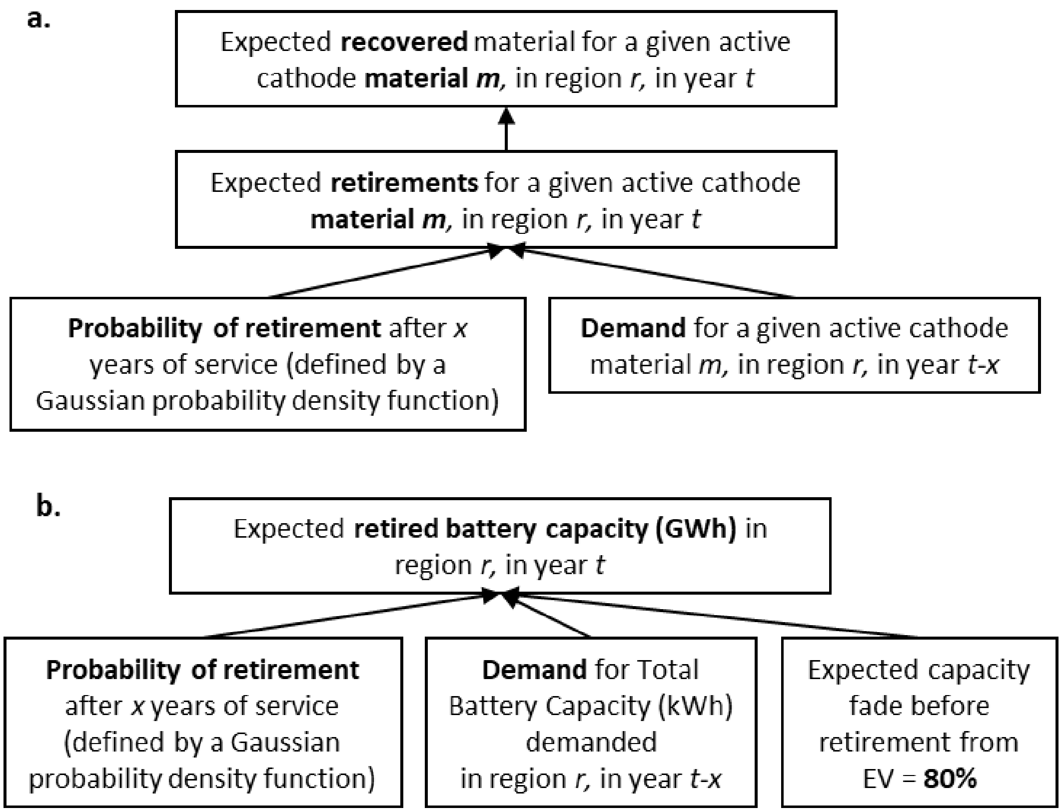

Figure 5.2: LIB Cathode MFA Retirement Model for a) EV LIB cathode materials, and b) EV LIB energy storage capacity.

The potential circularity of a given cathode material *c* in year *t* and region r is calculated by determining the difference between the demand in a given year and expected retirements:

$$Circularity_{c,t,r} = Demand_{c,t,r} - Recoverable_{c,t,r} \qquad \text{Eq. 5.1}$$

The following subsections describe the data, models, and key assumptions that comprise the MFA model.

5.5.2. Regional EV Sales Forecast

Prior to 2016, Europe and the US were the largest consumer of EV's, with sales increasing gradually. Sales in China spiked in 2015, and from 2017 on has comprised more than 50% of the market. The RoW has historically been a small portion of the global EV market, with only about 7% of global EV sales in 2018; however, it is also a large potential market for EV sales growth, especially as EV costs decrease.

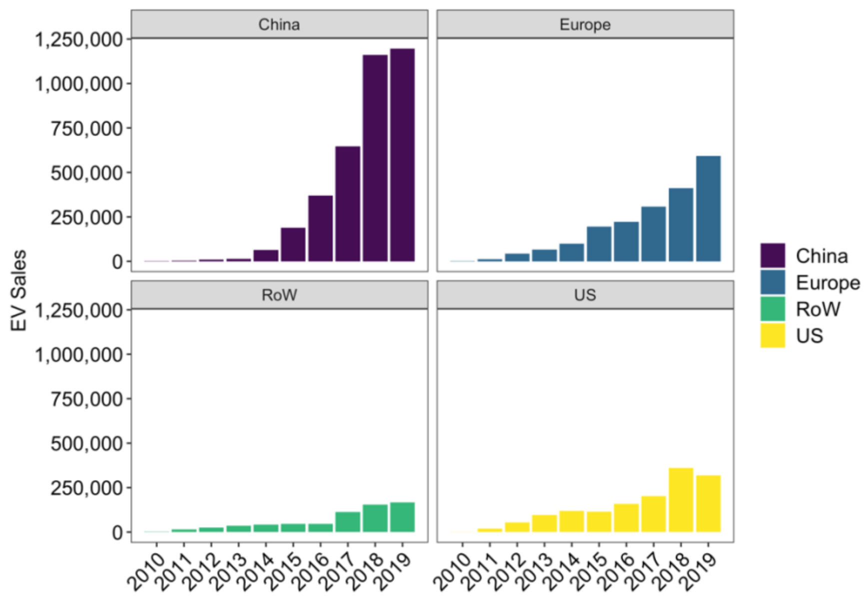

Figure 5.3: Historical EV sales per year until 2019. Reported by EV Volumes.

Due to the uncertainty of future EV sales in all regions, four EV sales scenarios are used for the period of 2020 to 2040 in China, Europe, US, and the RoW (Figure 5.4). Two are based on technology diffusion models developed as part of this research, and two are based on previously estimated forecasts:

- S1: Diffusion Model with policy-based targets - 2040 market share for EVs defined by policy targets.

- S2: Diffusion Model with market-data only - 2040 market share determined through regression analysis of historical sales.

- S3: IEA 2020 GEVO Mobility Model (MoMo) forecast(International Energy Agency (IEA), 2020) - based on the 30 @ 30 scenario where all countries commit to achieving 30% EV sales in 2030.

- S4: BNEF 2019 forecast (Bloomberg New Energy Finance, 2020)- based on an expected market trajectory for EVs.

S1 and S2 forecasts were produced using the Bass diffusion model (Equation 5.2). Bass diffusion models result in a characteristic S-shaped adoption curve, and split adopters of a new technology into innovators and imitators, limited by an ultimate number of potential adopters (Bass, 1969). The Bass model is specified by a coefficient of innovation *p*, a coefficient of imitation *q,* and the number of ultimate adopters or the potential market share of a technology m. Results show the sales per year *s(t)* and the cumulative sales over years *S(t)* (Bass, 1994).

$$s(t) = pm + (q - p)S(t)\left(\frac{q}{m}\right)S(t)^2 \qquad \text{Eq. 5.2}$$

Total light duty vehicle sales data were obtained for the same years (Bloomberg New Energy Finance, 2019), and used to determine the EV market share for each region (Table A in Appendix A). The resulting EV and PHEV sales as a percent of all historical light duty vehicle sales data was used to estimate the coefficients using least squares regression (Equation 5.3).

$$s(t) = \beta_0 + \beta_1 S(t) + \beta_2 S(t)^2 \qquad \text{Eq. 5.3}$$

The *m* value for Equation 5.3 is calculated by Equation 5.4 for S2, while S1 takes the policy targets of each region as the *m* value. The *p* and *q* values are then calculated using Equations 5.5-5.6. The resulting *p*, q, and *m* coefficient estimates are shown in Table 5.1. For each region in S1 and S2, EV sales were then forecast through 2040 based on expected sales of light duty vehicles (Bloomberg New Energy Finance, 2019).

$$m = \frac{-\beta_1 \pm \sqrt{{\beta_1}^2 - 4\beta_0\beta_2}}{2\beta_1} \qquad \text{Eq. 5.4}$$

$$p = \frac{\beta_0}{m} \qquad \text{Eq. 5.5}$$

$$q = p + \beta_2 \qquad \text{Eq. 5.6}$$

Table 5.1: The coefficient values used to forecast EV sales using the Bass diffusion model in the policy-based (S1) and market- based (S2) scenario.

Scenario	Coefficient	Europe	China	US	RoW
Policy-based (S1)	p	0.0013	0.0002	0.0041	0.0019
	q	0.3718	0.6676	0.3670	0.4116
	m	0.8000	0.6000	0.6000	0.5000
Market-based (S2)	p	0.0013	0.0002	0.0041	0.0019
	q	0.3718	0.6676	0.3670	0.4116
	m	0.6568	0.7324	0.2436	0.0883

Scenario S3 is based on the International Energy Administration (IEA) model MoMo. The MoMo forecast depicts a scenario where all countries achieve 30% EV sales for new vehicles in 2030 (30@30). Scenario S4 uses a market-based forecast published by BNEF (2020).

Figure 5.4 shows the resulting EV sales scenarios for the four evaluated regions. The RoW sales has the most heterogeneity between scenarios. S3 (IEA forecast) shows the RoW having nearly double the next highest estimate (S1 - Bass based on policy), while S2 (Bass based on historical sales only) is the only scenario for the RoW to have the smallest market. Sales are currently low in the RoW but there is a large market potential. If sales follow typical diffusion rates of technology (S2), and there are no policy initiatives, sales will likely stay low over the next twenty years. S4 (BNEF) forecasts the market to have a delayed uptake in later years resulting in a steep incline. Overall, the policy-based scenarios (S1 and S3) have the highest global sales.

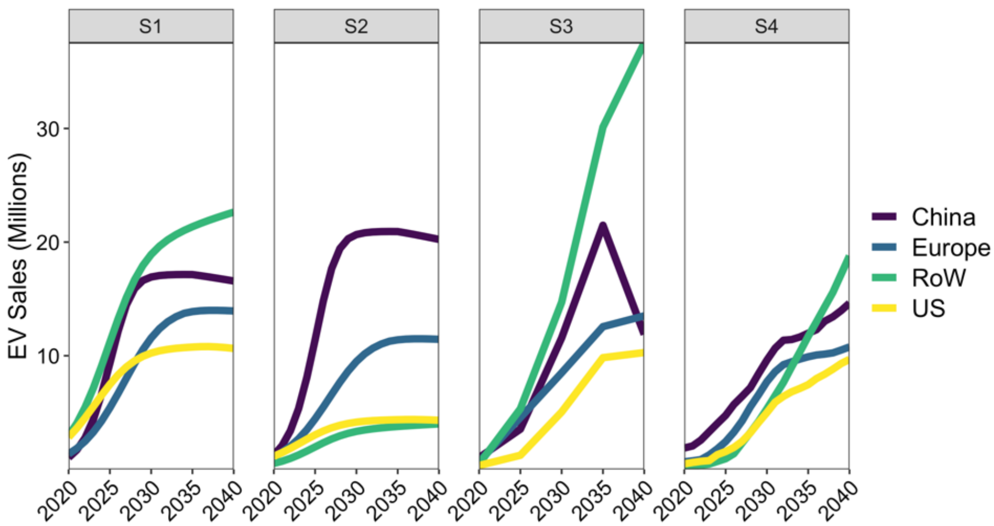

Figure 5.4: The regional EV sales under each sales forecast (Bloomberg New Energy Finance, 2020).

5.5.3. Battery Capacity Estimation

EV sales alone are insufficient for determining the total battery capacity sold in a region. A weighted average of the historical EV battery capacity per year and region is calculated using data from EV Volumes (2020). Regression analysis is then used to forecast future average capacity, taking into consideration two coefficients: year and region.

The capacity forecast model is assumed to have constant variance, normality, and independence of the errors. The residual plot (Figure 5.5) indicates normality and constant variance by the randomly distributed points forming a band of approximately the same size around zero.

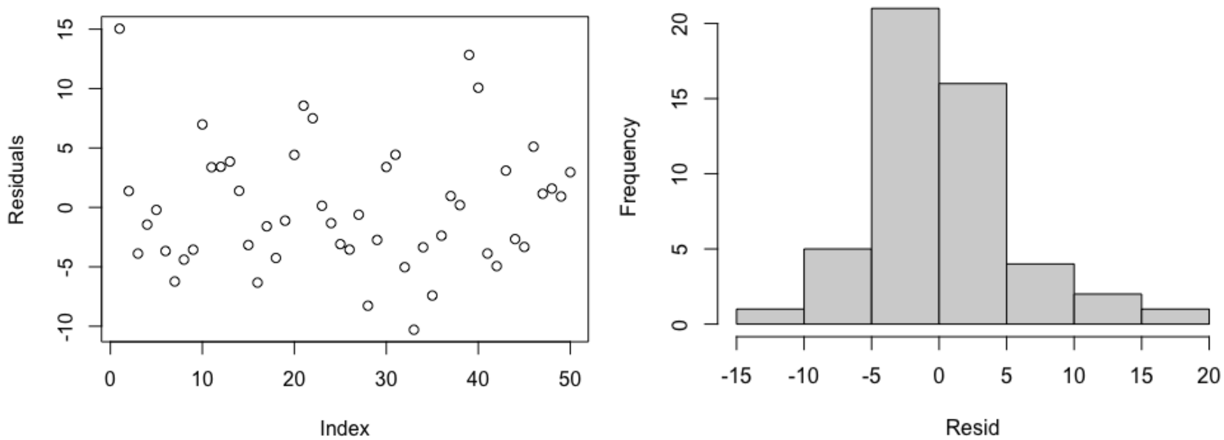

Figure 5.5: Residual plot and histogram of the multinomial regression model of capacity ~ year + region.

The average EV LIB capacity is projected to 2040 for each region demonstrating differing capacity per region, which matches the historical trend. The average capacity in kWh per region (Figure 5.6) is used to convert sales to total kWhs produced per year for each cathode scenario. These are comparable to other analysis; current estimates from the International Energy Agency (2020) estimate between 75 kWh and 100 kWh up to 2040 differing by region and year and Ambrose et al. (2020) at 100 kWh as a global average.

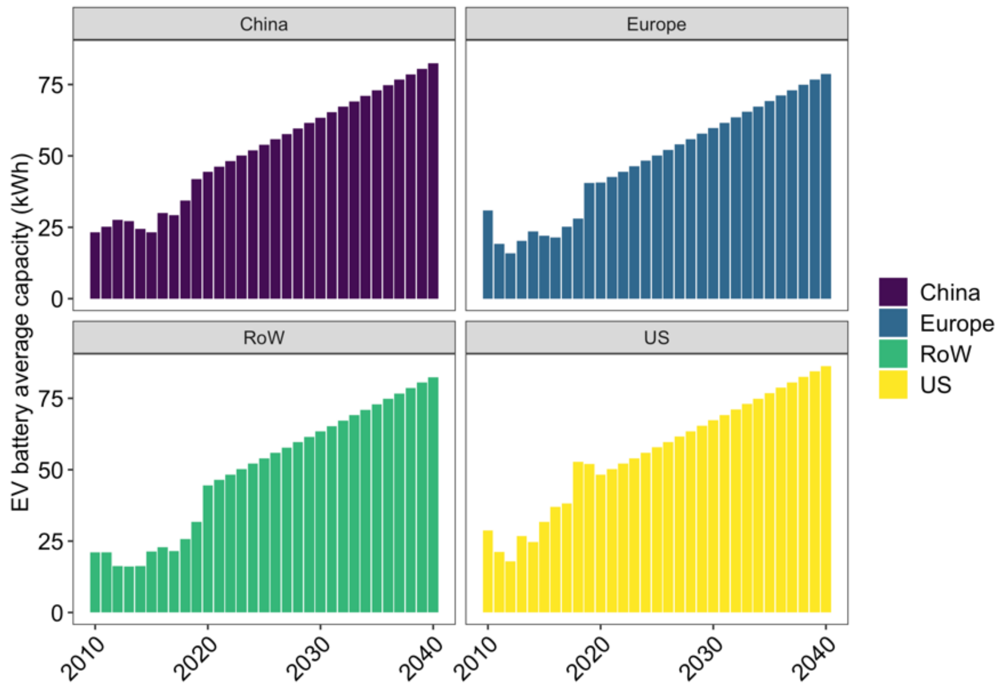

Figure 5.6: The average capacity of LIBs used in light duty EVs and forecasted until 2040.

5.5.4. Historical and Future LIB Cathode Chemistry

The estimated market share of cathode chemistries by region between 2010 and 2020 is shown in Figure 5.7, revealing substantial differences in chemistry by region and overtime. For example, China is essentially the only consumer of LFP batteries, and their market share dropped from more than 75% in 2011 to less than 10% in 2020, with a market dominated by NMC chemistries. Due to Tesla using LFP for the Model 3 and Model Y standard range models, some forecasts have predicted it to again have widespread use (C. Xu et al., 2020). Although, historical sales show the US market becoming increasingly dominated by NCA due to Tesla's large market share, with the NMC chemistry gaining popularity among other auto manufacturers. These trends show continued and rapid evolution in different chemistry adoption and regional differences may persist in the future.

Regional EV sales and battery type (LCO, LMO, NCA, and LFP) are taken from EV Volumes (2020); the share of NMC 111, 523, 622 and 811 that comprise NMC category developed using data from BNEF (2018) and Benchmark Mineral Intelligence (2020). The data for 2020 only includes the month of January.

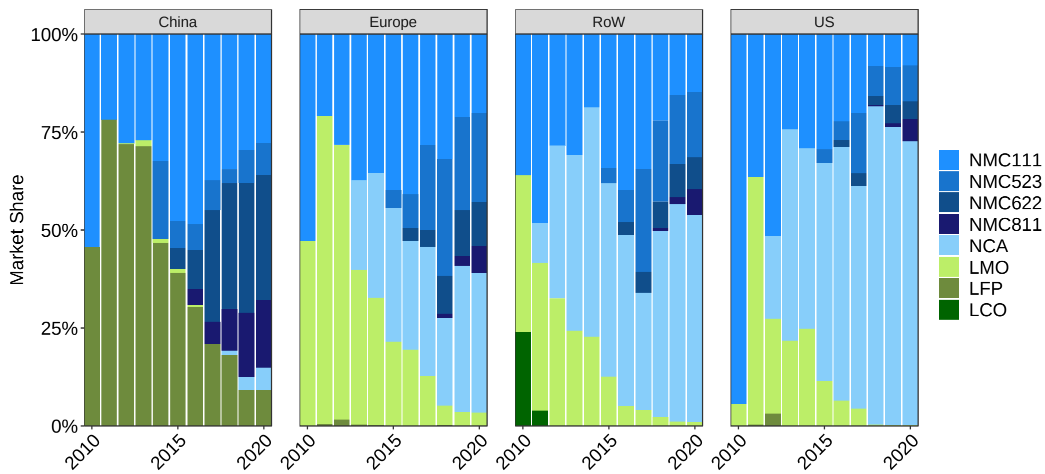

Figure 5.7: The cathode chemistry of historical sales for each region.

To observe the effect of changing cathode chemistries, and in recognition of the significant uncertainty in how this change may occur, six scenarios are considered:

C1: 2020 market share in each region is held constant until 2040 (EV Volumes, 2020).

C2: Market forecast based on an estimate by Benchmark Mineral Intelligence (2020).

C3: Current market shares in all regions transition linearly to 100% NCA by 2040.

C4: Current market shares in all regions transition linearly to 100% NMC 811 by 2040.

C5: Current market shares in all regions transition linearly to 100% NMC 622 by 2040.

C6: Current market shares in all regions transition linearly to 100% LFP by 2040.

This scenario analysis takes a bounded approach by providing extreme cases (C1, C3:C6) of cathode chemistry market change, and a market-based forecast (C2) considered to be the baseline, or most likely case (though not in a probabilistic sense). Holding the market share constant (as in scenario C1) provides a lower bound scenario to compare with futures where changes in cathode chemistry over time lead to the dominance of either NCA (C3), NMC 811 (C4), NMC 622 (C5), or LFP (C6) by 2040, all of which contain a relatively low amount of cobalt, or in the case of LFP, do not contain any. C1 is a worst-case scenario for cobalt, representing high demand due to the dominance of NMC 111. Scenarios C3:C6 represent unrealistic cases with 100% market penetration of each selected chemistry in 2040. Despite the low probability of a single-chemistry future, scenarios C3:C6 examine the potential effects of cathode chemistry changes have on circularity.

5.5.5. Battery pack composition

Material intensity estimates (kg/kWh) from Argonne National Laboratory's BatPaC (2020) model are used to convert kWh of batteries into kg of materials demanded (Table 5.2). The lithium weight includes materials in both the electrolyte and the cathode; the nickel, cobalt, and manganese weight includes material in the cathode; the aluminum weight includes material in the current collectors, cell terminals, thermal conductors, and model and battery enclosures; the copper weight includes the material in cell current collectors, terminals, thermal conductors, and the module and battery enclosures; and the graphite weight represents material in the anode.

Table 5.2: Lithium-ion battery pack composition by weight (kg/kWh).

	LFP	*LMO*	*NCA*	*NMC111*	*NMC532*	*NMC622*	*NMC811*
Lithium	0.095	0.106	0.102	0.141	0.136	0.118	0.100
Nickel	0.000	0.000	0.672	0.351	0.508	0.531	0.600
Cobalt	0.000	0.000	0.127	0.352	0.204	0.178	0.075
Manganese	0.000	1.396	0.000	0.328	0.285	0.166	0.070
Aluminum	3.528	3.369	2.920	3.110	3.070	3.017	2.921
Copper	0.946	0.863	0.564	0.677	0.661	0.605	0.549
Graphite	1.085	0.911	0.978	0.978	0.981	0.960	0.961

5.5.6. Retired Material

Retired material estimates hinge on the lifespan of EVs and any second-life uses or other delays to final disposition of a battery. In their LIB MFA studies, Song et al. (2019) and Yano et al. (2016) estimate EV LIB lifespan distributions using a three parameter Weibull density function (Equation 5.7) comprised of a location parameter a (the minimum possible life, 0); a scale parameter b that represents when in time a large portion of EVs will fail (i.e., the average lifetime); and shape parameter c, which is calculated using the US scrap rate from Jacobsen et al. (2015) (Equation

5.8). Due to differences in vehicle lifetime by region, the Weibull density function for each region must be calculated separately. Here we assume b is 15 years in the US and Europe (Hooftman et al., 2020; Staff, 2016), 14.5 years in China (Hao et al., 2011), and 16 years in RoW. Figure 5.8 demonstrates the Weibull distribution in the US.

$$fx(x|a,b,c) = \frac{c}{b}\left(\frac{x-a}{b}\right)^{c-1} exp\left(-\left(\frac{x-a}{b}\right)^{c}\right), x \geq a. \quad \text{Eq. 5.7}$$ [56]

$$c = ln\ ln\left(\frac{1}{1 - scrap\ rate}\right) = 2.07 \quad \text{Eq. 5.8}$$

In this analysis, we assume no second life uses and insignificant storage times, except for in Figure 5.12 where the impact of a prolong lifespan is explored. LIBs are therefore assumed to go directly to recycling or landfill when they are retired from use in the EV. The quantity of retired batteries per year is then calculated using the Weibull distribution.

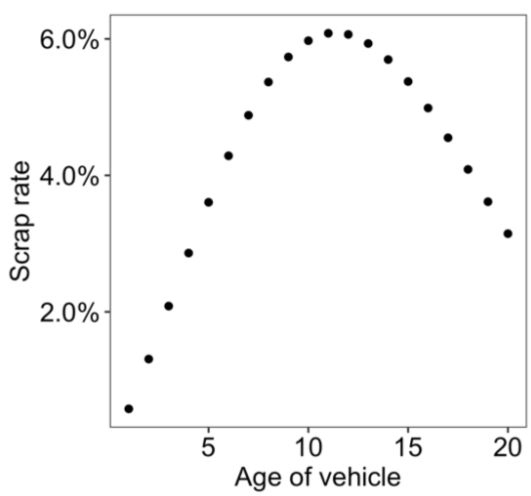

Figure 5.8: The vehicle scrappage rate in the US based off the Weibull distribution.

5.5.7. Recycling Efficiency

We assume a material recovery rate of 95% for all cathode materials, which is consistent with reports from recent hydrometallurgical recycling processes (e.g., Lithion (2019), Northvolt (2020)). This represents a best-case scenario and is meant to provide insight into how much material could theoretically be recovered if all retired EV batteries were collected and recycled in a state-of-the-art facility. In practice, retired EVs could follow any number of unregulated end-of-life pathways, and the collection rate for EV batteries is uncertain. Furthermore, metal recovery rates will likely be dictated by economic feasibility, rather than scientific potential; for example, lithium will only be recycled if the value of recovered material is high enough for recyclers to recoup the recycling cost.

5.6. Results

5.6.1. Quantity of Material Demanded and Retired

The results of the MFA include annual and cumulative demand for materials, as well as annual and cumulative retirement of materials from EV LIBs. This analysis will focus specifically on the circularity potential of the cathode materials cobalt, lithium, manganese, and nickel. Table 5.3 shows the material demanded and retired in the year 2040 under the scenario combination of S1 and C2, referred to as the baseline scenario.

The retired supply represents the total amount of potentially recoverable material in a given year, assuming a 100% collection rate and 5% loss during recycling, an overestimate of even the best real-world case (for example, lead acid batteries in the US (US EPA, 2020)). This is also under the assumption that used vehicles are not exported from the region of their initial sale, a gross simplification considering the US, European Union, and Japan are reported to have exported a total of 14 million vehicles between 2015 and 2018 (Baskin et al., 2020). Under these idealized conditions, material from retired batteries in 2040 could supply 58% of material demand in China, 60% of demand in the US; and 48% of pack material demand in Europe. If this constraint is lifted, the exporting companies will likely have lower circularity potentials, while importing countries will acquire more recyclable materials.

Table 5.3: EV LIB cathode materials demanded and retired in 2040 under the baseline scenario (S1 and C2).

		Material Name and Flow (thousand metric tons)			
		Cobalt	***Lithium***	***Manganese***	***Nickel***
China	Demand	168.37	150.70	162.05	698.50
	Recycled	118.04	90.37	117.23	383.47
Europe	Demand	135.23	121.01	130.15	561.02
	Recycled	74.38	58.73	74.68	267.13
RoW	Demand	229.90	205.74	221.28	953.80
	Recycled	123.89	98.77	108.27	468.67
US	Demand	113.20	101.30	108.95	469.62
	Recycled	73.47	60.24	55.21	300.75

5.6.2. Effect of cathode chemistry development

The potential for material circularity is highly dependent on cathode chemistry and its evolution over time. Due to the in-use residence time of batteries, the supply of materials reflects previous battery chemistry paradigms, and thus allows for circularity in certain metals as LIB chemistries reduce demand for cobalt and other high value materials over time. Figure 5.9 shows that substantial market penetration of NMC 811 (scenario C4), NCA (scenario C3) and LFP (scenario C6) increases the potential for retired materials to meet demand, particularly for manganese and cobalt (along with nickel in scenario C6). The NMC 811 cathode chemistry uses one third the cobalt of NMC 111, the most common NMC battery today (Ding et al., 2019), illustrating why circularity for some materials could be possible under conditions of changing cathode chemistries.

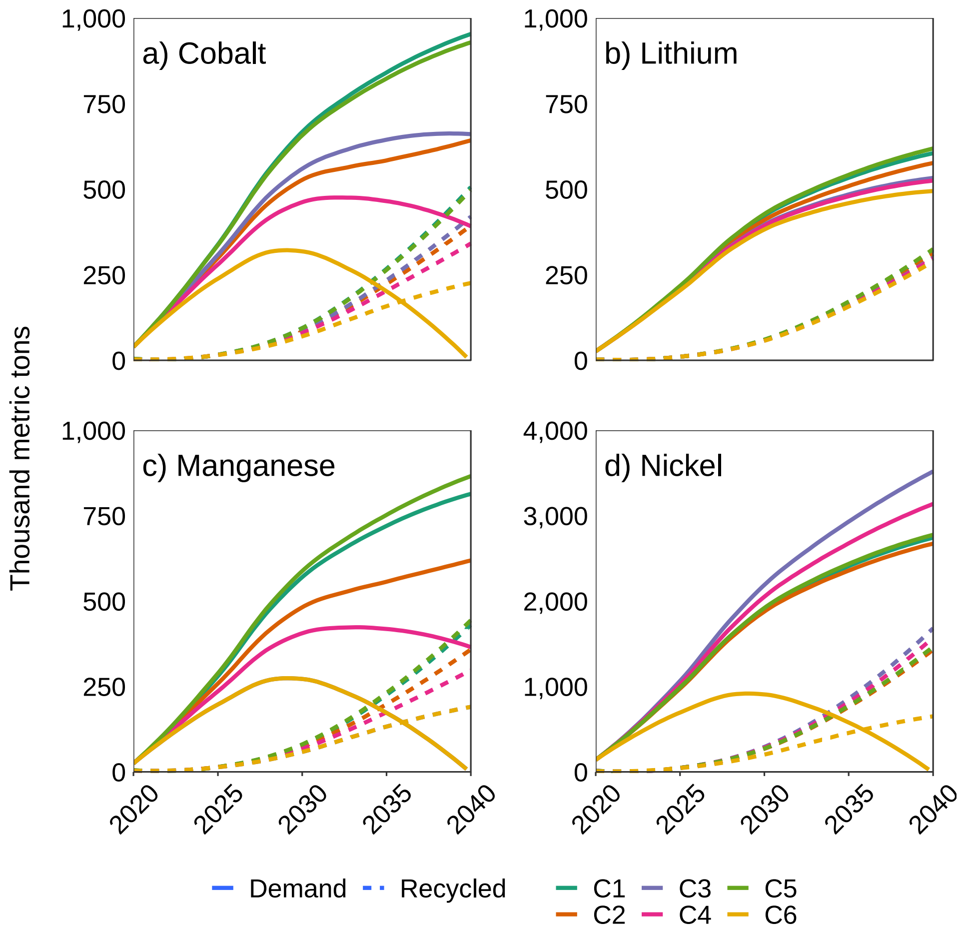

Figure 5.9: Global EV cathode battery material demand and retired supply in the policy-based sales scenario (S1) from 2020 to 2040.

Scenario C4 yields greater potential for circularity in cobalt (Figure 5.9a) and manganese (Figure 5.9c) compared to a future where today's chemistry mix is held constant over time (C1). Because NCA does not use manganese, the quantity of retired manganese in scenario combination S1-C3 surpasses demand in 2036. Scenario C6 has the greatest circularity potential of cobalt, manganese, and nickel due to the LFP cathode chemistry not including cobalt or manganese, but the cathode instead consisting of lithium and iron. The complete set of potential circularity results are provided in Table B of Appendix A. Each cathode scenario results in differing average material weights in 2040 (Table 5.4). The low average kg/kWh of these scenarios is the driver behind higher circularity.

Table 5.4: Average material weight of retiring supply in 2040 (kg/kWh).

Cathode scenarios:	***C1***	***C2***	***C3***	***C4***	***C5***	***C6***
Aluminum	2.956	3.012	2.920	2.921	3.017	3.528
Cobalt	0.182	0.123	0.127	0.075	0.178	-
Copper	0.610	0.616	0.564	0.549	0.605	0.946
Graphite	0.958	0.971	0.978	0.961	0.960	1.085
Lithium	0.116	0.110	0.102	0.100	0.118	0.095

Manganese	0.156	0.119	-	0.070	0.166	-
Nickel	0.524	0.511	0.672	0.600	0.531	-

The excess supply of manganese illustrates a key challenge to closed-loop recycling; as cathode chemistry changes, materials that are used today may become irrelevant to battery production in the future. The reverse can also be true – changing chemistries can increase demand for some materials. In 2040, for example, demand for lithium in scenario C3 is nearly double that of C4, and under both the C3 and C4 scenarios, the quantity of nickel demanded by 2030 will steadily increase, far outpacing cobalt, lithium, and manganese. This growing demand has implications for the future environmental impacts of primary nickel. Nickel is increasingly mined from laterites, which are more energy intensive to process compared with sulfide ores that have historically supplied nickel plus coproduct cobalt (J. B. Dunn et al., 2015). With the exception of aluminum, scenario C4 requires less total material over time compared to other scenarios. This is mostly because NMC 811 is more energy dense than other chemistries (see Table 5.2).

5.6.3. Impact of Sales Projections

The rate of EV adoption influences the timeframe of battery retirement. A faster adoption rate (as in scenarios S1 and S2) results in higher initial material demand and therefore a larger retired supply available for recycling or second use in 2040 (illustrated for cobalt in Figure 5.10 of the supplementary materials). The faster rate of adoption is then followed by a flattening out as the expected maximum market share is reached in S1 and S2. This is produced as a result of the S-shaped adoption curve of the Bass diffusion model which enables potential circularity of cobalt in all regions for scenario C4 (100% NMC 811 by 2040), and also results in near circularity for lithium (see China in Figure B of Appendix A). Scenarios S3 and S4 assume exponential growth and therefore a slower initial rate of adoption, and a delay in material availability, though a strong inflection for China around 2035 under S3 also leads to potential circularity.

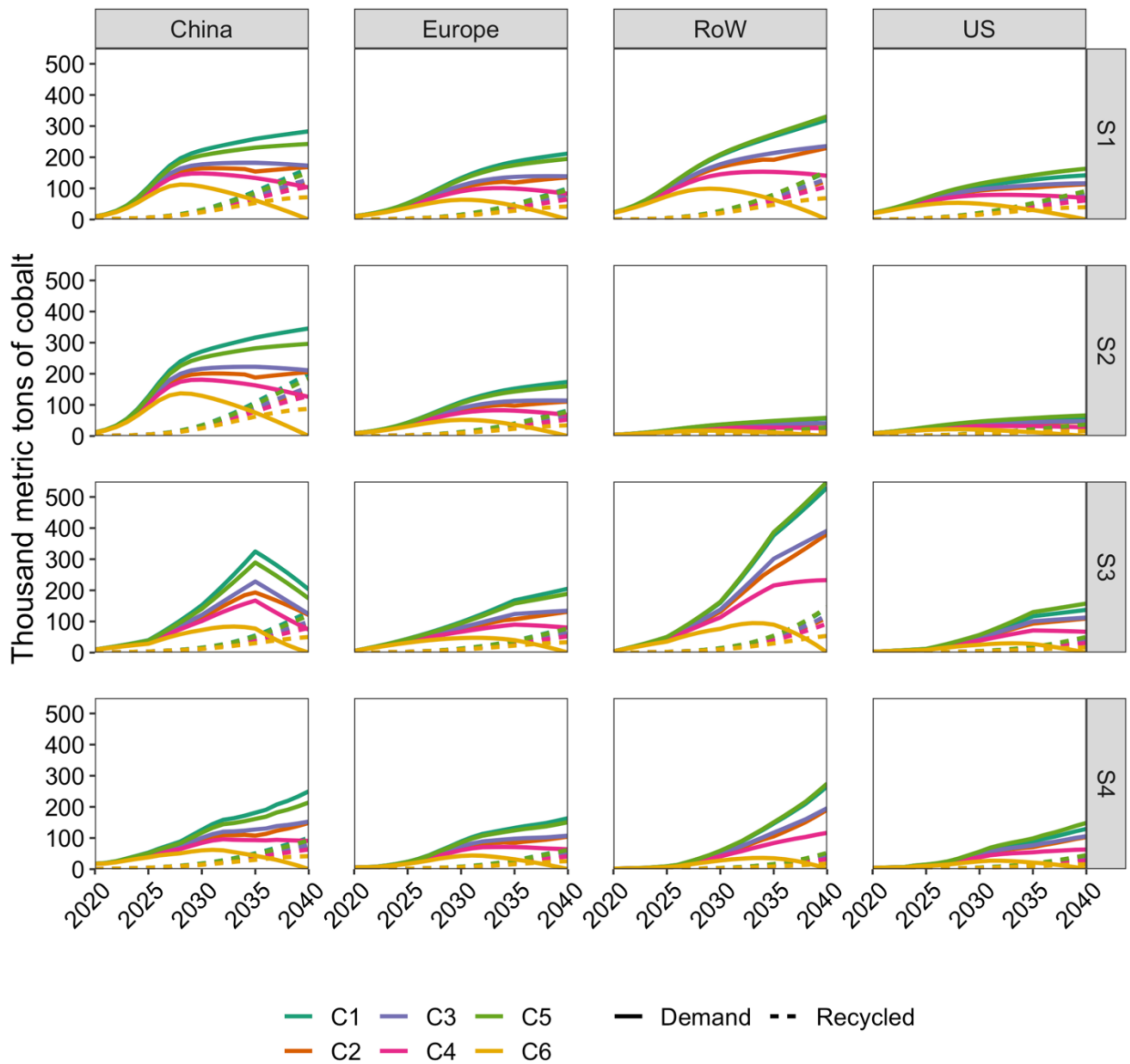

Figure 5.10: The potential circularity of cobalt from 2010 to 2040 in each region and under each cathode and sales scenario.

5.6.4. Retired GWhs and extended lifetime

Assuming LIBs are retired at a 20% battery capacity loss, between 1000 to 2000 GWh of LIBs will be retired globally in 2040. Under the baseline scenario, 20% of retired capacity will be retired in the US, 19% in Europe, 29% in China, and 31% in the RoW (Figure 5.11). After use in EVs, batteries may either be recycled, disposed of, or enter cascaded reuse. Cascaded reuse extends the lifetime of batteries by repurposing remaining capacity in less strenuous applications, such as stationary energy storage or grid services applications. Reuse can avert production of new batteries thereby decreasing life cycle impacts at a systems level (Richa, Babbitt, & Gaustad, 2017a). However, reuse postpones the recycling and production of recycled materials for manufacturing and may delay the adoption of more efficient batteries in stationary sources, both of which could have significant environmental benefits as well.

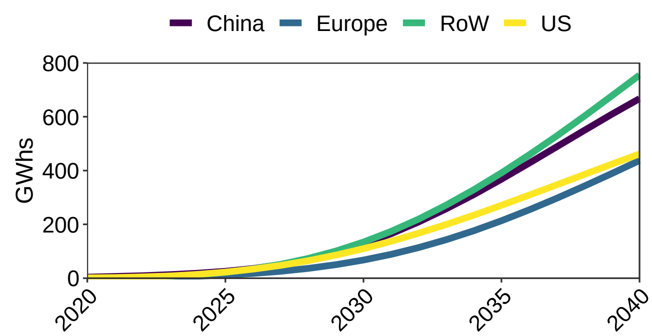

Figure 5.11: GWhs of retired EV battery capacity per region under the baseline sales scenario and assuming a 20% capacity loss from the point of manufacturing to retirement from the EV.

Lifespans vary depending on the second-life application, but range from 6 years for grid regulation services to 30 years in EV charge support (Casals et al., 2019). Additional years to the lifespan delays the year at which the material will reach circularity. This will essentially decrease the circularity of each material. Figure 5.12 demonstrates cobalt circularity will decrease from 60% to 37% in 2040 if six years are added to lifespan under the baseline scenario, and to 4% if fifteen years are added. An MFA is insufficient for determining the environmentally preferred use of a retired battery; a life cycle assessment or other environmental impact-oriented assessment is needed. This tension between the environmental benefits of reuse or recycling occurs for other product systems and materials as well, such as aluminum (G. Liu et al., 2013).

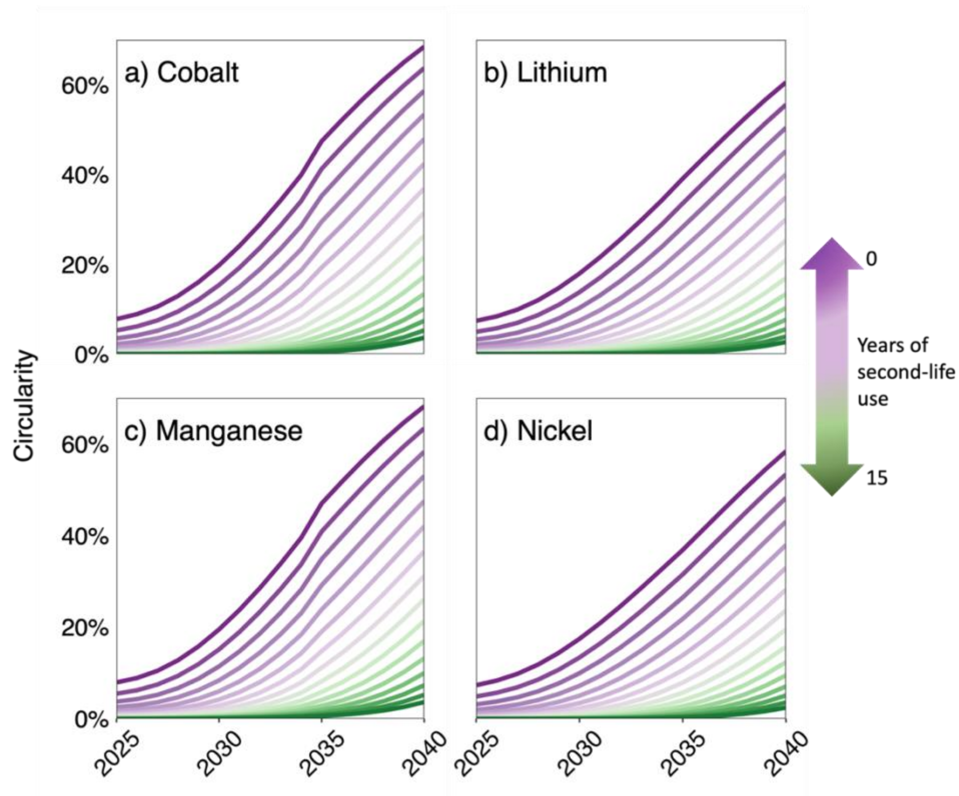

Figure 5.12: The circularity potential of materials as additional years are added to the lifespan of the LIB in the baseline scenario (S1 and C2).

5.6.5. Geography of EV Demand versus LIB production

Total material demand from 2010 to 2040 is dispersed globally, with China representing 28%, the RoW representing 32%, Europe representing 20%, and the US representing 20%. This does not align with the location of production; while China does not supply the majority of ore, it controls 47% of LIB manufacturing (EV Volumes, 2020), and 44% of cobalt refining (Statista, 2016). The vulnerability of material supply is exacerbated by the geographic concentration of manufacturing of LIBs and refining of materials. A 2019 study by Gulley et al. (2019) arrives at similar findings and predicts China may reserve production of cobalt for its domestic manufacturers. As illustrated in Figure 5.13, there are large differences between material demand of regions and the location of LIB battery manufacturing, cobalt refining, and LIB recycling. The manufacturing data is sourced from EV Volumes (2020) and the LIB recycling data is sourced from Circular Energy Storage (Melin, 2020).

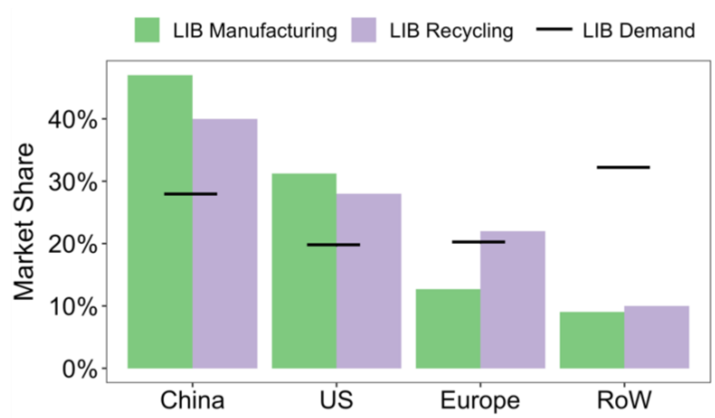

Figure 5.13: Comparison of LIB manufacturing capacity, recycling capacity, and material demand.

5.7. Discussion

5.7.1. Barriers to circularity

The results suggest that there is potential for near circularity of cobalt in China, Europe, and the US in 2040. A key driver is the adoption of lower-cobalt chemistries over time that lead to a convergence of demand and supply, a similar finding to Baars et al.'s (2021) study of LIB material flows in the EU. While circularity is, in theory, possible, a number of practical barriers exist. The first is one of geography; demand does not correspond to current manufacturing capacity in most regions (Figure 5.13). Recycling and manufacturing facilities will need to be developed in each region if they are to meet a greater portion of their own demand, rather than perpetuating the concentration of production and demand in different regions that exists today.

The second barrier is one of economics and policy. Cobalt is the most expensive cathode material, and its recovery is a key motivator for recycling. However, due to high economic and social cost, future batteries will evolve to lower or zero content, thus presenting a problem for a sustained market-driven recycling industry (J. B. Dunn, Gaines, Barnes, et al., 2012; E. A. Olivetti et al., 2017), and necessitating policies to support a robust recycling industry and encourage material circularity. LIB recycling is an emerging industry and policymakers have the opportunity to secure a material supply by supporting domestic recycling (Hao et al., 2017; International Energy Agency (IEA), 2020), and the need for policy intervention is heightened given the likelihood of lower value materials in future EV LIBs.

5.7.2. More robust policy needs to be enacted to encourage circularity

China has already enacted policies that require design for disassembly and domestic recycling (Chinese Ministry of Industry and Information Technology, 2018) and the EU recently updated the Battery Directive which focuses on using extended producer responsibility, recycling rates, and recycled content requirements to create a robust LIB recycling industry (European Commission, 2020). India, Japan, and several other countries also have LIB end-of-life policies that have not been sufficient to drive sufficiently high rates of recycling (International Energy Agency (IEA), 2020). The US trails behind with no national policy that requires or incentivizes material circularity (Gaines et al., 2018; C. J. Xu et al., 2017), although there have been major efforts to decrease the cost of recycling, specifically through the development of direct recycling by the ReCell center. Despite the lack of effective policy in much of the world, a substantial number of batteries will be reaching their end-of-life in the next 20 years, which coincides with the large increase in material demand, and presents an opportunity for effective recovery. Thus, there is still sufficient time to enact policy to achieve greater LIB material circularity and realize the potential economic and environmental benefits of creating a domestic supply of secondary materials in high-demand countries and regions.

5.7.3. Future research

LIB cathode material recovery and recycling could meet a large fraction of future material demand, thereby decreasing demand for virgin material, reducing battery waste, and potentially reducing the impacts of new batteries if recycling is less environmentally intensive than primary production. Future research that incorporates realistic collection and recovery rates is necessary to accurately calculate the recycled material available for use in manufacturing new batteries, and comprehensive approaches to understanding environmental impacts of alternatives, such as life cycle assessment, are required to anticipate the best use of retired EV batteries. Additionally, analysis that reviews the impact of increased energy density due to battery design and the substitution of silicon for graphite in the anode would add to this research. This was not assessed in this paper due to the focus on the impact of changing cathode chemistries on material circularity.

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6. Electric vehicle lithium-ion battery recycled content standards for the US – targets, costs, and environmental impacts

6.1. Scope and purpose

This section builds upon on the work in Section 5 and calculates potential recycled content standards for the US. In addition, the environmental impacts and economics of battery recycling are calculated and evaluated for recycling US supplies, in both the US and in China.

This section is adapted from the following publication:

Dunn, J., Kendall, A., Slattery, M. Electric vehicle lithium-ion battery recycled content standards for the US – targets, costs, and environmental impacts (Accepted).[b]

6.2. Introduction

One potential policy lever to encourage recycling is a recycled content standard (RCS). RCSs mandate a percent of constituent material in a product to be from recovered sources, which can increase recycling rates by creating a market for the reclaimed material. The US has implemented this type of standard for the newsprint, plastic, and glass industries (Aunan & Martin, 1994), but has not passed or proposed RCSs for LIBs; however, the European Union, an important EV LIB market, has included an RCS as part of their revised battery regulation (European Commission, 2020).

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[b] Jessica Dunn contributed through conceptualization, software, formal analysis, writing – original draft; Margaret Slattery contributed conceptualization; Alissa Kendall contributed through conceptualization, supervision, funding acquisition, writing – review

While federal LIB End-of-life policy has yet to be passed, the US has begun exploring the national interest of establishing a secure LIB supply chain. The Biden Administration's Executive Order 14017, "America's Supply Chains," required a 100-day analysis of supply chains within the US, including large capacity LIBs. This report states the US battery supply chain is highly exposed to risk and the US currently cannot supply all materials domestically. The report further concludes that this risk is an adverse side effect of the historical prioritization of efficiency and low cost over sustainability, thus resulting in reliance on low-cost providers overseas, instead of investing in a domestic supply.

In the vacuum of federal policy, states within the US are exploring policies to increase the recycling rates of LIBs. The State of California's 2019 Assembly passed Bill No. 2832 which created a stakeholder advisory group tasked with recommending policy to the 2022 legislature that will lead to as close to 100% reuse and recycling as possible of End-of-life EV batteries (*Assembly Bill No. 2832*, 2018). The advisory group discussed RCSs, although they did not recommend it as a policy, expressing hesitancy due to a lack of knowledge around the optimal level of RCSs for the US, and an unknown cost of recycling (Kendall et al., 2022).

6.3. Literature review

There is currently no academic literature that analyzes the proposed EU RCSs, calculates appropriate standards for the US, or assesses the environmental and economic use of these standards for LIBs. Prior analyses have estimated the future demand of materials to manufacture LIBs for the US (Richa et al., 2014; Shafique et al., 2022; C. Xu et al., 2020), China (W. Liu et al., 2021; Shafique et al., 2022; Song et al., 2019), the EU (Baars et al., 2021), and South Korea (Kim et al., 2018), as well as the circularity potential of these materials (i.e., the potential for retired supplies to meet the material demand) (Baars et al., 2021; J. Dunn et al., 2021; Richa et al., 2014;

C. Xu et al., 2020). These estimates have demonstrated the potential for the recovered materials from retired EVs to provide a substantial source of supply. Dunn et al. (2021) forecast a wide range of circularity potentials for the US in 2040 for the materials cobalt (35% to 93%), lithium (35% to 68%), nickel (35% to 69%), manganese (29% to 64%), and aluminum (34% to 64%). Xu et al. (2020) estimate a wide range of global circularity potentials in 2050 for lithium (>30% to 50%), cobalt (>40% to 70%), and nickel (>30% to 55%). These large spreads from both Dunn et al. and Xu et al. are due to uncertainty in the future cathode market shares, sales forecasts, and the portion of batteries used in second-life applications. While these circularity estimates are informative, they are based exclusively on the quantity of material available and do not reflect economic feasibility or realistic collection and processing recovery rates. A more tightly defined range representing the near-term circularity potential is needed to guide policy discussions and developments. Thus, this research estimates feasible US RCS for cobalt, lithium, and nickel, that can serve as targets in the discussion or development of RCSs for light-, medium- and heavy-duty EV LIBs in the US market.

Feasibility of RCS is explored by estimation of the cost and environmental impacts, which include life cycle emissions of CO_2e, SO_X, and NO_X, from recycling LIB materials to battery grade quality. Prior research has demonstrated that recycling is environmentally preferable over landfill disposal, with differing impacts dependent on the recycling process, cathode chemistry, and carbon intensity of the grid (Ciez & Whitacre, 2019; J. B. Dunn, Gaines, Sullivan, et al., 2012; Ellingsen et al., 2014; Gaines, 2018; Gaines et al., 2010; Mohr et al., 2020; Rajaeifar et al., 2021). This paper adds to the LCA literature by calculating the environmental impacts of recycling LIBs retired in the US, either domestically or in China. In addition, the economics of recycling a mixed cathode chemistry stream of LIBs is calculated. While it is currently disputed if recycling of LIBs is profitable, previous research has attempted to capture the economics of LIB recycling (Table 6.1) (Bernhart, 2019; Ciez & Whitacre, 2019; Foster et al., 2014; Gaines & Cuenca, 2000; Hanlon, 2016; Ma et al., 2018; Mossali et al., 2020; Qiao et al., 2019; Rahman et al., 2017; C. R. Standridge et al., 2016; Steward et al., 2019; X. Wang, Gaustad, Babbitt, Bailey, et al., 2014; Z. Wang, 2020; Xiong et al., 2020). Choubey et al. (2017) is the only study to analyze a mixed cathode stream, reporting a profit from hydrometallurgical processing.

Table 6.1: Literature review of the economics of recycling lithium-ion batteries.

Author	***Recycling Technique***	***Recycled material***	***Cathode chemistry***	***Revenue, cost, or profit/loss***	***$/kg***
Standridge et al. (2014)	Unknown	Full pack	LCO	Loss	-$9.14
Choubey et al.(2017)	Pyro + Hydro	Full pack	NMC 111	Profit (2020 values)	$8.43
Choubey et al. (2017)	Pyro + Hydro	Full pack	NMC 111	Profit (2016 values)	$2.31
Roland Berger (2019)	Hydro	Full pack	NCM 622	Profit	$0.53
Ciez and Whitacre (2019)	Direct	Cathode	NMC and NCA	Cost	$6.00
Ciez and Whitacre (2019)	Direct	Cathode	LFP	Cost	$17.00
Qiao et al. (2019)	Hydro	Full pack	NMC	Profit	$0.74
Xiong et al. (2020)	Hydro	Full pack	NMC 111	Profit	$1.29
Hanlon (2016)	Hydro	Full pack	LCO, NMC, LMO, LFP	Cost	$3.97
Rahman et al. (2017)	Unknown	Full pack	LCO	Savings	48.8%
Foster et al. (2014)	Unknown	Full pack	NMC111	Loss	-$9.19

Wang et al. (2014)	Hydro	Full pack	LMO, LCO, LFP	Revenue	.95 - 9.81
Ma et al. (2018)	Hydro	Full pack	LFP, NMC 111	Profit	.59 - 3.04

In this analysis, three different recycling processes are considered: hydrometallurgical, pyrometallurgical, and direct recycling. These results are then compared with the material value and avoided emissions of recovered materials. Because the cost and environmental impact of recycling is a function of where recycling occurs, three scenarios are modeled that consider the location of recycling and mode of transportation, an aspect that has historically been overlooked in LIB End-of-life cost estimates (Slattery et al., 2021). Recycling is modeled to occur in the US under two possible transport modes, truck or train, or is modeled to occur in China. China is currently the only market with significant LIB recycling infrastructure.

6.4. Materials and Methods

Estimates of feasible RCS for cobalt, nickel, and lithium used in LIB traction batteries are calculated for the US using material flow analysis (MFA) from 2020 to 2050. Results from the MFA are then used in the Argonne National Lab's EverBatt (2021a) and GREET (2021b) models to estimate the cost and environmental impact of recycling the batteries retired until 2050.

6.4.1. Material flow analysis

MFA is used to forecast the demand for new materials and the quantity of retired and reclaimed materials for light-, medium-, and heavy-duty vehicles until 2050. Process a in Figure 6.1 calculates the demand of materials (t) per year to manufacture the lithium-ion batteries. Process b calculates the reclaimed material which can then be used in the manufacturing process.

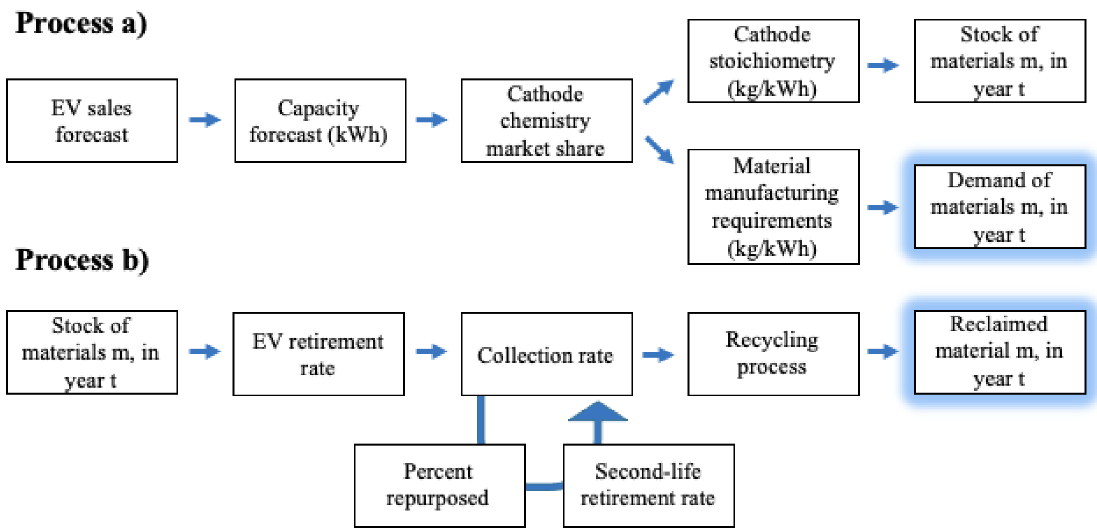


Figure 6.1: The material flow model.

The demand for new materials is calculated based on EV sales, capacity of batteries, cathode chemistry, and manufacturing requirements. The quantity of materials reclaimed from recycling is calculated based on the EV lifespan, LIBs used in a second-life application, second-life lifespan, recycling process, collection rate, and manufacturing scrap rate. Scenario analysis is used due to the uncertainty of these inputs, resulting in 864 different scenarios as described in Table 6.2.

Table 6.2: Scenarios used in the material flow analysis.

Model input	***Scenarios***
EV sales forecast	Two scenarios taken from the International Energy Agency's Mobility Model (IEA MoMo) (International Energy Agency, 2020) (Figure 6.2) 1) Stated Policies Scenario (STEPS) 2) Sustainable Development Scenario (SDS)
Cathode chemistry forecast	Two scenarios taken from Xu et al. (C. Xu et al., 2020) (Figure 6.3) 1) NCX: Chemistries containing nickel and cobalt dominant in 2050 2) LFP: Lithium-iron-phosphate (LFP) chemistry dominant in 2050
Percent repurposed	10%, 25%, 50%
Failure rate of 2nd life	A lognormal distribution that is based on the average cycles completed per year: 365 cycles, 183 cycles, and 92 cycles
Recycling process	Hydrometallurgical, pyrometallurgical, and direct recycling
Collection rate	1) Step increase from 65% in 2025 to 90% in 2050 by 5% increments 2) Flat collection rate from 2020 to 2050 analyzed for 7 different rates/scenarios: 65%, 70%, 75%, 80%, 85%, 90%, 95%.

EV sales data and forecast

The historical sales data for light-, medium-, and heavy-duty EVs and plug-in hybrid EVs is gathered from EV Volumes (2020). To predict future sales, two scenarios are considered based on the IEA MoMo forecast for light-, medium- and heavy-duty vehicles (Figure 6.2). The first is based on MoMo's STEPS, which represents the policies and goals of the US. The second scenario is based on MoMo's SDS, which ensures sharp reductions in air pollutants and meet the global climate goals of the Paris Agreement (International Energy Agency, 2020).

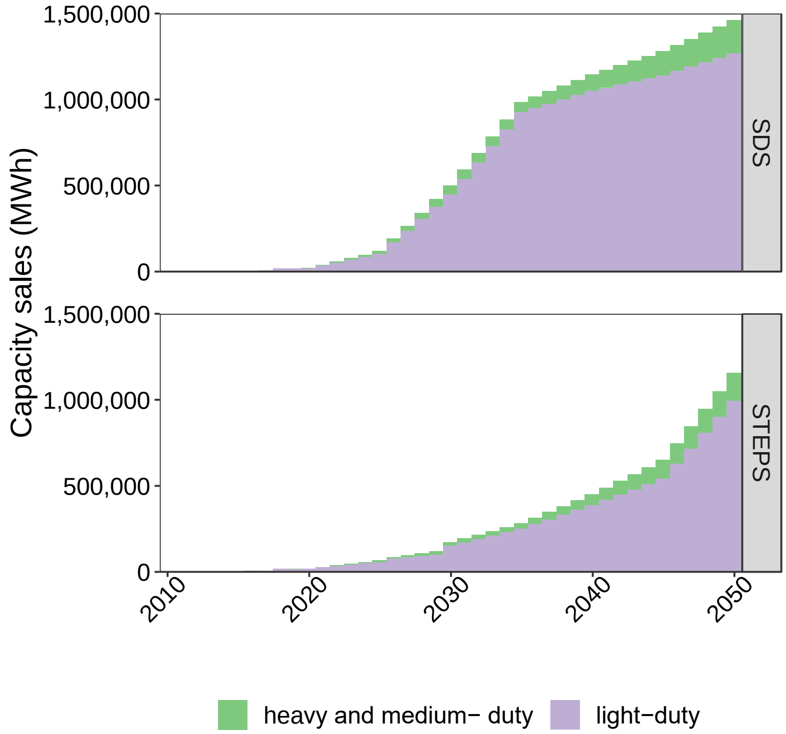


Figure 6.2: The heavy-, medium-, and light-duty EV sales scenarios from 2010 to 2050.

Cathode chemistry

Cathode chemistry is based on historical EV Volumes data until 2020 and a forecast for future years (EV Volumes, 2020). From 2021 to 2050 the forecast represents the two scenarios NCX and LFP from Xu et al (2020). The NCX scenario has the chemistries containing nickel and cobalt (e.g., NMC 632, NMC 811, and NCA) as the dominant cathode chemistries, while the LFP scenario has lithium iron phosphate (LFP) as dominant. The 2050 cathode chemistry percentages are taken from each scenario and linear interpolation was used from 2021 until 2050 (Figure 6.3).

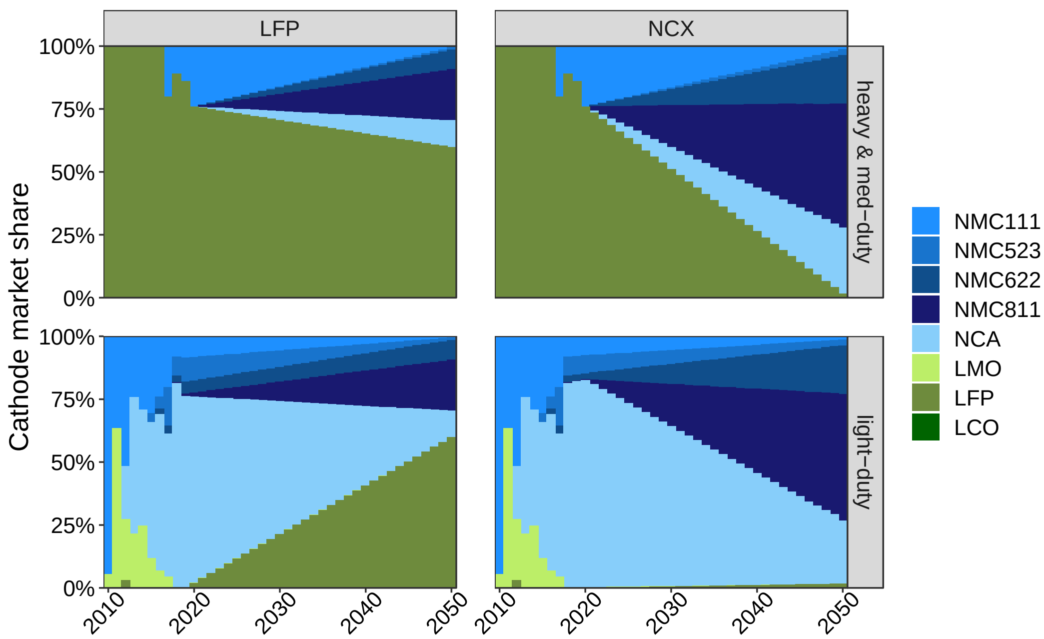
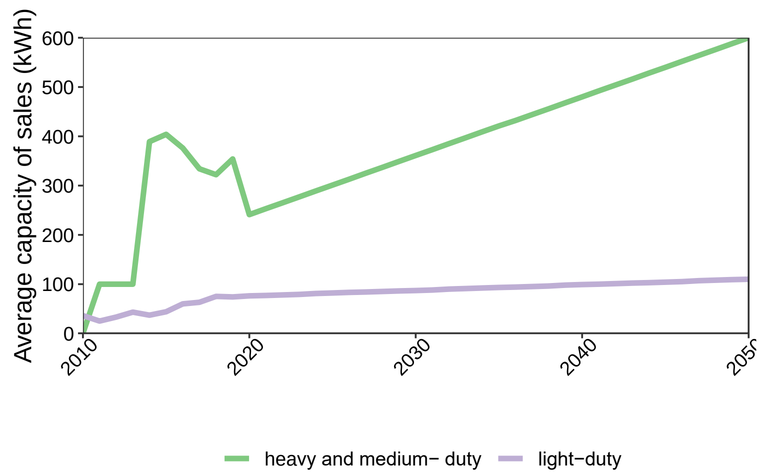


Figure 6.3: Cathode chemistry market share forecast for NCX and LFP scenarios.

Battery capacity

The average battery capacity is calculated using EV Volumes data from 2010 to 2020 (EV Volumes, 2020). From 2020 to 2050 regression analysis is used to forecast the light-duty sector. The heavy- and medium-duty forecast was created using linear interpolation to a 600 kWh battery in 2050 (Figure 6.4).

Figure 6.4: The average capacity of lithium-ion batteries in light-, medium-, and heavy- duty EVs.



EV lifespan

A Weibull distribution is used to estimate EV lifespan. The average lifespan used for light-duty vehicles is 15 years while the average lifespan used for medium- and heavy-duty vehicles is 10 years (Staff, 2016; Statista, 2016).

Second-life use and lifespan

Due to the infancy of the second-life industry, the percent of batteries that will be repurposed and the lifespan of second-life battery systems is uncertain. This analysis uses several scenarios for the repurposing and the failure rate of second-life batteries (Table 6.2).

The failure rate is calculated using a lognormal distribution of cyclical aging based on cyclical aging research of failure rates by Johnen et al. (2020) ($\mu = 7.038$ and σ=.064 when End-of-life = 50% capacity). In Johnen et al. (2020) batteries are charged and discharged between a minimum and maximum state of charge. The probability of failure is based on the number of equivalent full cycles completed, thus scenarios representing various applications are calculated based on the average cycles completed per year (Table 6.2).

Collection rate

The collection rate of LIBs represents the percentage of LIBs retiring that are collected and eventually recycled. Collection rates are uncertain due to a lack of reporting and uncertain export rates. Due to this uncertainty, the collection rate is evaluated under several scenarios. First, a flat rate for all years is assessed for the following levels: 60%, 65%, 70%, 75%, 80%, 85%, 90%, and 95%. Then, an increasing collection rate is assessed, replicating the EU requirements of 65% in 2025 and 70% in 2030, increasing by 5% increments every five years until 95% is met and held constant. It is assumed recyclers will accept all cathode chemistries collected. The cathode chemistry of batteries and scrap collected for recycling are represented in Figure 6.5.

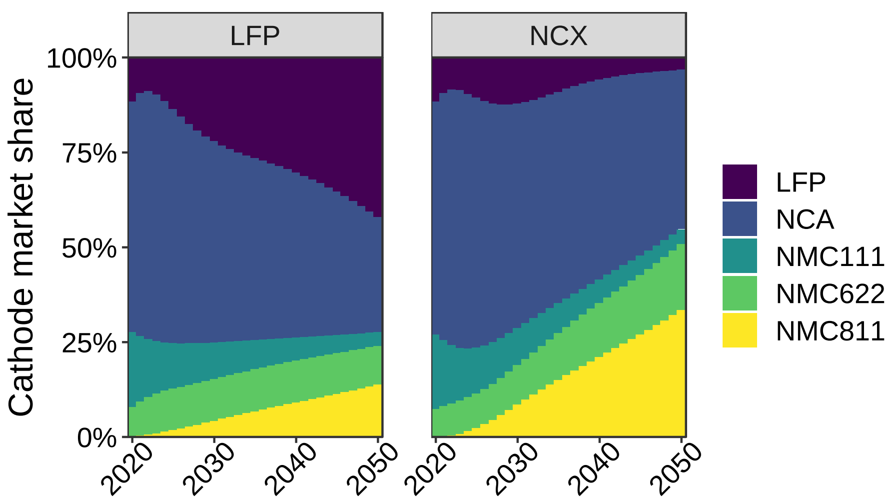


Figure 6.5: The cathode chemistry mix of batteries and scrap collected for recycling in the NCX and the LFP scenario.

Recycling processes and efficiency

The recycling processes included in this analysis are hydrometallurgy, pyrometallurgy, and direct recycling. Each use different methods for metal recovery, result in different yields, and produce different products.

Pyrometallurgical processing has been common in the recycling of electronics for metals recovery. Prior to pyrometallurgical treatment, batteries can be mechanically treated by sorting and crushing, and then subjected to temperatures of 150 to 500° Celsius to remove electrolyte and organic solvent. The pyrometallurgical process consists of heating the LIB to temperatures of 1400 to 1700° Celsius to create a copper-nickel–cobalt–iron alloy of the recovered materials and a slag of the unrecovered materials, including lithium. The alloy produced is a mixture of metals, but can be run through an additional hydrometallurgical process to recover the constituent target materials of cobalt, nickel, and copper (Assefi et al., 2020).

The hydrometallurgical recycling process also requires pre-treatment, which typically consists of discharging, dismantling and/or mechanical crushing, and sorting the following from the rest of the materials: active cathode, anode, electrolyte, copper foils, and aluminum foils. Next, the electrolyte is recovered, and the cathode active materials are separated from the aluminum foil by a dissolution process using organic solvents. The hydrometallurgical process then begins by leaching with inorganic or organic acids to create a solvent containing the materials. The materials cobalt, nickel, manganese, and lithium are then recovered from the solution using solvent extraction, chemical precipitation and/or electrochemical deposition (Yao et al., 2018).

The direct recycling method similarly begins with discharging, physical separation, electrolyte recovery, and delamination. At this point, the anode and cathode are separated using froth flotation, followed by binder removal, and then relithiation. Relithiation processes are still in the research and development stage. Several different methods are being researched, including thermal, hydrothermal, redox mediator, ionothermal, and electrothermal processes. In addition, the ReCell center is currently researching the possibility of upcycling cathodes to different stoichiometry, for example taking an NMC111 cathode and upcycling the cathode to an NMC811 cathode (Gaines et al., 2021).

The recycling efficiencies for pyrometallurgy, hydrometallurgy, and direct recycling are taken from the Argonne National Lab model, EverBatt, and are included in Table 6.3 (Argonne National Lab, 2021a).

Table 6.3: Recovery efficiency of pyrometallurgical, hydrometallurgical, and direct physical recycling taken from EverBatt (Argonne National Lab, 2021a).

	Pyrometallurgical	*Hydrometallurgical*	*Direct Physical*
Copper	90%	90%	90%
Steel	90%	90%	90%
Aluminum		90%	90%
Graphite		90%	90%
Plastics		50%	50%
Li+ in product		90%	40%
LCO			90%
NMC(111)			90%
NMC(532)			90%
NMC(622)			90%
NMC(811)			90%
NCA			90%
LMO			90%
LFP			90%
Co2+ in product	98%	98%	
Ni2+ in product	98%	98%	
Mn2+ in product		98%	
Electrolyte Organics		50%	50%

Material loss during manufacturing

The demand of materials is calculated based on the sales, capacity of batteries, and the cathode chemistry. To properly calculate the demanded materials, the material loss during manufacturing must be included. The Argonne National Lab BatPac model estimates a yield rate of 92.2% for all cathode materials, a number also used by Ciez and Whitacre (2017), and which is adopted here as well (Argonne National Laboratory, 2020; Ciez & Whitacre, 2017). In addition to the loss of cathode materials, 5% of finished cells are discarded in the final manufacturing step due their inability to retain a charge (Ciez & Whitacre, 2017). This loss is included in the sales forecast as well as the available materials for recycling.

6.4.2. Recycled content standards

Recycled content is the fraction of recovered material within a product. To calculate the recycled content that could be achieved for future LIBs, the supply of recovered material and the demand of materials for manufacturing needs to be determined. The recycled content is calculated for all scenarios listed in Table 6.2 from 2020 to 2050. This model assumes a closed loop recycling system for US batteries.

Equation 6.1 is used to calculate the recovered material *(m)* of nickel, cobalt, and lithium, for the year *(t)* from 2020 to 2050, for a given scenario *(s)* listed in Table 6.2, and for each recycling process (r) of pyrometallurgy, hydrometallurgy, or direct recycling. The recovered material is calculated by taking the retired supply for the year *(t)*, material *(m),* and the scenario *(s)*, and multiplying it by the material *(m)* recycling efficiency of the recycling process *(r)*.

$$m = material: nickel, cobalt, and\ lithium$$

$$t = year: 2020\ to\ 2050$$

$$r = recycling\ process: pyrometallurgy, hydrometallurgy, and\ direct\ recycling$$

$$s = scenario; the\ scenarios\ listed\ in\ table$$

$$\sum retired\ supply_{t,m,s} * collection\ rate * recycling\ efficiency_{m,r} = reclaimed\ material_{t,m,r,s} \qquad \text{Eq. 6.1}$$

Equation 6.2 calculates the manufacturing material demand for each material *(m)* and year *(t)* by multiplying the material demand by 1 plus the material loss, thereby accounting for the additional material needed to manufacture the batteries.

$$\sum material\ demand_{t,m,s} * (1 + manufacturing\ loss)_{t,m} = manufacturing\ material\ demand_{t,m,s} \qquad \text{Eq. 6.2}$$

In Equation 6.3, the recovered materials is divided by the manufacturing demand to calculate the percent of recycled content. This calculation is done for each material *(m),* year *(t),* recycling process *(r*), and scenario *(s).*

$$\sum \frac{reclaimed\ material_{t,m,r,s}}{manufacturing\ material\ demand_{t,m,s}} = recycled\ content\ (\%)_{t,m,r,s} \qquad \text{Eq. 6.3}$$

After the RCS for all the scenarios and materials are calculated, a 95% confidence interval is used to calculate a feasible RCS bound. The impact of the scenarios to the RCS are then analyzed. Based on this analysis, the final RCS uses an incremental collection rate beginning with 65% in 2025 and increasing to 90% in 2050, all other scenarios are included in the final RCS calculation except the use of pyrometallurgical recycling. The RCS targets assume the recycled material is battery grade, meaning batteries do not need to be designed around using lower grade material in manufacturing. This assumption is supported by industry declarations, such as the creation of battery cathode material by NorthVolt (2021) and the announcement of a partnership between Tesla and Redwood Materials (Korosec, 2022).

6.4.3. Cost & environmental impact of recycling

The cost and environmental impacts of recycling were determined using the EverBatt and GREET models (Argonne National Lab, 2021a, 2021b). The cathode chemistry market share of retired batteries from the MFA was used to determine the cost and environmental impact of recycling 1 kg of pack-level LIB materials for each year and scenario (Figure 6.6). While batteries can be measured by kg and kWhs, kg was used because it represents the mass of materials handled and reclaimed. If kWhs was used, the cost of recycling batteries with less nickel and cobalt (e.g. NMC811) would decrease compared to higher nickel and cobalt batteries (e.g. NMC111) due to their higher energy density. Similarly, the cost of recycling LFP batteries would likely increase due to the lower energy density of the batteries.

The avoided emissions from using recovered materials were determined by taking the materials recovered and calculating the equivalent virgin material impacts using the GREET life cycle inventory.

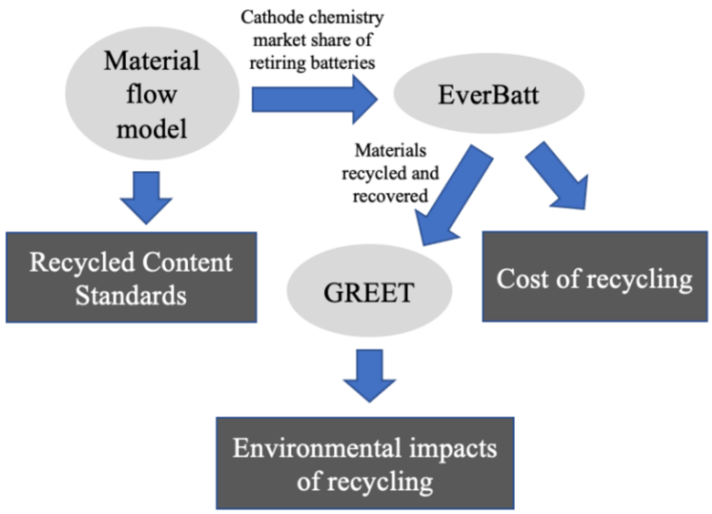


Figure 6.6: The interconnection of models used to determine the RCS, cost, and environmental impacts of recycling.

EverBatt and GREET are limited by the ability to calculate results for only one scenario at a time. To calculate results for this study, EverBatt was run iteratively 2,604 times using a macro to determine the scenario outputs. The transportation distance, mode of transportation, location of recycling, and recycling facility size were varied.

The default variable and fixed costs in EverBatt were kept (Table 6.4), except for the dollar value of materials, the labor rate of LIB disassembly in China, and the amount recyclers pay, or are paid, for LIBs at their end-of-life (i.e. the recycling fee). The labor rate for LIB disassembly is $7.50 per hour when occurring in China. This was calculated by using the ratio of US to China labor rates during the recycling phase. The recycling fee has been changed to zero for all chemistries to compare the value of recovered materials and the cost to recycle, without inflating profit or loss by including an additional transaction between the supplier and recycler. In addition, these values are removed because the source and process EverBatt used to calculate these values are not reported. This points to another limitation of the EverBatt model; not all inputs have an explanation and documented source. While these limitations exist, the models provide detailed and changeable variables that significantly aided in creating the scenarios in this analysis.

Table 6.4: The fixed and variable costs of recycling in the US and China. All values are taken from EverBatt, except for the $7.50 for direct labor for disassembly in China.

	US	*China*
Equipment cost adjustment (%)	100%	60%
Direct labor for manufacturing ($/hr)	$20.00	$3.00
Direct labor for disassembly ($/hr)	$50.00	$7.50
Electricity cost ($/kWh)	$0.07	$0.09
Natural gas cost ($/MMBTU)	$3.84	$12.00
Water cost ($/gal)	$0.01	$0.00
Landfill cost (tip fee $/ton)	$55.36	$10.00
Wastewater discharge cost ($/gal)	$0.01	$0.00

The material value used to calculate the total value of recovered materials (Table 6.5) was based on the five-year average of USGS costs (Table A in Appendix B). The cathode chemistry value was calculated using EverBatt. These values differ from Ciez and Whitacre (2019) which estimate the cost of manufacturing 1 kg of NMC111 to be $24, NCA to be $21, and LFP to be $20.

Table 6.5: The value of materials represents an average price from USGS between 2016 to 2020.

Material	*$/ kg*
Co2+ in product	59.74
Ni2+ in product	15.15
Mn2+ in product	0.28
Lithium Carbonate	16.74
Lithium Hydroxide	25.11
LCO	44.39
NMC(111)	24.67
NMC(532)	21.32
NMC(622)	22.15
NMC(811)	20.10
NCA	22.56
LMO	16.00
LFP	14.00

Location and Transportation Scenarios

Three scenarios representing the location of recycling, transportation distance, and mode of transportation are used to calculate the cost and environmental impact of recycling in EverBatt.

1) **Domestic – truck scenario:** The LIB is recycled within the US and transported via truck. The distance from end-use to collection is 50 miles, collection to disassembly is 50 miles, and from disassembly to recycler is 1,000 miles.

2) **Domestic—train scenario:** The LIB is recycled within the US and transported via train and truck (U.S. Department of Transportation, 2021). The distance from end-use to collection is 50 miles via truck, collection to disassembly is 50 miles via truck, and from disassembly to recycler is 1,000 miles via train.
3) **China—truck and ocean tanker scenario:** The LIB is transported from the US to a recycling facility in China via ocean tanker. The distance from end-use to collection is 50 miles and collection to disassembly is 50 miles, both via truck. The batteries are shipped from Los Angeles to Shanghai, China which is 19,270 nautical miles. It is assumed the battery will be trucked an average of 260 miles to the LA port and then 740 miles from Shanghai to the Hunan province where Brunp recycling is located.

Recycling facility yearly throughput

The facility throughput per year is varied from 1,000 to 10,000 metric tons (t) per year, increasing by 1,000 t increments, and then from 10,000 to 50,000 t per year, increasing by 10,000 t increments.

Disassembly

The level of disassembly is dependent on the recycling process used. Pyrometallurgical recycling can begin at the module level. Hydrometallurgical is discussed in academic literature as requiring disassembling to the cell level, although recyclers, such as Li-cycle, state their process disassembles to the module level (Karidis, 2020). This analysis assesses the economics of hydrometallurgical and pyrometallurgical recycling when disassembled to the module level. Direct recycling is still in the development stage and must be disassembled to the cell level.

In EverBatt, disassembly is modelled to be performed by hand, thus the cost consists mostly of labor. Recycling in the US is estimated to cost $50 per hour and recycling in China is estimated to be $7.50 per hour. Due to the lack of battery standardization, it is difficult to automate this step. Research is currently underway for self-learning robotics to potentially decrease the cost of this labor-intensive process (Neumann et al., 2022).

Recycling profit sensitivity analysis

Sensitivity analysis was used to evaluate the impact of recycling cost and material value on the profitability of recycling. The following inputs were both increased and decreased by 20% within the EverBatt model: value of cobalt, value of nickel, value of lithium, distance transported, hourly labor wage, and equipment cost. Due to the high volatility of commodity prices, the impact of cobalt, nickel, lithium, and manganese at their high and low prices since 2000 was additionally analyzed (Table 6.6 calculated from Table B – C in Appendix B).

The profits (or loss) of the baseline scenarios are calculated by the following equation for each year (t), recycling process (r), and cathode scenario (c):

$$value\ of\ recovered\ materials_{t,r,c} - \ cost\ of\ recycling_{t,r,c} = profit_{t,r,c} \qquad \text{Eq. 6.4}$$

The profits (or loss) from each sensitivity analysis is then calculated by the following equation for each year (t), recycling process (r), cathode scenario, and sensitivity analysis scenario (s):

$$value\ of\ recovered\ materials_{t,r,c,s} - \ cost\ of\ recycling_{t,r,c,s} = profit_{t,r,c,s} \qquad \text{Eq. 6.5}$$

The percent change demonstrates the impact of the input on the total profitability. It is calculated using the following equation:

$$\frac{profit_{t,r,c} - \ profit_{t,r,c,s}}{profit_{t,r,c}} = Percent\ change_{t,r,c,s} \qquad \text{Eq. 6.6}$$

Table 6.6: The high and low material prices used in the sensitivity analysis.

	High (kg)	*Low (kg)*
Co in product	\$112.69	\$23.79
Ni in product	\$59.20	\$9.40
Mn in product	\$0.76	\$0.16
Lithium Carbon	\$31.00	\$2.99
Lithium Hydroxide	\$31.75	\$4.49

6.4.4. Environmental impact of recycling

The environmental impacts of collection and transportation, disassembly, and recycling, were calculated with EverBatt using data from GREET (Argonne National Lab, 2021b). To calculate the environmental impacts avoided, the amount of recovered materials from 1 kg of battery recycled at end-of-life was taken from EverBatt, and the pollution from manufacturing these materials from virgin sources was calculated using GREET.

6.5. Results

6.5.1. Achievable recycled content standards

Achievable RCSs for the US are estimated to be between 11-12% for cobalt, 7-8% for lithium, and 10-12% for nickel in 2030, which then increase to 45-52%, 22-27%, and 40-46% respectively, in 2050 (Figure 6.7 and Table D in Appendix B). If the RCS for each of the scenarios is evaluated independently, the cathode chemistry forecast, sales forecast, collection rate, type of recycling, and repurposing scenarios are the large influencers (Figure A of Appendix B).

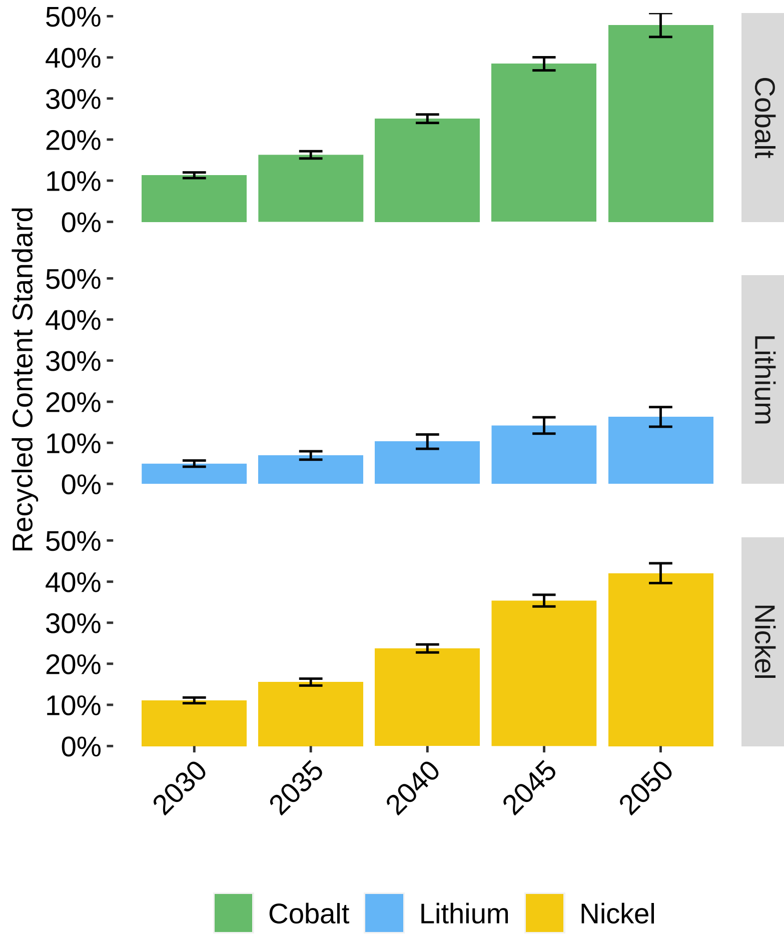


Figure 6.7: Achievable recycled content standards for lithium-ion batteries in the US. The error bars represent 95% confidence the proposed RCS.

The cathode scenarios significantly alter the estimated RCS for cobalt and nickel; the NCX dominant scenario results in a lower percentage (35-37% for cobalt and 32-34% for nickel in 2050) compared to the LFP dominant scenario (49-53% for cobalt and 43-45% for nickel in 2050). This is due to higher demand when cathodes containing cobalt and nickel are dominant (Table E in Appendix B). The sales scenarios influence the RCS for all materials, with slower future growth in material demand for the SDS scenario, which results in a higher RCS (55-58% for cobalt and 48-51% for nickel in 2050) than the STEPS scenario (32-33% for cobalt and 29-

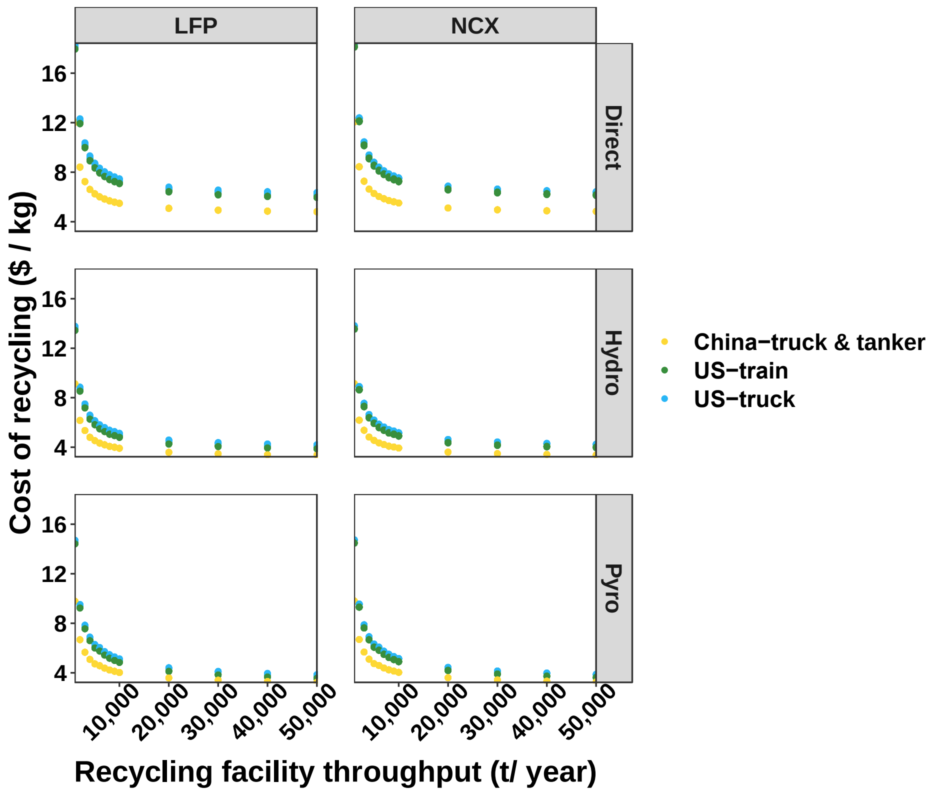
30% in 2050) (Table F in Appendix B). In addition to the sales and cathode forecasts, the collection rate determines the amount of material available for recycling; a higher collection rate results in a higher RCS.

Another important and uncertain variable in future material recovery is the mix of recycling processes in operation. Pyrometallurgical recycling does not recover lithium, and when it is removed from the RCS estimation, the confidence interval for lithium is highest (Table G in Appendix B). Due to the phasing out of pyrometallurgical recycling, it is not used to estimate achievable RCS for the US.

6.5.2. Recycling Cost

The total cost of recycling LIBs at their end-of-life includes their transportation, collection, disassembly, and recycling. The cost of recycling is highly affected by economies of scale and costs decrease exponentially until the throughput of the facility reaches ~10,000 t/year (Figure 6.8).

Figure 6.8: The cost per kg of recycled LIB in 2020 at material throughputs between 1,000 and 50,000 metric tons (t).



The location of recycling, mode of transportation, and level of disassembly also impact costs (Figure 6.9). China has a lower recycling cost, despite the added transport distance, due to a lower cost of labor and equipment costs. Figure 6.8 includes three steps of disassembly: removal from the car, disassembly to module level, and disassembly to the cell level. While all disassembly costs are included in Figure 6.9, hydrometallurgical does not always need to be disassembled further than the module level (Karidis, 2020). These high costs associated with disassembly are one of the reasons research has focused on mechanical disassembly, design for recycling, and beginning pre-treatment with the least amount of handling (Neumann et al., 2022).

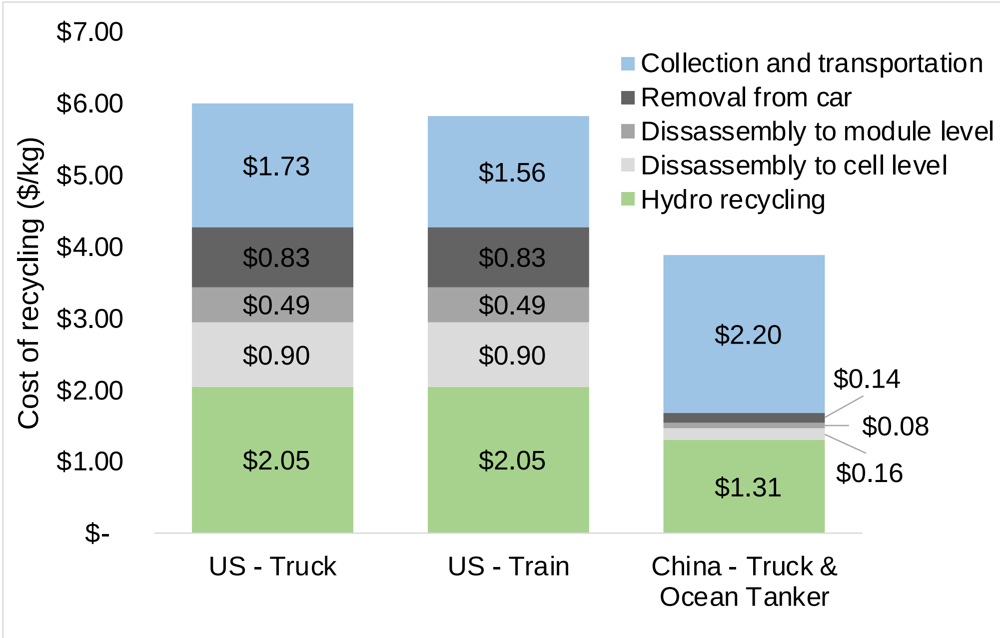


Figure 6.9: The cost ($/kg) of a hydrometallurgical recycling facility in 2020.

Recovered material value

The recovered material value is highly dependent on the materials recovered and commodity pricing. Direct recycling recovers the most value by recovering the whole cathode, reducing the need for further processing before it can be used as an input to battery manufacturing. In addition, the cathode chemistry mix of retiring materials changes the amount of high value materials such as cobalt and nickel within the batteries. Since the NCX scenario contains a constant cobalt and nickel supply, the value stays higher than the LFP scenario (Figure 6.10. In the LFP scenario, the number of cobalt-containing cathodes decreases overtime, replaced by iron, a material that is not recovered.

The economics of recycling lithium-ion batteries

All recycling processes are profitable after material throughput thresholds are met. Based on the EverBatt cost assumptions, recycling in the US in 2020 became profitable at or above ~8,000 t/year for hydrometallurgical, ~7,000 t/year for direct, and ~20,000 t/year for pyrometallurgical recycling, while using truck transportation. The location of recycling has a considerable impact to the cost of recycling due to the lower cost of labor in China. Recycling in China is profitable at throughput levels of ~3,000 t/year, ~3,000 t/year, ~4,000 t/year, respectively.

Figure 6.10 represents the cost of recycling in a 10,000 t/year facility and the material value and the cost of processing are held constant to 2020 values, which does not consider the economies of learning. This figure demonstrates the effect of evolutions in cathode chemistry; the value of recovered materials in the LFP scenario declines over time, while the NCX scenario stays relatively constant. This divergence is due to the LFP chemistry not containing the two highest valued materials, cobalt and nickel. Thus, the NCX scenario is economical for all years in the hydrometallurgical scenario, and LFP is only economical until 2028-2031. These results align with more recent analysis that demonstrate a profit from using hydrometallurgy to recycle cobalt containing chemistries (Choubey et al., 2017). These results suggest lower profitability than those documented by Choubey et al. (2017); the cost of recycling is relatively similar and the value of recovered materials in our analysis is considerably lower.

Direct recycling is profitable in the NCX scenario and is the most economical recycling process in the LFP scenario. While direct recycling is more costly, there is high value in recovering the whole cathode. This increased value leads to profitability in the LFP scenario until 2038. Despite direct recycling being the preferable choice for LFP, recycling of the LFP chemistry independently is not profitable. Xu et al. (2020) has different findings, demonstrating a net profit from recycling LFP due to the exclusion of disassembly and transportation costs.

Despite this overall higher expense for domestic processing in comparison to the recycling in China, US-based recycling can still be a profitable venture when the recycling mix includes some NMC chemistries. Cost estimates are in Table H in Appendix B.

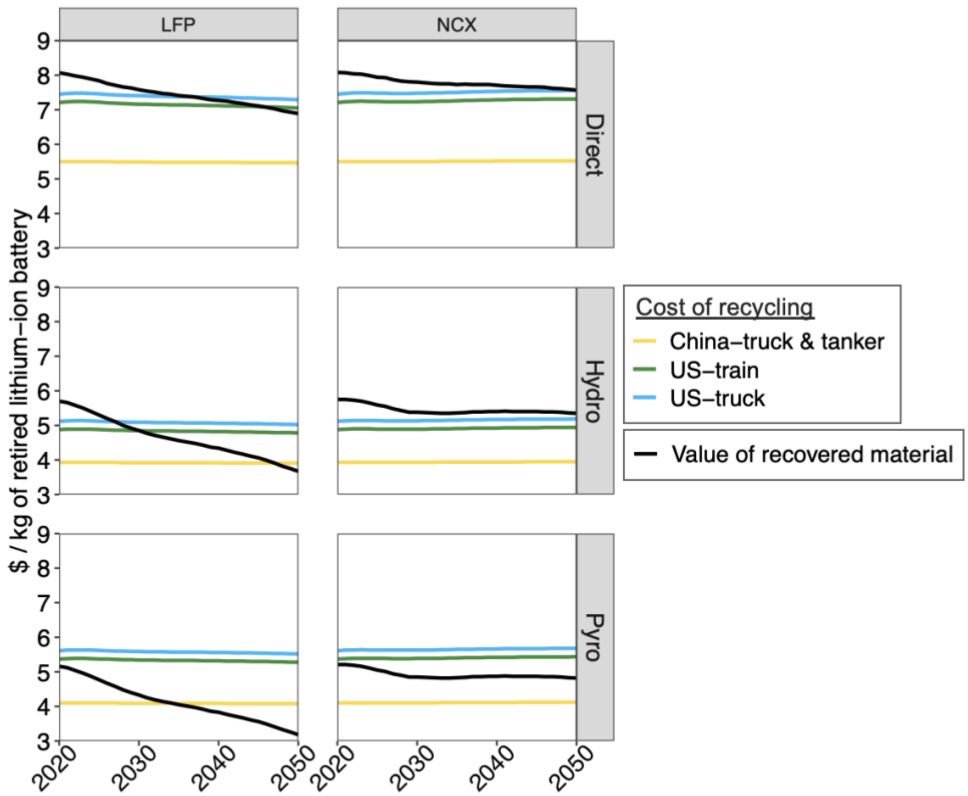
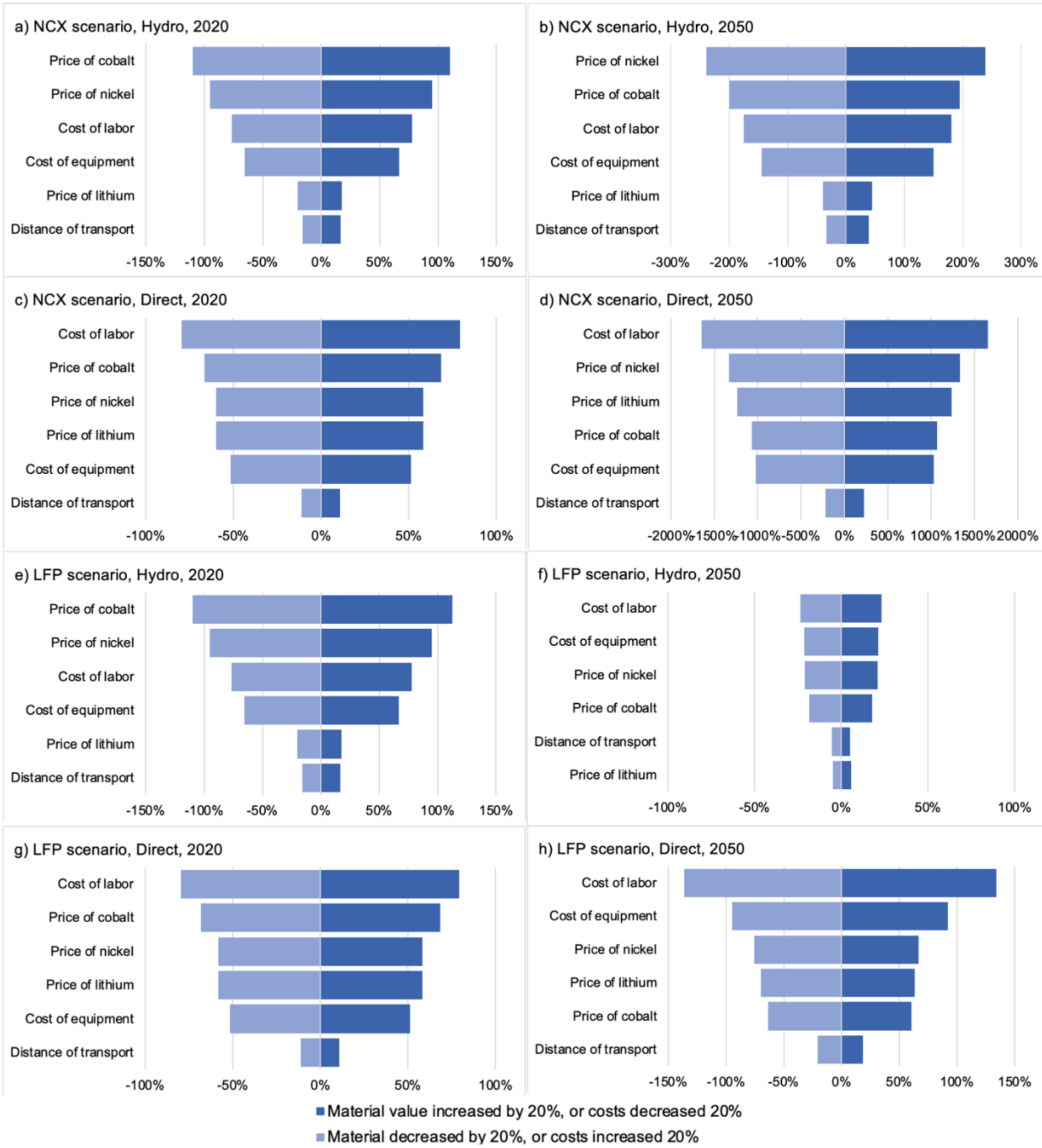


Figure 6.10: The economics of recycling lithium-ion batteries.

Sensitivity analysis

A sensitivity analysis was applied to evaluate the impact of recycling cost and material value inputs on the profitability of recycling. When inputs were both increased and decreased by 20% within the EverBatt model, the value of cobalt has the largest impact on hydrometallurgical recycling profit in 2020 and the cost of labor has the largest impact on direct recycling profit. In 2050, nickel is the most influential parameter for hydrometallurgical recycling profit in the NCX scenario. This is a direct result of the decreased use of cobalt and increased use of nickel over time in NCX chemistries. The cost of labor is the most influential parameter for all other scenarios in 2050 due to the use of manual disassembly. Figure 6.11 presents scenario results of domestic recycling with truck transportation. The x axis represents the percent change in profits or losses in $ (value of recovered materials – cost of recycling) from the baseline scenario and the y axis represents the input changed in EverBatt (Argonne National Lab, 2021a).

Figure 6.11: The recycling profit sensitivity analysis results.



In addition, the impact of modeling cobalt, nickel, lithium, and manganese at their historic high and low prices since 2000 was analyzed. Nickel had the largest impact on profitability both in 2020 and 2050 due to the historical high of $42.44 per kg in 2006. While the LFP dominant scenario has only a small amount of nickel-containing chemistry, it has a larger impact than the other materials due to the comparably higher value increase. These results demonstrate high sensitivity of the LIB recycling industry to potentially volatile commodity prices, as well as significance of the industry's increased reliance on nickel. Full results are in Table I in Appendix B and the data repository.

6.5.3. Scale of retirement in the US and infrastructure build-out

An estimated 3,000 to 10,000 t of LIB battery packs retired in 2020, too small of a quantity to support the necessary throughput for more than one facility to run at breakeven, if handling only retired EV batteries (Table 6.7). Currently, manufacturing scrap and consumer electronics are the bulk of materials processed by LIB recyclers (Carney, 2021). EV LIB retirement rapidly increases to 19,000 - 73,000 t in 2025, 71,000 - 404,000 t in 2030, and 1.2 – 8.5 million t in 2050 (Table 6.6). These estimates are larger than those previously reported by Richa et al. (2014) of 14,000 - 193,000 t in 2030 and 38,000 – 344,000 t in 2040 which only consider light-duty vehicles, assumes a smaller battery capacity of 29-51 kWh, and a lower EV sales forecast taken from the US Energy Information Administration's 2012 estimates.

Table 6.7: The pack-level retired material per year in metric tons (t). Full results in Table J of Appendix B.

Year	*Min (t/year)*	*Max (t/year)*
2020	3,172	9,537
2025	19,107	73,148
2030	70,575	403,945
2035	187,572	1,491,295
2040	393,365	3,594,527
2045	706,456	6,218,250
2050	1,153,014	8,545,553

This rapid increase in LIB retirement demonstrates that increased recycling capacity is likely necessary to support the near future LIB retirement. Publicly reported planned capacity in North America is estimated at approximately 122,000 t/years of recycling and an additional 25,000 t/year of mechanical crushing (Table K of Appendix B).

6.5.4. Environmental impacts of recycling lithium-ion batteries

As found in several previous studies, recovered material from recycling is environmentally less intensive than producing material from virgin ore (Ciez & Whitacre, 2019; J. B. Dunn et al., 2015; Mohr et al., 2020; Richa, Babbitt, & Gaustad, 2017). This analysis concludes recycling in the US results in less pollution than recycling in China (Figure 6.12) because of a shorter transportation distance and a less fossil fuel intensive electricity grid. Transportation is modelled as 1,000 miles by truck and 19,270 nautical miles by ocean tanker when recycling LIBs in China and a shorter distance of 1,000 miles by truck when recycling in the US. The electricity grid emissions factor is modelled at 724 g CO_2/kWh in China and a lower value of 426 g CO_2/kWh in the US (Argonne National Lab, 2021b)

Pyrometallurgical processing results in more CO_2e emissions than the other recycling technologies, while hydrometallurgy results in higher SO_X emissions. Direct and hydrometallurgical recycling recover more material than pyrometallurgical, thus offsetting more virgin material and associated emissions. This is contradictory to the results found by Richa et al. (2017), which showed hydrometallurgical recycling to offset less emissions. The difference is primarily due to Richa et al.'s assumption that manganese is not recovered by hydrometallurgical processing. Ciez and Whitacre (2019) show pyrometallurgical recycling and the recycling of LFP do not result in avoided emissions. Differences among their study and this one are largely due to their scope, which includes the manufacturing of the whole cell, while this analysis only accounts for the emissions of manufacturing the recovered materials.

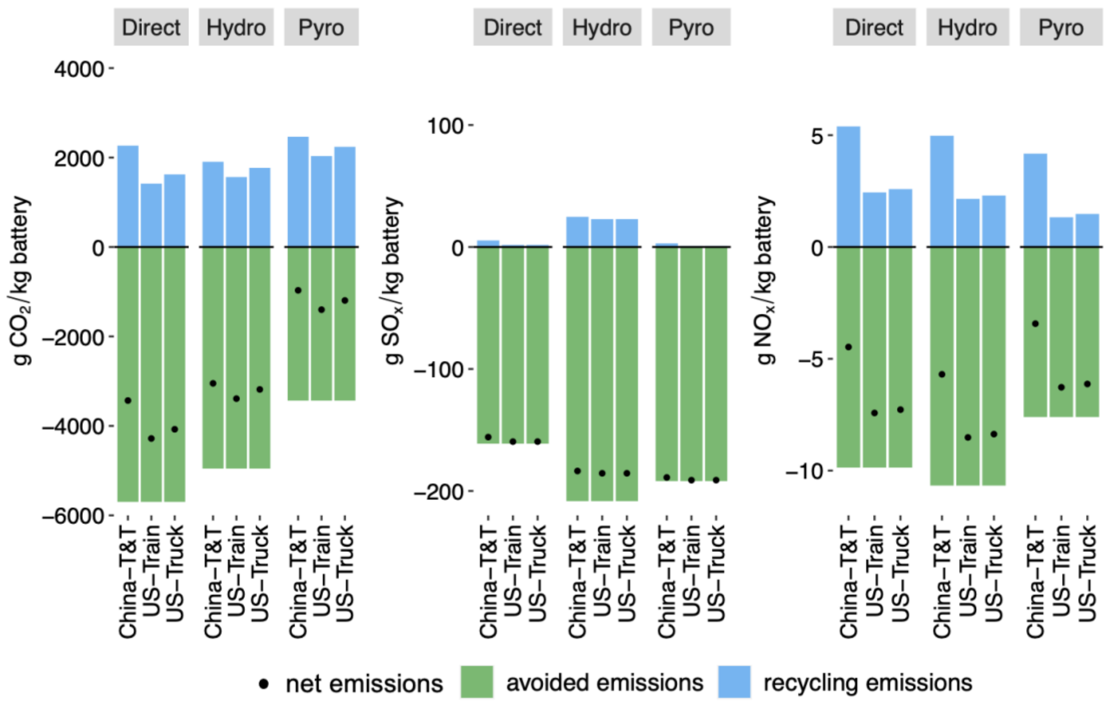


Figure 6.12: The environmental impact of recycling lithium-ion batteries in 2020.

The avoided emissions in Figure 6.12 represent the environmental impacts if the materials recovered were from virgin ore (green), the recycling emissions are the emissions resulting from the recycling process (blue), and the net emissions represent the emissions saved because materials were recycled instead of mined. The x-axis represents the location and transportation scenarios. Recycling in China and transporting via truck and ocean tanker is abbreviated to "China- T&T".

6.6. Limitations of study

The impact of emerging technologies such as solid-state, lithium-sulfur, or sodium-ion batteries was not included in this analysis. If the dominant technology changes, the relevancy and level of RCS will need to be reconsidered. For example, solid-state batteries which are currently under development, would drastically decrease the demand for critical materials, therefore the current retiring supply would provide recovered materials for a higher feasible RCS (Watanabe et al., 2019). This is not the case for lithium, if it replaces graphite in the anode for solid-state batteries. In addition, the recycling processes will likely require modification. In the case of solid-state batteries, even if the same materials for recovery are desired, modifications are required due to the presence of a solid electrolyte such as glass or ceramic (Schwich et al., 2020).

We also do not include the potential impact of recycled materials from other product systems; for example, consumer electronics or stationary storage. While the LIB market is historically dominated by consumer electronic sales, and therefore is currently the bulk of retired supply, EV sales are now the large majority (Bloomberg New Energy Finance (BNEF), 2019). If consumer electronics were included in this analysis, they would likely increase the cobalt RCS due to lithium cobalt oxide being the common cathode chemistry (Fu et al., 2020; Gaines et al., 2021). Stationary storage is currently a comparably small market, equal to an estimated 3% (1,688 MWh) of total EV capacity in 2019 (U.S. Energy Information Administration, 2021). This is forecasted to increase along with EVs to be equal to about 3-5% of total EV sales until 2050 (664 GWh for 2-6 hour storage) (Frazier et al., 2020).

Lastly, the purity of material recovered impacts the value of materials, and recent studies show the shredding versus disassembling reduces purity, thus changing the economics (Thompson et al., 2021). While this is an important finding, it has not been considered in this analysis.

6.7. Discussion

6.7.1. RCS and other international policies to increase material circularity

This research calculates feasible RCSs in the US to be 11-12% for cobalt, 7-8% for lithium, and 10-11% for nickel in 2030, which increase to 15-18% for cobalt, 9-11% for lithium, and 15-17%

for nickel in 2035. These are slightly different from the proposed EU standards of 12% for cobalt, 4% for lithium, and 4% for nickel in 2030, and 20%, 10%, and 12%, respectively, in 2035. The variance between regions is likely due to political considerations and different calculation inputs. To reach the EU RCS for lithium, a process which recovers the material must be used. This indicates a push away from pyrometallurgical and towards hydrometallurgical and direct recycling. Future recycled lithium availability will depend on whether hydrometallurgical continues to be the dominant technology. Cathode chemistry trends will likely determine if battery manufacturers prefer cathodes recovered through direct recycling or their constituent materials through hydrometallurgical recycling. Trends towards LFP will likely indicate direct recycling is preferable due to the constituent materials having relatively low values. Both processes are desirable in a trend towards NMC811 due to the high value of materials and the ability to upcycle. The high cobalt chemistries currently phasing out, such as NMC111, are upcycled by adding nickel to achieve a lower ratio of cobalt in the recovered cathode (Gaines & Wang, 2021).

In addition to RCS, the EU has proposed several policy mechanisms to achieve a circular economy, including extended producer responsibility, collection rates, material recovery rates, and emission requirements. The proposed EU collection rates are those used in this analysis, at 65% in 2025 and 75% in 2030 (European Commission, 2020). Considering the EU is the second largest EV market in the world, these policies will impact EV manufacturers globally (Melin et al., 2021). If the US does not implement similar requirements, the battery and material suppliers unwilling to reduce social and environmental impacts may divert their attention to sales in the US, while the companies focused on a sustainable supply chain may focus their efforts on the EU. Policy harmonization across regions could engender a global shift in supply chain and manufacturing requirements, thus positively decreasing the regulatory uncertainty for manufacturers.

There are other policy mechanisms that can also increase battery circularity. Government subsidization of recycling matched with a recycling requirement has been demonstrated by China, resulting in the growth of the industry (International Energy Agency, 2020). Another route for increasing recycling is creating a collection and recycling program funded by environmental collection fees charged at the time of sale. This has not been demonstrated for any LIB recycling program, although is a solution for e-waste in California (California Department of Tax and Fee Administration, 2021).

6.7.2. Closed-loop recycling assumptions and real-world demonstrations

The potential circularity reported in this analysis assumes closed-loop recycling, in which battery materials recovered from LIB waste are used in manufacturing LIBs. While RCS may encourage a circular economy on a global scale, it will only contribute to a domestic circular economy if manufacturing is done domestically. The current lack of cathode manufacturing in the US means recovered materials may be exported or used by other industries.

Due to the criticality of these materials, it is advantageous for the US to develop a domestic cathode manufacturing industry and thus increase material security. Northvolt, located in Sweden, has a closed-loop system which combines LIB recycling and manufacturing (Northvolt, 2021). Within the US, Redwood Materials, a hydrometallurgical recycler, recently announced they are building capacity to manufacture cathode and copper foil from recovered materials, which will be sold to Panasonic, the battery supplier to Tesla (Carney, 2021; Korosec, 2022). If other US-based recyclers and EV manufacturers follow suit, the industry has the potential to create a closed-loop system for secondary material generated from retired LIBs.

6.7.3. LIB recycling economics and global material flows

This analysis shows recycling in China is less expensive than in the US, although domestic recycling in the US is still profitable at economies of scale (Table 6.1). The known recycling facilities planned for development all have capacities over the calculated economical threshold in this paper, excluding pilot facilities. Not previously discussed in this analysis is the spoke and hub model, a method used by Li-cycle. In this method, LIBs are shredded at a smaller facility (5,000 t/year spokes), then aggregated at a larger facility (60,000 t/year hub) which performs hydrometallurgical recycling.

The cathode chemistry, and specifically the amount of cobalt, also significantly affects the economics of recycling. The sensitivity analysis demonstrates when cobalt batteries are a significant portion of the waste stream, the value of cobalt is the largest influencer of profits. As the portion of cobalt declines, the value of nickel and the cost of labor becomes more influential. This is especially important considering recent warnings of future class 1 nickel shortages due a lack of necessary processing capacity to support rapidly increasing demand from LIBs (Campagnol et al., 2017; The White House, 2021).

While the cost of recycling in the US is higher than recycling in China, domestic recycling results in lower emissions due to decreased transportation and a cleaner electricity source. The uptake of EVs is based on the need to reduce climate change and emissions from transportation, therefore it is essential to apply those principles to the End-of-life as well. One opportunity to achieve lower impact and cost is mode shifting from truck to train transportation. LIB End-of-life transportation by train is not common practice, but it does result in the least environmental impact and also reduces transportation costs.

6.8. Conclusion

This research calculates achievable RCSs for the US that can support decision-making in the policymaking process. The analysis finds recycling is economical in the near term. While domestic recycling is ideal to increase energy material security and lower the life cycle environmental impact of these materials, the recycling of LIBs in China is less expensive than in the US. The LIB recycling facility capacity in the US will also have to rapidly increase to support future retirements, if domestic recycle of EV batteries is a priority. To ensure that recovered materials stay domestic, recycling facility development must also be coupled with the development of cathode and battery manufacturing capacity within the US (The White House, 2021). Therefore, policy is likely necessary to ensure the market does not result in exporting of retired batteries or their critical materials.

6.9. Data availability

The code and datasets used to generate these results are made available in the Dryad repository: https://datadryad.org/stash/share/X_DM7T32Z5pJPlaB1ibqXBjAxZL4BU3JxmNgDmC7nbI

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7. Should high cobalt EV batteries be repurposed? Assessing the impact of technological innovation on the waste hierarchy.

7.1. Scope and purpose

Recovered materials from the retiring stock of LIBs can contribute to a substantial portion of EV material demand, therefore decreasing the manufacturing impacts from EVs. Section 5 and 6 reviewed the lithium-ion battery (LIB) circularity potential, recycling economics, and recycling emissions. This previous research was completed under the assumption that it is preferable to repurpose a battery before recycling. This section assesses if the impact of new technology will change the preferable end of life route of recycling prior to reuse. It does so by comparing environmental tradeoffs between reusing a battery at the end of life, versus immediately recycling that battery to then manufacture a more technologically advanced battery with recycled content.

This section is adapted from a paper which will be submitted to The Journal of Industrial Ecology:

Dunn, J., Ritter, K., Kendall, A., Velázquez, J. Should high cobalt EV batteries be repurposed? Assessing the impact of technological innovation on the waste hierarchy.[c]

7.2. Introduction

As previously discussed, the electrification of transportation, enabled by the LIB, has the potential to drastically decrease carbon emissions (Rogelj et al., 2018). Life cycle assessment

[c] Jessica Dunn contributed through conceptualization, software, formal analysis, writing – original draft; Kabian Ritter contributed through formal analysis; Alissa Kendall contributed through supervision, funding acquisition, writing – review; Jesus Velázquez contributed through supervision, writing – review

(LCA) has been used to analyze the environmental impact of electric vehicles (EVs) to understand whether eliminating tailpipe emissions leads to real reductions in emissions on a life cycle basis, and to identify life cycle stages and materials that are hotspots of environmental impact (Bauer et al., 2015; Pero et al., 2018). Previous LCAs have demonstrated benefits are highly dependent on the source of electricity, the cathode chemistry used, and the miles travelled (Archsmith et al., 2015). Second to electricity generation, except in cases of low-carbon electricity, the upstream production of battery materials represents the majority of impacts. Specifically, impacts from the cathode active material, aluminum, and energy for cell production (Dai et al., 2019).

In addition to environmental impacts from battery production, research has demonstrated the social impacts related to the mining of materials are detrimental to the wellbeing of miners and the surrounding communities. In particular, the mining of cobalt is associated with many human rights abuses, including the exploitation of child labor (Sovacool, 2019). Batteries are being designed with less cobalt because of these associated impacts, the susceptibility to geopolitical risk due to the geographical concentration in the Democratic Republic of Congo, and the increasing material prices (International Energy Agency, 2020; Sovacool, 2019; The White House, 2021).

Impacts from battery manufacturing and concerns over resource constraints will continue to increase with LIB demand. As these LIBs age, a wave of batteries will begin retiring, and governments and industries are attempting to figure out the best use for this retired supply. The waste hierarchy is a guiding principle that has long been used by the European Union and United States Environmental Protection Agency as a prioritization framework for handling waste. This hierarchy is now being applied to retired LIBs and is as follows: prevent, reduce, reuse, recycle, recover, and dispose (Environmental Protection Agency, 2021; European Commission, n.d.). The order is based on the ability of processes to save the most resources and result in the least environmental impact (van Ewijk & Stegemann, 2016). The benefits of repurposing LIBs for stationary storage prior to recycling has been validated by Richa et al. (2017) in an LCA specifically reviewing the waste hierarchy and circularity, as well as in other LCAs more broadly focused on LIB second-use applications (Bobba et al., 2018; Casals et al., 2017; Cusenza et al., 2019; Richa et al., 2015).

Despite these earlier findings, the waste hierarchy for end-of-life LIBs has recently been questioned due to the rapidly changing LIB technology (Emma Wiesner & NorthVolt battery, 2020; Harper et al., 2019). Previous studies do not consider that the hierarchy may not apply under these changing technology conditions. For example, cathode chemistries are being designed with lower cobalt content and forecasts show that if the low cobalt chemistry becomes the dominate cathode, cobalt circularity of 80% could be achieved within the battery ecosystem in 2040, in comparison to 50% circularity with the current cathode market share (J. Dunn et al., 2021). This is the direct result of a high cobalt cathode battery (NMC111) being able to supply enough cobalt to produce four low cobalt batteries (NMC811). In addition, batteries using silicon (Si) instead of graphite (Gr) in the anode are in development, resulting in a potentially large increase in energy density.

The overall lower material demand per kWh for new batteries could offset future demand and decrease long-term extraction (Dunn et al., 2021). An LCA is needed of waste hierarchy pathways for batteries being retired in order to understand whether the reduced cobalt chemistries and changing anode material might change the preferred waste management option.

This research will review the environmental impact of this technological innovation to evaluate if recycling should be prioritized for these high cobalt batteries, or if their lifespan should be extended through repurposing. An assessment of the environmental impacts cannot provide a full view of the tradeoffs, considering it does not include social impacts and the economics, although this analysis will provide partial insight into the optimal waste hierarchy. It will also contribute to a greater discussion around the evaluation and potential flexibility of the waste hierarchy.

7.2.1. Literature review

Lithium-ion battery manufacturing life cycle assessments

A literature review of LIB LCA articles by Peters et al. (2017) found that between 2000 to 2016 there were a total of 36 LCAs that provide a detailed outline of their LIB production modeling. The reviewed LCAs examine various cathode chemistries, including LFP, LCO, NCA, and NMC, but do not detail the stoichiometry of the NMC chemistry studied (i.e, NMC111, NMC523, NMC622, and NMC811). Out of the 36 studies, five compare cathode chemistry impacts. Notably, Ambrose and Kendall (2016) compare NCA, NMC, LMO, LFP, and LMO/LTO, demonstrating chemistries containing cobalt have a higher percent contribution to battery production GHG emissions.

Since Peters et al.'s literature review, a number of new LCAs have been published which have explored the use of a silicon anode. Kallitsis et al. (2020) analyzed the impact of varying NMC stoichiometries and the substitution of a Gr anode with a Gr and Si mix, finding the mixed anode results in a 40-50% decrease in all ecotoxicity 2008 ReCiPe midpoint categories. Si is a hopeful replacement for Gr because it has a higher theoretical capacity and can potentially increase the energy density of LIBs. These batteries are currently at pilot scale and plagued with the issue of anode volume change when cycling, leading to cracking and early failure of the battery.

Beyond the Si and Gr mix, there are various types of Si anodes in development to accommodate the issue of volume expansion. Nanostructured anodes, including Si nanotubes and nanowires, are more porous than the typical Gr anode and considered to be a promising technology (Zuo et al., 2017). The downfall of these anodes includes the added required area, and thus the loss of energy density provided by silicon (Piwko et al., 2017). Recent LCAs have found in some cases these anodes increase the environmental impacts due to 1) this lack of density improvement, and 2) the impact of process silicon into a nanostructured anode (Wu & Kong, 2018). Deng et al. (2019) found a lithium-ion battery with a silicon nanotube anode has potential benefits if produced at industrial scale, but at current scale is still lacking. While Wu and Kong (2018) found Si nanowire anodes result in the higher impacts than Gr and lithium metal due to the processing of Si powder into silicon nanowire. Columnar Si thin film anode appears to be a better option due to its high specific capacity and ability to have a higher anode to cathode ratio (Piwko et al., 2017). In this study we model the columnar Si thin film anode due to the potential life cycle environmental benefits it can potentially provide over the nanostructured silicon anodes.

Lithium-ion battery end-of-life life cycle assessments

The repurposing and remanufacturing of LIBs have been evaluated by a plethora of LCAs, demonstrating that in most cases it is environmentally advantageous to extend the lifespan of the product before end-of-life disposal (Bobba et al., 2018; Casals et al., 2017; Cicconi et al., 2012; Cusenza et al., 2019; Faria et al., 2014; Genikomsakis et al., 2014; Richa, Babbitt, & Gaustad, 2017; Richa et al., 2015; Sathre et al., 2015). The LCA results vary depending on the reference scenario, second-life use, allocation factor, and other assumptions made in the analysis, such as lifespan. Various second-life storage applications have been analyzed, including solar PV system support (Bobba et al., 2018; Kamath, Shukla, et al., 2020), load shifting (Sathre et al., 2015), peak shaving (Berzi et al., 2020; Faria et al., 2014; Kamath, Shukla, et al., 2020; Sathre et al., 2015), and EV fast-charging stations (Kamath, Arsenault, et al., 2020). Sathre et al. (2015) found that when the storage is used for diurnal energy shifting it enables an increase of renewables on the grid. Richa et al (2015) did a similar analysis and found the load shifting results in a reduced net cumulative energy demand due to the ability to store renewable energy instead of curtailing it (Richa et al., 2015). Several authors have also pointed out that second-life applications that support carbon-intensive fuels increase overall carbon emissions, and are not advised (Bobba et al., 2018; Casals et al., 2017).

In the waste hierarchy, after the battery has been used to its fullest extent through reuse and repurposing, the recycling of LIBs is a preferred alternative to landfill disposal. Pyrometallurgy is the most energy-intensive recycling technique, resulting in a higher environmental impact than the alternatives of hydrometallurgy and direct cathode recycling (Anwani et al., 2020; Richa, Babbitt, & Gaustad, 2017). These processes also differ in their material recovery; the pyrometallurgical process cannot recover lithium, while the hydrometallurgical process does. Direct recycling, also called direct cathode recovery, recovers the cathode active materials through physical separation, and after the lithium is replenished, can be reused in the manufacturing of new batteries (Gaines et al., 2018; Harper et al., 2019). Some studies have demonstrated that using pyrometallurgical recycling instead of disposal results in no reduction, or even an increase, of lifecycle GHG emissions depending on the carbon intensity of the energy used in the process (Ciez & Whitacre, 2019). While this is important to consider when choosing a preferable recycling technique, it is also important to note that all recycling processes result in a large decrease of ecotoxicity and material depletion potential due to avoided raw material production (Richa et al., 2015).

7.2.2. Contribution to literature

The literature review shows an extensive body of LCA work spanning all life cycle phases of the LIB (production, use in an EV, repurposing, second-life use, and recycling). However, the influence of technological innovation on the tradeoff between repurposing and recycling has not been explored by LCA. Kamath et al. (2020) also identified this gap in the literature, stating that repurposing may divert flows of end-of-life batteries away from recycling, and the relationship between recycling and repurposing needs to be evaluated. In addition, Harper et al. (2019) notes that American Manganese, a LIB recycling company, has asserted that high cobalt chemistries should be sent directly to recycling at the end of their first life to boost cobalt supplies (Moonshot Exec, 2018). Other recycling companies have echoed this idea, maintaining that materials should be recycled directly after their first use to produce low cobalt chemistry batteries (Emma Wiesner & NorthVolt battery, 2020).

This research will address this gap in the literature by evaluating the environmentally preferable end-of-life route for high cobalt chemistries. This analysis is needed now, because a large portion of batteries containing NMC111 cathode chemistry will be retiring in the coming years, while the batteries manufactured to replace them and grow the market will be lower cobalt chemistries like NMC811 (Dunn et al., 2021). This research measures the life cycle environmental tradeoff between two end-of-life management routes; (i) recycling high cobalt chemistry batteries immediately after first use to produce new, less material-intense batteries, and (ii) repurposing high cobalt chemistries for second-use applications followed by recycling.

7.3. Methods

LCA evaluates environmental impacts throughout the life cycle of a product and throughout the supply chains that support a product system. The LCA conducted in this research largely conforms to ISO 14040 standards, and the following sections step through the three primary phases of the LCA methodology; goal and scope definition, life cycle inventory analysis (LCI), life cycle impact assessment (LCIA), and interpretation of the results.

7.3.1. Goal and Scope definition

This study is a comparative LCA with the goal of identifying if it is environmentally preferable to extend the life of a high cobalt battery, or recycle it after first use. The intended audience includes policymakers, who are responsible for legislating and recommending end-of-life pathways, as well as parties responsible for retired batteries that will have to decide if the battery should be repurposed or recycled.

The service provided by these two pathways is provision of stationary storage services for electricity produced by solar photovoltaic panels. The functional unit is 1 kWh of cycled electricity through an LIB in a stationary storage application. The system boundary of this analysis starts with a retired EV battery; thus, the production of the EV battery is outside the scope of analysis and extends through to final disposition of the LIBs via recycling. The compared pathways are modeled as follows (Figure 7.1):

> *Recycling Pathway:* An LIB containing a high cobalt cathode chemistry is retired from use in an EV at 80% capacity and then sent directly to hydrometallurgical recycling. A LIB containing a lower-cobalt chemistry is manufactured for stationary storage use. When the LIB manufactured for stationary storage reaches 50% capacity, the battery is recycled using hydrometallurgical processing.

> *Repurposing Pathway:* An LIB containing a high cobalt cathode chemistry is retired from use in an EV at 80% capacity. It is then used in a second-life stationary storage application. When the LIB reaches 50% capacity, the battery is recycled using hydrometallurgical processing.

In the Repurposing Pathway, the battery is repurposed as a stationary energy storage system prior to recycling, thus providing an additional product function compared to the Recycling Pathway, where the retired EV battery is immediately recycled. Repurposing the battery entails 1) transporting the used LIB to a new location for an estimated 1000 km; 2) removal from the EV, which could occur before or after transportation; 3) testing the state of health through a full charge and discharge; and 4) the connecting of packs together to create a larger system, including the connection of the packs with an HVAC and computer system. Since the new stationary storage LIB also requires an external stationary storage casing, connecting busbars, HVAC, wiring, and a computer system, these are outside the system boundary, similar to the approach taken by Le Varlet et al. (2020).

For the compared pathways, the service provided must be identical. Therefore, in the Recycling Pathway, a new LIB stationary storage system is included within the system boundary to deliver stationary storage services.

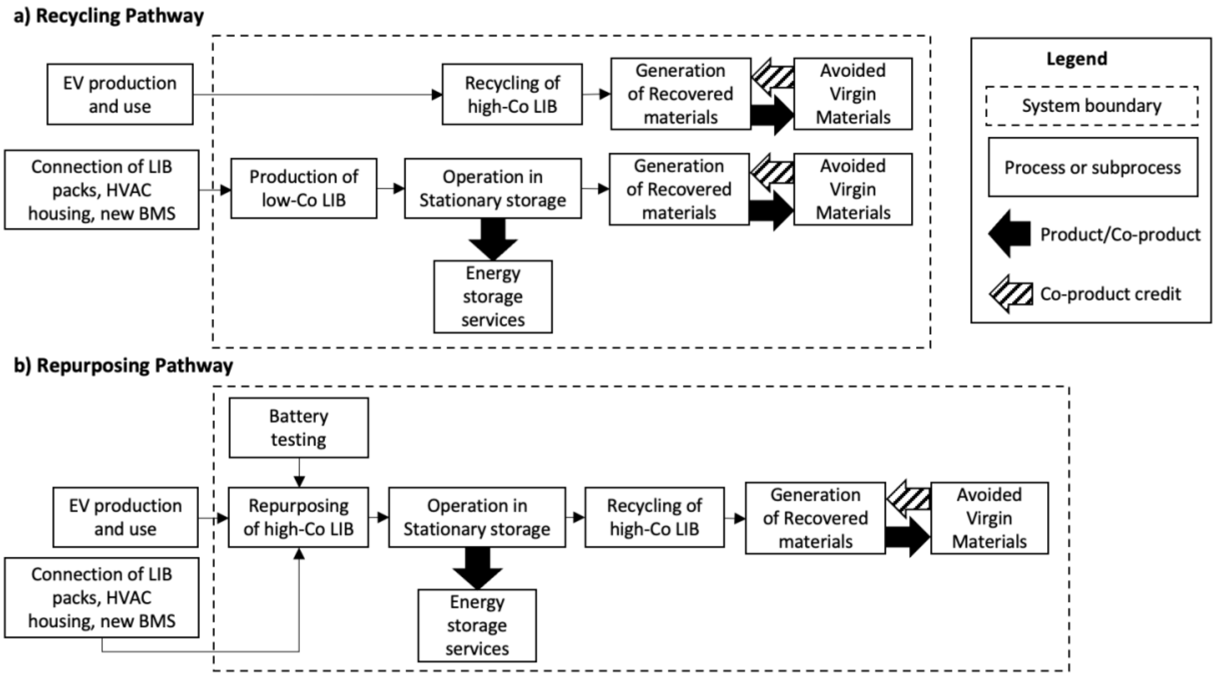

Figure 7.1: The two pathways modeled in this LCA. a) the Recycling Pathway and b) the Repurposing Pathway.

Scenario development

The future dominance of different LIB cathode materials, anode materials, and recycled content, is uncertain. This will affect the characteristics of future LIB stationary storage systems that will be manufactured. In addition, current and previously manufactured EV LIBs have taken on a number of different chemistries, even within the NMC family of LIB chemistries, making the chemistry of a retired EV LIB uncertain as well. To address these uncertainties, scenario analysis is used to examine the effect of potential technology evolution for the new stationary storage LIB manufactured in the Recycling Pathway, as well as the chemistry of the retired EV battery. There are a total of 24 scenarios (Table 7.1): four scenarios vary the cathode chemistry of the retired EV LIB and the chemistry of the new LIB manufactured; two scenarios vary the anode material of the new LIB manufactured (for stationary storage); and three scenarios vary the level of recycled content in the new manufactured LIB. For the Repurposing Pathway, there are a total of three scenarios which represent different LIB cathode chemistries.

Table 7.1: The total scenarios used in the comparative LCA.

Recycling at end-of-first life scenarios	
Cathode	Scenario 1: NMC111 battery recycled, NMC811 produced Scenario 2: NMC111 battery recycled, NMC622 produced Scenario 3: NMC622 battery recycled, NMC811 produced Scenario 4: NMC111 battery recycled, NMC111 produced
Anode	Scenario Gr: LIB produced with graphite anode Scenario Si: LIB produced with columnar silicon thin film anode
Recycled content	Scenario A: 100% virgin material Scenario B: 50% recycled cobalt, nickel, manganese, lithium, and copper material Scenario C: 100% recycled cobalt, nickel, manganese, lithium, and copper material
Repurposing at End-of-life scenarios	
Cathode	Scenario D5: NMC111 battery Scenario D6: NMC622 battery Scenario D7: NMC811 battery

7.3.2. Life Cycle Inventory Analysis

Lithium-ion battery life cycle inventory

Life cycle inventory (LCI) development began with modeling of an LIB with a NMC111 cathode and a Gr anode (NMC111-Gr). The reference LCI for this LIB is from Kallitsis et al. (2020), which updated an LCI by Ellingsen et al. (2014) with newer reference datasets from Ecoinvent version 3.5. The NMC111-Gr battery modelled is 253 kg, 26.6 kWh and has a 95% round trip efficiency.

Altering the cathode LCI

The cathode and anode scenarios in Table 1 were created by altering the $N_xM_yC_z$ ratio and anode material of NMC111-Gr. The six cases studied are: NMC111-Gr, NMC622-Gr, NMC811-Gr, NMC111-Si, NMC622-Si, and NMC811-Si. All dimensions and mass of the cell are constant, except for the active electrode material. This simplifying assumption, also used by Kallitsis et al. (2020), allows consideration of the effects of changing the anode and cathode in isolation of other changes.

To calculate the amount of nickel, manganese, and cobalt for each cathode chemistry scenario, the mass for each cathode chemistry, NMC111, NMC622, and NMC811, is calculated. Due to their compositional differences each mass will vary slightly and is calculated using Equation 7.1:

$$N_xM_yC_z = LiNi_xMn_yCo_z(O_2) \qquad \text{Eq. 7.1}$$

Using these values for the required mass of precursors, the inventories of the unique NMC chemistries were calculated based on the well-studied co-precipitation reaction (Equation 7.2):

$$x\ NiSO_4 + y\ CoSO_4 + z\ MnSO_4 + 2\ NaOH \rightarrow Ni_xCo_yMn_z(OH)_2 + Na_2SO_4 \qquad \text{Eq. 7.2}$$

Altering the anode LCI

A major consideration in calculating the anode mass is the theoretical capacity difference between Gr and Si. Si has a theoretical capacity that is an order magnitude higher than Gr (~3700 mAh g^{-1} and 372 mAh g^{-1} respectively), a major benefit of transitioning from a Gr to columnar Si thin film anode. This capacity difference translates to less active material needed in the anode to deliver the same energy density (Baasner et al., 2020).

As a result, to translate the base case configuration, NMC111-Gr to NMC111-Si, a simplifying assumption is applied: the mass of Si required to deliver the same theoretical capacity as Gr is lower by an order of magnitude. In addition, the density difference between Gr and Si is assumed to be negligible (2.27 and 2.33 g cm^{-3} respectively), and thus is not considered. This calculation process uses the theoretical capacity (TC) and mass loading (ML) of Gr and Si, as illustrated in Equation(s) 7.3 and 7.4:

$$TC_{Gr}\ (mAh\ g^{1-}) \times ML_{Gr}(\mathrm{g}\ cm^{2-}) = TC_{Si}\ (mAh\ g^{1-}) \times ML_{Si}(\mathrm{g}\ cm^{2-}) \qquad \text{Eq. 7.3}$$

$$ML_{Si}(\mathrm{g}\ cm^{2-}) \times \text{NMC111 Gr area}\ (cm^2) = Mass\ of\ Si\ (g) \qquad \text{Eq. 7.4}$$

This mass of Si, calculated in Equation 7.4, is then scaled for pack level analysis and the mass of cathode material is adjusted to maintain the 1.2 N/P ratio typical of Si. Finally, both masses were normalized to maintain overall pack level mass.

The manufacturing process for columnar Si thin film anode was also modified in the LCI. Manufacturing includes depositing Si on a roughened copper foil current collector, and then a laser is used to etch block wise structures in the Si (Piwko et al., 2017).

Due to the energy density variance, the capacity of the battery is calculated using equations from a model by Wentker et al. (2019) and in Section S1 of Appendix C. In addition, the LCI of all batteries modelled in this study can be found in Tables S2-1 to S2-19 in Appendix C, and a detailed explanation of stoichiometry calculations in Section S3.

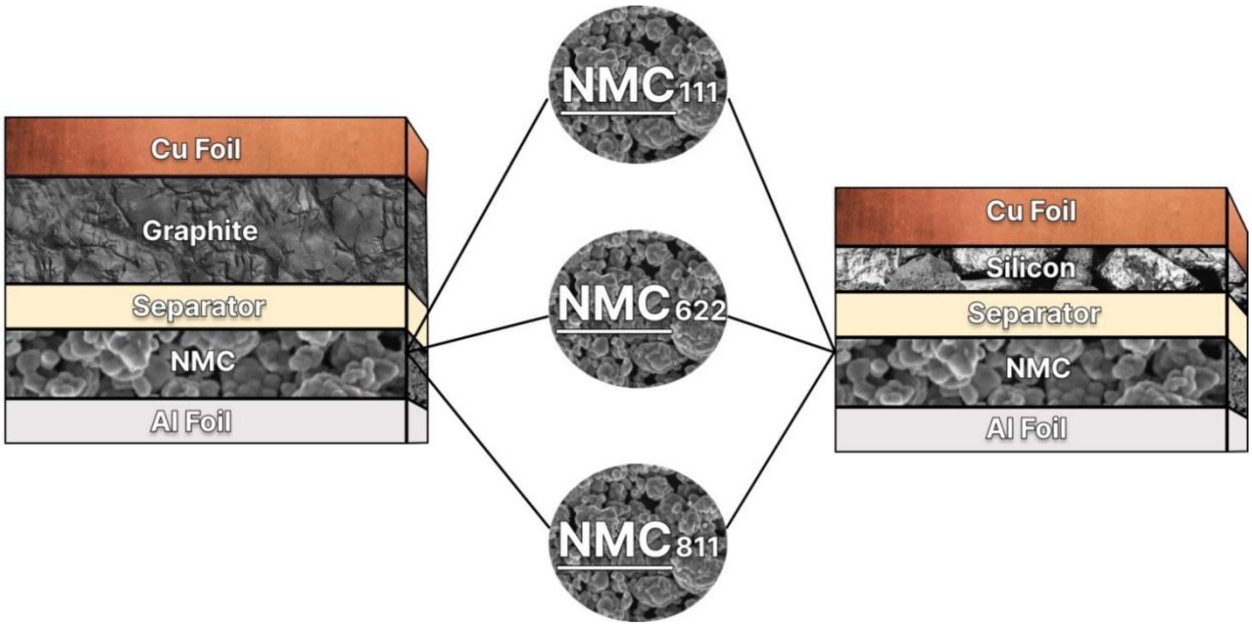

Figure 7.2: Visual depiction of the 6 cell chemistries: NMC111-Gr, NMC622-Gr, NMC811-Gr, NMC111-Si, NMC622-Si, and NC811-Si.

Stationary storage use-phase inventory

The use-phase of the stationary storage LIB consists of charging and discharging electricity generated by a 530 kWh solar photovoltaic panel in the Western Electricity Coordinating Council (WECC) grid region of the United States. The delivered electricity is calculated by using the rated capacity of the battery (C_r), capacity at the current cycle *y* (C_y), 95% depth of discharge (DoD), and efficiency at the current cycle *y* (E_y). Equation 7.5 represents the kWh delivered (discharged) and Equation 7.6 represents the kWh consumed. The two equations differ by the efficiency, which decreases as the battery ages, thus requiring more kWh consumed per kWh discharged.

$$kWh\ delivered = \sum_y^c (C_r * C_y * DoD * E_y) \qquad \text{Eq. 7.5}$$

$$kWh\ consumed = \sum_{y}^{c}(C_r * C_y * DoD) \quad \text{Eq. 7.6}$$

Degradation of the battery over the lifespan is modelled using a capacity and energy efficiency fade. Capacity fade is modelled using Equation 7.7 from Yang et al (2018) and used to calculate C_y. The degradation rate is linear, until it decreases rapidly due to a "knee" at ~70%. The battery is taken to 50% capacity due to this being the lowest the equation can accurately predict to. The input parameters are in Table 7.2 and represent cell 1 in the study by Yang et al. (2018).

$$C_y = \alpha_1 + \alpha_2 * \ln(y + m) + \alpha_3 * \ln(1 - \alpha_3 * y) \quad \text{Eq. 7.7}$$

Table 7.2: the input parameters into equation S17.

Parameters	*Inputs*
α_1	1.1978
α_2	-0.0365
α_3	0.1309
α_4	8.93E-04
m	200

The energy efficiency fade is modelled as a linear decline, similar to Richa et al. (2017) and Ahmadi et al. (2014), to calculate E_y in Equation 7.5. Efficiency fade is correlated with capacity fade, and batteries are expected to be retired from first use at 80% capacity and 80% efficiency (Ahmadi et al., 2014). The rate of efficiency decline per cycle (D_y) is calculated by determining the slope of the linear equation, when a battery has reached 80% degradation. Since capacity and efficiency reach approximately 80% at the same cycle, this is determined to be at 756 cycles based on Equation 7.7, resulting in a capacity fade rate of 1.98E-4 using Equation 7.8. Richa et al (2017) used a similar approach and determined a capacity fade rate of 5.13E-5.

$$.80 = .95 - D_y(756) \quad \text{Eq. 7.8}$$

The Repurposing Pathway has a substantially lower number of cycles due to the capacity and efficiency fade (Yang et al., 2018). The resulting lifespan kWhs delivered, as shown in Table 7.3, is used to calculate the functional unit.

Table 7.3: The lithium-ion battery capacity and total delivered kWh throughout the stationary storage lifespan.

Battery chemistry	*Capacity (kWh)*	*Total delivered kWh from 100 - 50% capacity*	*Total delivered kWh from 80 - 50% capacity*
NMC111 - Gr	26.6	19,644	4,624
NMC622 - Gr	27.9	20,604	4,850
NMC811 - Gr	30.7	22,672	5,337
NMC111 - Si	34.3	25,326	5,962
NMC622 - Si	37.1	27,400	6,450
NMC811 - Si	42.0	30,985	7,294

7.3.3. Recycled materials inventory and allocation

Prior to recycling, the battery is removed from the EV and manually disassembled to the module level and mechanically crushed. Hydrometallurgical recycling is then used to recover the constituent materials. The allocation of recovered materials from recycling is done assuming an open loop process with no market disequilibrium using the BPX 30-323-0 method (Allacker et al., 2014). In both pathways the recycled material is credited with the amount of material recovered based on the ability to substitute virgin materials. The 50/50 allocation approach is used to split the burdens and credits from recycling between the prior and subsequent product (Allacker et al., 2014). The end-of-life LCIs can be found in Tables S4-1 and S4-2 of Appendix C.

7.3.4. Life cycle impact assessment and interpretation of results

Life cycle impacts are calculated using ReCiPe 2016 midpoint and endpoint characterization factors. The midpoint characterization factors include: metal depletion factor (MDP), freshwater eutrophication (FEP), human toxicity (HTP), global warming potential (GWP), fresh water ecotoxicity (FETP), terrestrial ecotoxicity (TETP), freshwater consumption (WCP), human toxicity non-cancerous (HTPnc), Land use (LOP), Fossil depletion (FDP), Ionizing radiation (IRP), photochemical ozone formation (POFP), terrestrial acidification (TAP), photochemical ozone formation- ecosystem (EOFP), marine eutrophication (MEP), fine particulate matter formation (PMFP), marine ecotoxicity (METP), ozone depletion (ODP). The endpoint characterization factors include: Damage to Resource Availability, Damage to Human Health, and Damage to Ecosystems.

Both Kallitsis et al (2020) and Ellingsen et al. (2014) use the previous version, ReCiPe 2008 midpoint characterization factors. To understand the effect of updating the EcoInvent inventory data, we also calculated the results using ReCiPe 2008 midpoint characterization factors. The comparison of the ReCiPe 2008 midpoint characterization factors for the NMC111-Gr LIB from Ellingsen et al, Kallitsis et al., and this paper, can be found in Table S5-1 of Appendix C.

Due to the large number of impact categories considered in this analysis, only the endpoint characterization factors and the midpoint factor GWP are presented in the article. The LCI results and all impact category results can be found in the data repository associated with this article.

7.4. Results

In all impact categories the Repurposing Pathway results in less environmental impact than the Recycling Pathway (MDP, FEP, HTP, GWP, FETP, TETP, WCP, HTPnc, LOP, FDP, POFP, TEP, EOFP, MEP, PMFP, METP, ODP, Damage to Resource Availability, Damage to Human Health, Damage to Ecosystems). This is due to the very low impact of repurposing and the fact that generating recycled material does not show a significant benefit even when it is incorporated into a new battery with lower-cobalt chemistries. The net benefit from recycling the battery at end-of-life leads to negative impacts in the Repurposing Pathway. The recycling benefits are doubled in the Recycling Pathway due to two batteries being processed, although this is offset by the impacts of manufacturing which lead to a net positive impact. The benefits from recycling are larger in the Repurposing Pathway because this pathway is cycled less times (Table 7.3), thus the benefits are divided by a smaller number.

The choice of functional unit, 1 kWh cycled, also effects the two pathway's results; when a stationary storage battery is new, as in the Recycled Pathway, the application can be cycled more times, amortizing the burdens of battery production over more kWh cycled.

Due to the manufacturing of new batteries in the Recycling Pathway, the cathode and the other LIB components represent a large portion of impacts (Figure 7.3). Cathode production is larger in the Damage to Resource Availability and GWP characterization factors due to the use of cobalt, nickel, and aluminum. The other LIB components category is also significant and represents a larger impact in the Damage to Ecosystems characterization factor largely due to the aluminum, copper, and electricity usage in the cell container production. For the Damage to Human Health characterization factor, the cathode has higher impacts if virgin materials are used, but not if 100% recycled materials are used in the cathode.

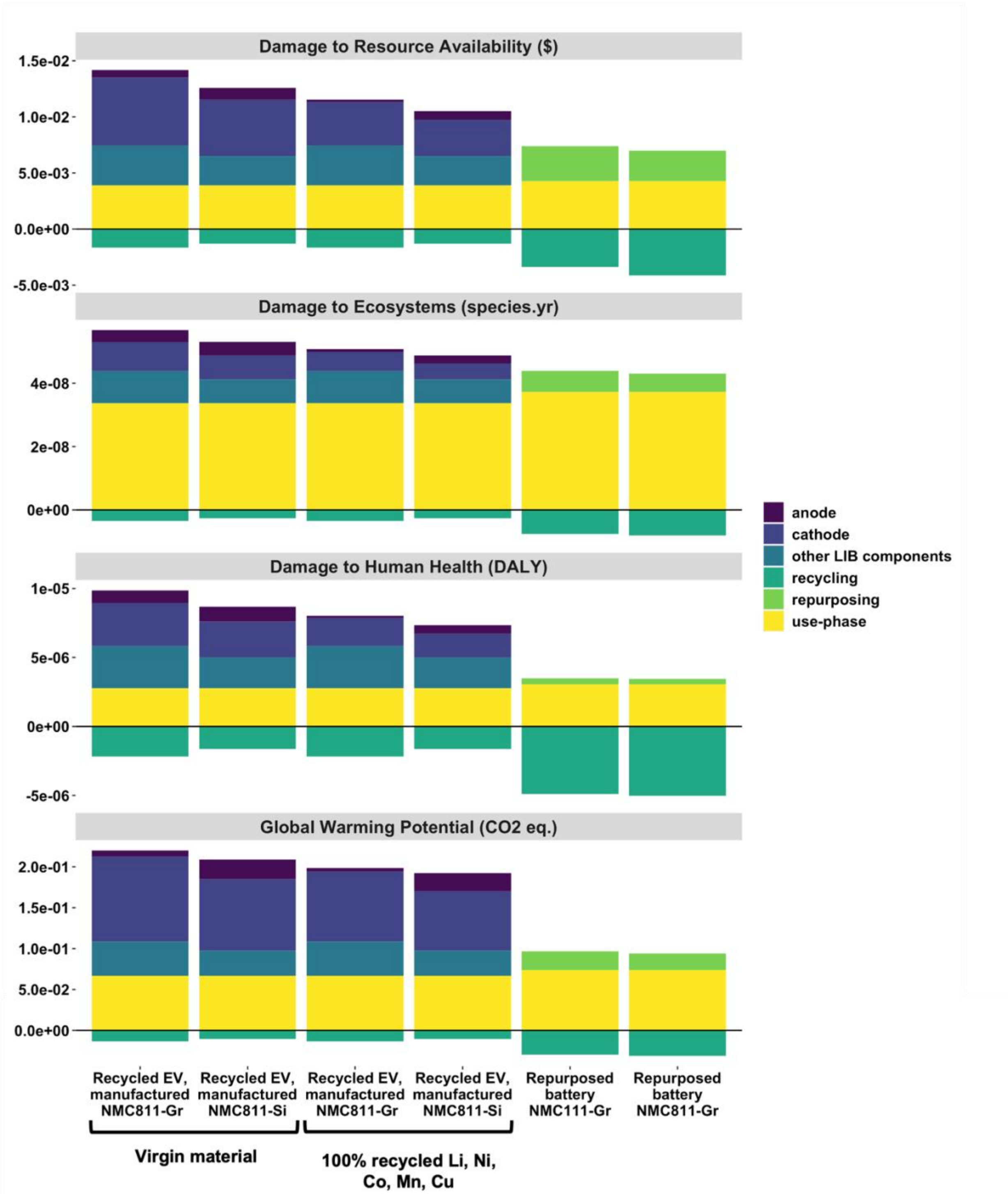

Figure 7.3: The impact of the Recycling Pathway scenarios (columns 1-4) and the Repurposing Pathway (columns 5-6).

7.4.1. Material substitution

While the analysis focused on the tradeoff between extending the lifespan of a battery through repurposing and sending the battery directly to recycling, it also explores the tradeoffs of material substitution.

Findings show that substituting the Gr anode with columnar Si thin film is beneficial in reducing environmental impacts of the battery. This is due to an increase in energy density, thus requiring less material per kWh. While this benefit is apparent, the technological improvement is not great enough to suggest the battery should skip the repurposing phase and go straight to recycling.

The manufacturing of an LIB with a NMC811 cathode, instead of NMC111, results in lower impacts to all endpoint characteristics (Figure 7.4 and Figure S6-1 in Appendix C), but the substitution of materials does not always result in the decrease of environmental impacts. Results show the NMC chemistry with decreased use of cobalt result in a net increase in some of midpoint impact indicators due to the high impact of nickel. The midpoint indicators where NMC 811 results in higher impacts than NMC111 include: MDP, PMFP, and TAP. NMC 811 has lower impacts in EOFP, FDP, GWP, HTP, MEP, ODP, TETP, FEP, LOP, POFP (Figure S6-2 in Appendix C).

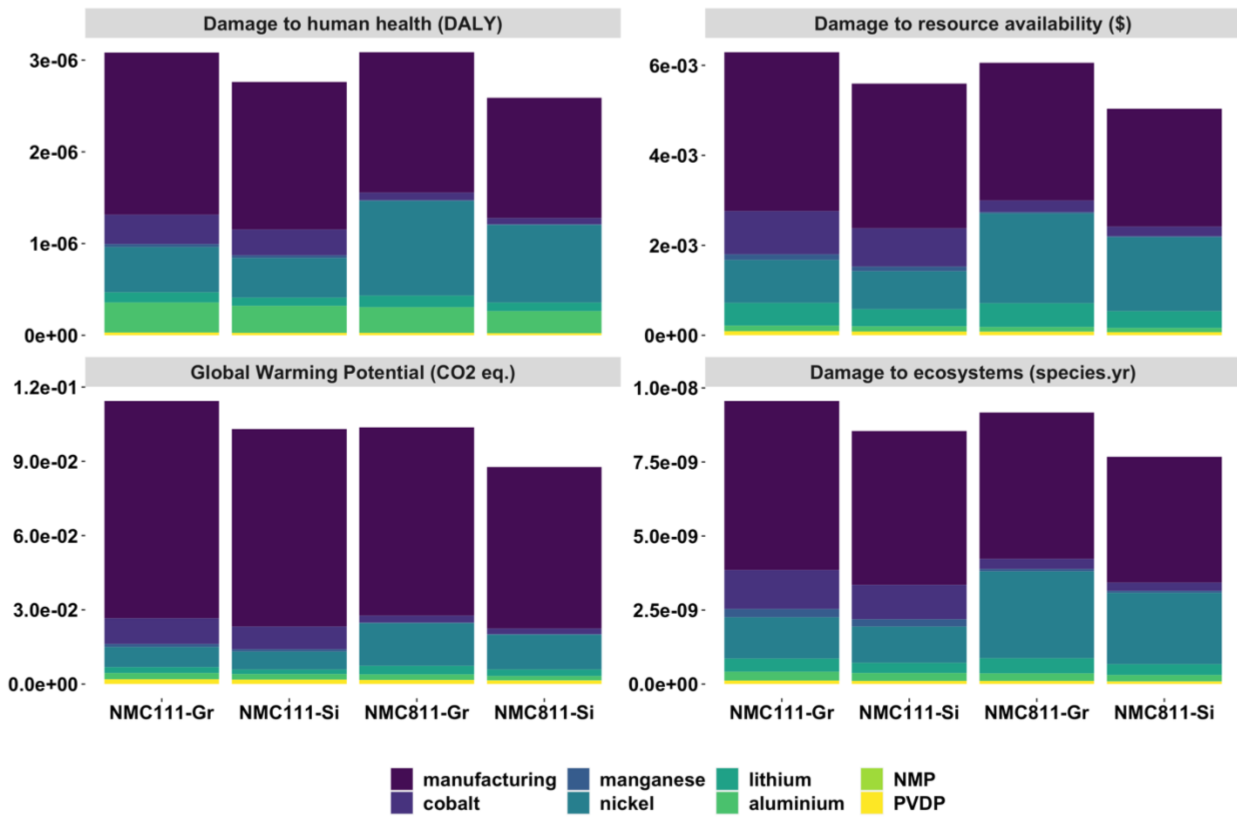

Figure 7.4: The ReCiPe endpoint impacts per kWh of the cathode in a 253 kg lithium-ion battery.

7.4.2. Recycled content

The impact of manufacturing a new battery is reduced with higher recycled content (i.e. as more recovered materials are used in manufacturing). All endpoint categories show the lowest impacts are from a battery produced with 100% recycled Li, Ni, Mn, Co, and Cu (Figure S6-3 in Appendix C). In FEP, HTPnc, and TETP, the anode represents the largest impact due to the use of copper foil, thus the decline of impacts associated with the use of recycled materials is the result of recycled copper (Figure S6-4 and S6-5 in Appendix C). Increased recycling does not considerably decrease the GWP, a category which has historically been given the most attention in LIB analysis.

7.4.3. The impact of capacity fade, efficiency fade, and the electricity source

The repurposing of LIBs is found to be better than manufacturing a new battery for the same stationary use, due to the repurposing process and ultimate recovery via recycling after stationary use. The Repurposing Pathway impacts are highly dependent on three interconnected influencers: capacity fade, efficiency fade, and electricity production source.

If only the capacity is decreased for the battery, and cycling efficiency stays at 80%, the impacts do not surpass the Recycling Pathways. If the cycling round trip efficiency is decreased, it results in a large increase to the life cycle impacts of the Repurposing Pathway.

When a repurposed battery has a low roundtrip efficiency it essentially wastes more of the electricity it is meant to store, and thus increases the environmental impacts of each kWh of electricity discharged. The point where the repurposing of a NMC111-Gr battery is less advantageous than the recycling and manufacturing of an NMC811-Si battery with 100% recycled content, was found for the endpoint indicators and GWP midpoint indicator. For Damage to Ecosystems, the Recycling Pathway is more advantageous at or below a cycling efficiency of 66%. The break-even point for Damage to Resource Availability is a lower efficiency of 43%. Due to the Repurposing Pathway having much less impacts to Damage to Human Health, even with decreased efficiency, the Repurposing Pathway is preferable.

The electricity used for battery cycling also has a large impact on the life cycle impacts of the Repurposing Pathway. If the battery charges using the US average grid, rather than electricity generated from PV, the Recycling Pathway becomes preferable at a higher cycling efficiency. For the Damage to Ecosystems indicator, the Recycling Pathway is preferable at an efficiency of 69%; Damage to Resource Availability at 74% and Damage to Human Health at 64%. Outputs from this analysis can be found in the data depository.

7.5. Discussion

7.5.1. The waste hierarchy

Findings show the currently understood waste hierarchy, of reduce, reuse, repurpose, and recycle, should continue to be used for EV LIBs retired under typically assumed conditions, when considering the material substitutions and impacts reviewed in this paper. These findings also show that there are conditions where directly to recycle batteries (i.e., those with low roundtrip efficiency) is preferable. The breakeven point for recycling versus repurposing is dependent on the generation resources for the electricity being stored during stationary use and the battery chemistry. Testing of batteries to access battery health information will be crucial in determining the best end-of-life route.

While findings show that in most cases it is better to repurpose batteries at their end-of-life, the theory that disruptive technology can change how we think about the waste hierarchy is worthy of further discussion. The focus of this analysis is on environmental impacts, although the influence of other factors such as social impacts and human rights issues, should also be considered.

Research by Rasmusssen et al. (2005) also addressed this question, stating the waste hierarchy should be considered a flexible guideline, and the socio-economic impacts should be reviewed along with the environmental. They cite the example of landfilling versus incinerating waste, claiming these processes have similar environmental impacts, although incineration is much more costly to society, thus landfilling should be prioritized. Bugge et al. (2019) addresses the waste hierarchy from a different angle, pointing out that potential abatement can go unrealized due to path dependency in the waste system when there is advancement in technology, production, infrastructure, logistics, and consumer practices. The transition from the internal combustion engine to the EV is a great example of how reorientation of the waste hierarchy for environmental gain is possible: when a high-polluting internal combustion engine vehicle reaches its end-of-life it is ideal to recycle and replace with an EV, rather than repair and continue driving the vehicle.

When a truly market disrupting and innovative technology is developed, the end-of-life for the batteries currently under production may need to be reconsidered (Figure 7.5). Thus, the life cycle impact of LIBs or other battery chemistry innovations that may be adopted in EVs should continue to be analyzed and the influence on the waste hierarchy should be examined. Continued research on the preferred waste hierarchy is particularly required for LIBs because of their long lifespan, use of critical materials, rapid growth in demand, and the rate of innovation.

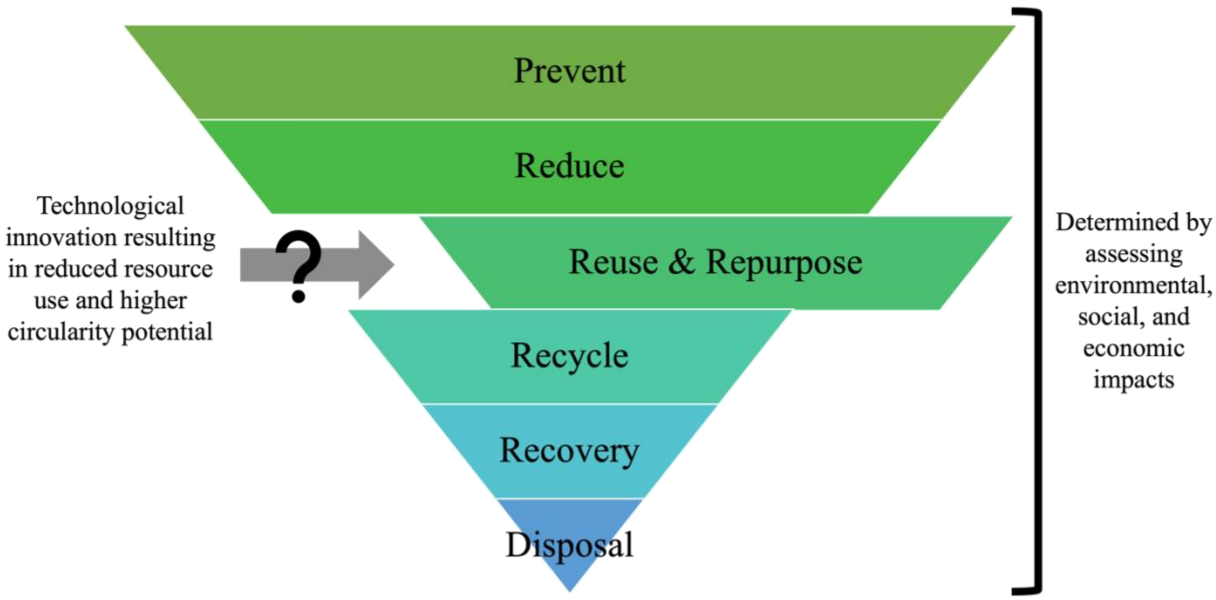

Figure 7.5: Technological development can impact the preferable waste hierarchy.

7.5.2. Social impacts

Environmental impacts represent only one element in the decision space for how to handle retired EV batteries. Literature documenting the humanitarian impacts from mining demonstrate that the review of social impacts is necessary to make definitive statements on the preferred end-of-life path for LIBs. The social impacts here are considered more broadly than the socio-economic impacts discussed by Rasmusssen et al. (2005), including the treatment and pay of laborers, the displacement of people, conflict, and corruption (Amnesty International & Afrewatch, 2016; Sovacool, 2019). While the environmental impact of cobalt is high, results demonstrate the manufacturing of low cobalt chemistries also have relatively high environmental impacts, mainly due to the energy requirements and the use of nickel, aluminum, and copper. These impacts are the reason using LIBs in a second-life application is better than recycling materials and producing a new LIB for stationary storage when examining only environmental impacts. Since this analysis does not consider the social impacts, it cannot speak to the socially preferable end-of-life route, and a social LCA or other social impact methods is needed for decision-making informed by both environmental and social impacts (Petti et al., 2018).

7.5.3. Nickel as a substitute for cobalt

The substitution of nickel for cobalt is seen to be advantageous because nickel can provide higher density at a lower weight. In addition, nickel does not have the geopolitical risk or the humanitarian concerns associated with cobalt. The NMC811 battery contains approximately four times less cobalt and nearly two times more nickel than the NMC111 battery (~.08 and ~.35 kg/kg respectively of cobalt and ~.60 and ~.35 kg/kg of nickel) (Argonne National Laboratory, 2020). The substitution has several corresponding environmental and economic impacts, pointing to the conclusion that materials used as substitutes must also be interrogated.

All material extraction comes with resource destruction and pollution, and nickel's impact is not considered low. The refining process of nickel production results in high SO_X emissions (244.18 g/kg) (Dai et al., 2019). The majority of nickel comes from Indonesia and the Philippines, where strip mining leaves little ability for rehabilitation and results in widespread biodiversity loss. This is especially harmful in these areas because the mines are located in tropical rainforests, a biome that is a large carbon sink and essential to slowing climate change, as well as the home to many endangered and endemic species (Barlow et al., 2016; Supriatna et al., 2020). Despite the need to preserve the habitat, little effort has gone into land rehabilitation strategies (van der Ent et al., 2013).

In addition, nickel is becoming a designated critical material due to increasing demand, which is projected to be higher than the global refining capacity (The White House, 2021). From the US perspective, due to its lack of nickel refining capacity, nickel supply relies on other nations and poses a risk to energy security. The increased demand for nickel and strain on resources is now leading to rapidly increasing costs.

7.5.4. Availability of recycled materials

Using recycled materials results in a large reduction of impacts associated. Prioritization of ensuring recycling of batteries at their end-of-life to create a circular LIB economy will help materialize these reduced impacts. The increased demand for batteries as EVs begins to replace internal combustion engines makes it so that all LIBs cannot be manufactured with 100% recycled materials. Under current technologies and chemistry, only approximately 25% - 65% of LIB materials can come from recycled content in 2040 (J. Dunn et al., 2021; C. Xu et al., 2020).

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8. Conclusion

This research used industrial ecology methods to review and recommend policies for increasing the sustainability of the LIB supply chain. Sustainability, in this instance, is used to describe the abatement of negative environmental and social impacts including reducing virgin material demand through enhanced circularity. Section 4 discussed LIB policy; Section 5 calculated global circularity potential of materials at a regional scale; Section 6 zoomed in on the US and forecasted RCS that could be met if the US were to implement the policy, as well as assessed environmental and social impacts of recycling versus virgin extraction; Section 7 assessed the impact of technological innovation and its potential to alter the standard waste hierarchy. These research questions were spurred by the broader policy and industry environment which is motivated to reduce the environmental and social impacts from EV production, and the need to ensure batteries are recycled at their end-of-life.

Appendices

A: Supporting Information for Section 5

Table A: Historical EV sales until 2018 as a percentage of total car sales.

Year	*Europe*	*China*	*US*	*RoW*
2010	0.0%	0.0%	0.0%	0.0%
2011	0.1%	0.0%	0.1%	0.1%
2012	0.2%	0.1%	0.4%	0.1%
2013	0.4%	0.1%	0.6%	0.1%
2014	0.5%	0.3%	0.7%	0.1%
2015	1.0%	0.8%	0.7%	0.2%
2016	1.1%	1.3%	0.9%	0.2%
2017	1.5%	2.2%	1.2%	0.4%
2018	2.0%	4.1%	2.0%	0.5%

Work Cited

Table B: The global circularity potential in each scenario in 2040.

	Al	*Co*	*Cu*	*Gr*	*Li*	*Mn*	*Ni*
C1							
S1	52%	52%	53%	52%	52%	52%	52%
S2	54%	54%	54%	54%	54%	54%	54%
S3	36%	37%	37%	36%	37%	40%	35%
S4	31%	32%	32%	31%	32%	33%	30%
C2							
S1	52%	60%	52%	52%	53%	57%	53%
S2	54%	64%	55%	54%	56%	63%	53%
S3	36%	41%	36%	36%	37%	39%	36%
S4	31%	35%	31%	31%	32%	35%	31%
C3							
S1	53%	63%	54%	52%	56%	0%	47%
S2	55%	67%	57%	54%	58%	0%	48%
S3	36%	43%	37%	36%	38%	0%	33%
S4	31%	37%	32%	31%	33%	0%	28%
C4							
S1	53%	85%	55%	52%	56%	80%	50%
S2	55%	91%	58%	54%	59%	93%	50%
S3	36%	56%	38%	36%	38%	54%	34%
S4	31%	49%	33%	31%	33%	48%	29%
C5							

S1	52%	53%	53%	52%	52%	51%	52%
S2	54%	56%	55%	54%	54%	57%	52%
S3	36%	37%	36%	36%	36%	36%	36%
S4	31%	32%	31%	31%	31%	31%	31%
C6							
S1	49%	0%	44%	50%	58%	0%	0%
S2	51%	0%	46%	52%	60%	0%	0%
S3	34%	0%	31%	35%	39%	0%	0%
S4	29%	0%	27%	30%	34%	0%	0%

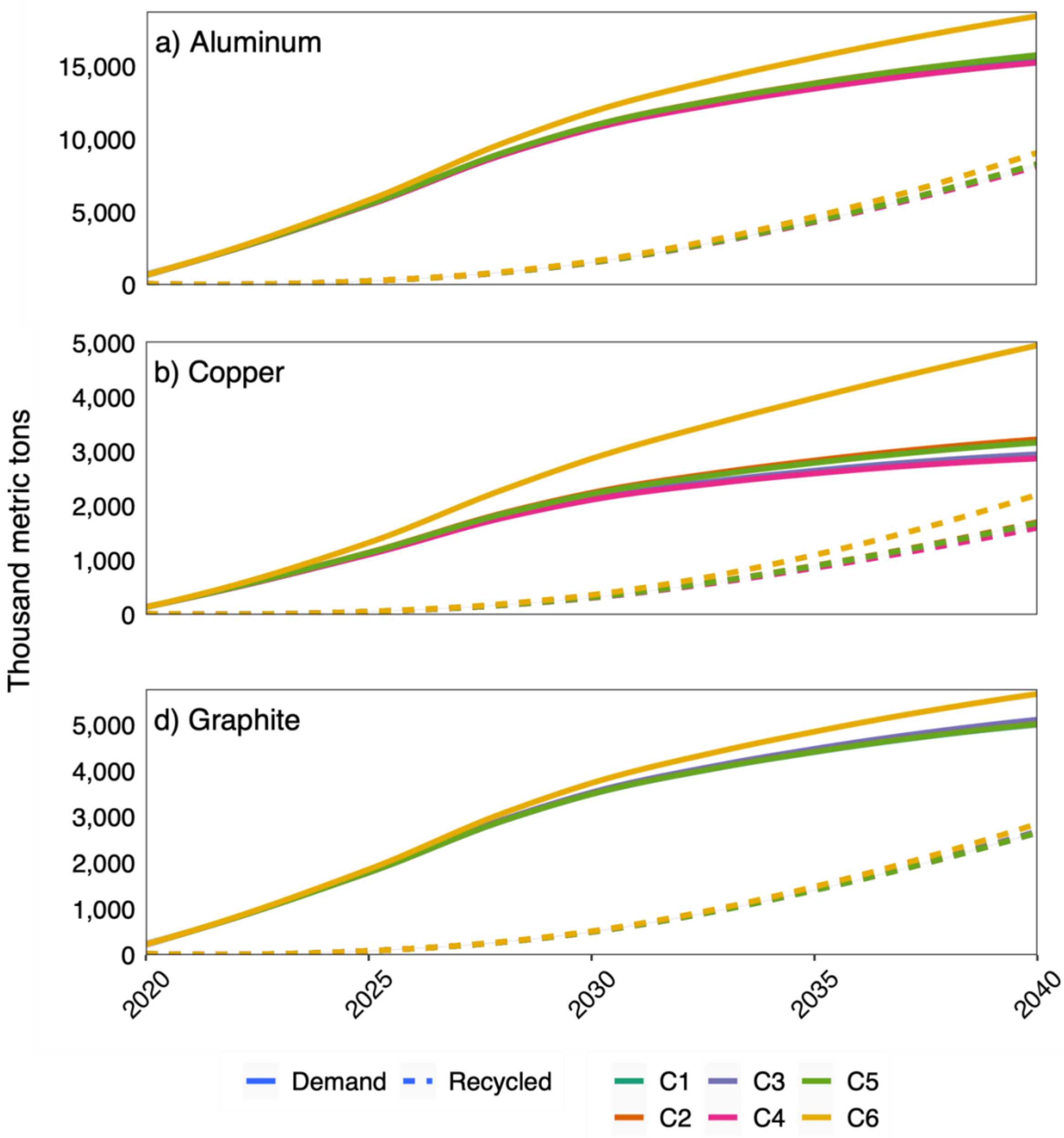


Figure A: Global EV battery material demand and retired supply of aluminum, copper, and graphite for light-duty EV battery production. This graphic represents the policy-based sales scenario (S1) from 2020 to 2040. The dashed lines represent recoverable material and the solid lines represent material demand.

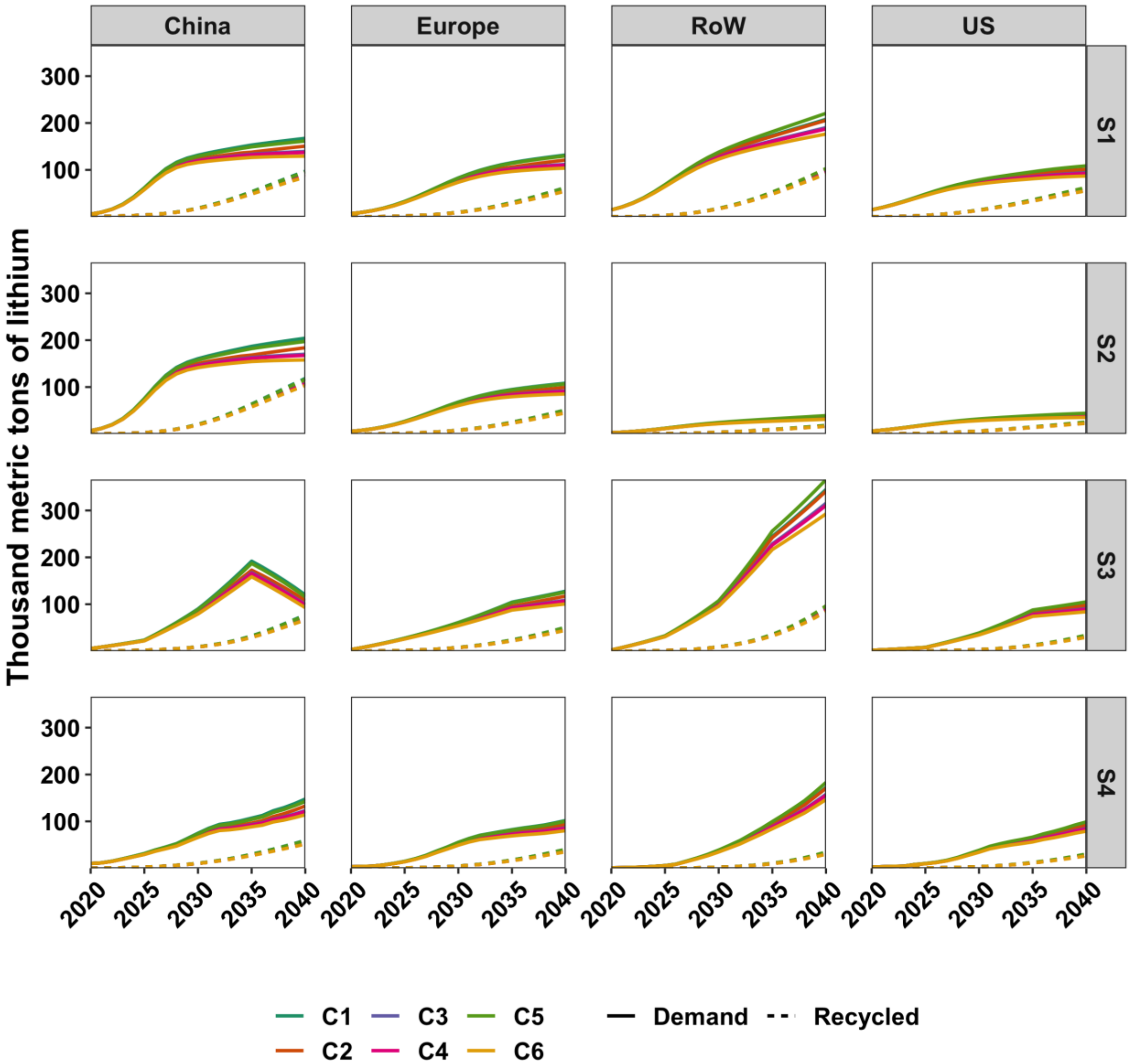


Figure B: The potential circularity of lithium from 2010 to 2040 in each region and under each cathode and sales scenario.

B: Supporting Information for Section 6

Table A: The cathode chemistry stoichiometry (kg/kWh) taken from BatPac (Argonne National Laboratory, 2020)

Cathode	*Li*	*Ni*	*Co*	*Mn*	*Al*	*Cu*	*Gr*	*Pack*
NMC111	0.141	0.351	0.352	0.328	3.110	0.677	0.978	9.432
NMC523	0.136	0.508	0.204	0.285	3.070	0.661	0.981	9.300
NMC622	0.118	0.531	0.178	0.166	3.017	0.605	0.960	8.977
NMC811	0.100	0.600	0.075	0.070	2.921	0.549	0.961	8.606
NCA	0.102	0.672	0.127	0.000	2.920	0.564	0.978	8.730
LMO	0.106	0.000	0.000	1.396	3.369	0.863	0.911	10.262
LFP	0.095	0.000	0.000	0.000	3.528	0.946	1.085	8.977

Work Cited

Table B: The value of materials from 2000 to 2020. These were adjusted to 2021 US dollars using the CPI inflation calculator (CPI Inflation Calculator, n.d.). The values are from the USGS material commodity spot prices (National Minerals Information Center & USGS, 2021).

	Cobalt		***Lithium carbonate***		***Nickel***		***Manganese***	
	$ / kg	*$ / kg (2021 value)*	*$ / kg*	*$ / kg (2021 value)*	*$ / kg*	*$ / kg (2021 value)*	*$ / kg*	*$ / kg (2021 value)*
2000	33.42	54.76	4.47	7.32	8.64	14.16	0.11	0.18
2001	23.26	36.74	1.90	3.00	5.95	9.40	0.12	0.19
2002	15.23	23.79	2.00	3.12	6.77	10.57	0.11	0.17
2003	23.37	35.57	2.00	3.04	9.63	14.66	0.12	0.18
2004	54.01	80.66	2.00	2.99	13.84	20.67	0.14	0.21
2005	35.19	51.04	3.00	4.35	14.74	21.38	0.23	0.34
2006	37.96	52.95	4.00	5.58	42.44	59.20	0.15	0.21
2007	67.35	92.03	6.10	8.34	37.22	50.86	0.15	0.21
2008	86.00	112.69	5.90	7.73	21.10	27.65	0.58	0.76
2009	39.68	51.98	5.00	6.55	14.94	19.57	0.32	0.42
2010	45.97	58.68	5.18	6.61	21.80	27.83	0.38	0.49
2011	39.66	49.81	5.18	6.51	22.89	28.75	0.32	0.40
2012	31.02	37.85	6.06	7.39	17.53	21.39	0.24	0.29
2013	28.42	34.14	6.80	8.17	15.02	18.04	0.22	0.27

2014	31.75	37.54	6.60	7.80	16.86	19.94	0.22	0.26
2015	29.63	35.07	6.50	7.69	11.83	14.00	0.18	0.21
2016	26.48	30.91	8.65	10.10	9.59	11.20	0.21	0.25
2017	59.46	67.72	15.00	17.08	10.40	11.85	0.29	0.33
2018	82.52	92.08	17.00	18.97	13.11	14.63	0.34	0.38
2019	37.37	41.06	12.70	13.96	13.90	15.27	0.27	0.30
2020	35.27	37.82	8.00	8.58	14.00	15.01	0.23	0.25
2021	60.00	60.00	31.00	31.00	19.00	19.00	0.16	0.16

Table C: The cathode chemistry value. This was derived using EverBatt (Argonne National Lab, 2021a) and the high and low values in Table x.

	Co high	*Co low*	*Li high*	*Li low*	*Mn high*	*Mn low*	*Ni high*	*Ni low*
LCO	$76.27	$22.75	$49.78	$39.20	$44.39	$44.39	$44.39	$44.39
LFP	$14.00	$14.00	$14.00	$14.00	$14.00	$14.00	$14.00	$14.00
NCA	$27.53	$19.18	$24.22	$17.40	$22.56	$22.56	$44.16	$19.74
NMC 111	$35.46	$17.35	$30.13	$19.40	$25.06	$24.64	$33.61	$23.50
NMC 532	$27.78	$16.93	$26.78	$16.06	$21.67	$21.29	$34.71	$19.57
NMC 622	$28.59	$17.78	$27.59	$16.91	$22.38	$22.13	$38.16	$20.06
NMC 811	$23.31	$17.92	$21.74	$15.02	$20.22	$20.09	$41.37	$17.32

Table D: The recycled content standards representing the 95% confidence interval. This calculation does not include the use of pyrometallurgical recycling and only includes the increasing collection rate from 65% in 2025 increasing to 90% in 2050.

	lower bound	***upper bound***
Cobalt		
2025	10%	11%
2030	11%	12%
2035	15%	18%
2040	24%	27%
2045	37%	41%
2050	45%	52%
Lithium		
2025	6%	7%
2030	7%	8%
2035	9%	11%
2040	14%	17%
2045	19%	23%
2050	22%	27%
Nickel		
2025	10%	11%
2030	10%	12%
2035	15%	17%

2040	23%	25%
2045	34%	38%
2050	40%	46%

Table E: The recycled content standards representing the 95% confidence interval for cobalt, nickel, and lithium parsed by cathode chemistry scenario. The "LFP forecast scenario" calculates the confidence interval for all scenarios excluding the NCX forecast, and the "NCX forecast scenario" calculates the confidence interval for all scenarios excluding the LFP forecast.

	LFP forecast scenario		***NCX forecast scenario***	
	lower bound	*upper bound*	*lower bound*	*upper bound*
Cobalt				
2025	11%	11%	11%	11%
2030	12%	13%	11%	12%
2035	17%	18%	15%	17%
2040	27%	28%	23%	24%
2045	40%	42%	31%	33%
2050	49%	53%	35%	37%
Lithium				
2025	4%	5%	4%	5%
2030	5%	6%	5%	6%
2035	7%	8%	7%	8%
2040	10%	11%	10%	12%
2045	13%	15%	13%	15%
2050	14%	17%	14%	17%
Nickel				

2025	11%	11%	11%	11%
2030	12%	12%	11%	12%
2035	16%	17%	15%	16%
2040	26%	27%	22%	23%
2045	37%	38%	29%	31%
2050	43%	45%	32%	34%

Table F: The recycled content standards representing the 95% confidence interval for cobalt, nickel, and lithium parsed by the sales scenarios: SDS and STEPS.

	SDS forecast scenario		***STEPS forecast scenario***	
	lower bound	*upper bound*	*lower bound*	*upper bound*
Cobalt				
2025	8%	8%	13%	14%
2030	8%	8%	16%	16%
2035	12%	12%	21%	22%
2040	24%	26%	26%	27%
2045	40%	42%	32%	34%
2050	55%	58%	32%	33%
Lithium				
2025	3%	4%	5%	6%
2030	3%	4%	6%	8%
2035	5%	6%	9%	10%
2040	9%	11%	10%	12%
2045	14%	17%	12%	14%
2050	18%	21%	11%	13%
Nickel				
2025	8%	8%	14%	14%
2030	7%	8%	16%	16%

2035	11%	12%	20%	21%
2040	23%	24%	25%	26%
2045	37%	39%	30%	31%
2050	48%	51%	29%	30%

Table G: The recycled content standards representing the 95% confidence interval for cobalt, nickel, and lithium parsed by recycling process. "All recycling processes" calculates the confidence interval for all scenarios, "direct recycling" calculates the confidence interval for all scenarios using direct recycling and excluding the other recycling processes, "hydrometallurgical recycling" calculates the confidence interval for scenarios including hydrometallurgical recycling and excluding the other recycling processes, and the same pattern continues for "pyrometallurgical".

	Direct recycling		***Hydrometallurgical***		***Pyrometallurgical***		***All recycling processes***	
	lower bound	*upper bound*	*lower bound*	*upper bound*	*lower bound*	*upper bound*	*lower bound*	*upper bound*
Cobalt								
2025	10%	11%	11%	12%	10%	11%	11%	11%
2030	11%	12%	12%	13%	11%	12%	12%	12%
2035	16%	17%	17%	18%	16%	17%	16%	17%
2040	25%	26%	27%	28%	25%	26%	26%	26%
2045	35%	37%	38%	41%	35%	37%	37%	38%
2050	41%	45%	45%	49%	41%	45%	43%	46%
Lithium								
2025	4%	4%	10%	10%	0%	0%	4%	5%
2030	5%	5%	10%	11%	0%	0%	5%	6%
2035	6%	7%	14%	16%	0%	0%	7%	8%
2040	10%	10%	21%	22%	0%	0%	10%	11%

2045	13%	13%	28%	30%	0%	0%	13%	15%
2050	14%	15%	31%	34%	0%	0%	15%	17%
Nickel								
2025	10%	11%	11%	12%	10%	11%	11%	11%
2030	11%	12%	12%	13%	11%	12%	11%	12%
2035	15%	16%	16%	18%	15%	16%	16%	16%
2040	23%	24%	25%	27%	23%	24%	24%	25%
2045	32%	34%	35%	37%	32%	34%	34%	35%
2050	37%	40%	40%	43%	37%	40%	39%	40%

Table H: The cost of recycling large-format lithium-ion batteries ($/kg)

	NCX cathode scenario			***LFP cathode Scenario***		
	Direct ($/kg)	*Hydro ($/kg)*	*Pyro ($/kg)*	*Direct ($/kg)*	*Hydro ($/kg)*	*Pyro ($/kg)*
China-truck & tanker						
2020	5.50	3.93	4.02	5.50	3.93	4.02
2030	5.50	3.93	4.03	5.49	3.92	4.02
2040	5.52	3.94	4.04	5.48	3.92	4.01
2050	5.52	3.95	4.04	5.46	3.91	4.01
US-train						
2020	7.21	4.88	4.88	7.21	4.88	4.88
2030	7.23	4.89	4.89	7.16	4.85	4.86
2040	7.29	4.92	4.91	7.12	4.82	4.84
2050	7.31	4.94	4.92	7.05	4.78	4.82
US-truck						
2020	7.45	5.12	5.12	7.45	5.12	5.12
2030	7.48	5.13	5.13	7.41	5.09	5.10
2040	7.53	5.17	5.15	7.36	5.06	5.08
2050	7.56	5.19	5.16	7.29	5.02	5.06

Table I: A sensitivity analysis of the impact to the profit (or loss) of recycling was produced using the commodity prices in table S6 and S7. The following tables demonstrate the deviation from the baseline scenario. Green indicates the largest impact, orange the second largest impact, and yellow the third largest impact.

	Direct		***Hydro***		***Pyro***	
	LFP	*NCX*	*LFP*	*NCX*	*LFP*	*NCX*
2020						
cobalt high	305%	305%	493%	493%	1407%	1407%
cobalt low	-207%	-207%	-333%	-335%	-950%	-957%
lithium high	135%	137%	80%	80%	0%	0%
lithium low	-242%	-243%	-78%	-78%	0%	0%
manganese high	7%	5%	2%	2%	0%	0%
manganese low	0%	0%	0%	0%	0%	0%
nickel high	838%	837%	1365%	1368%	3900%	3907%
nickel low	-110%	-112%	-178%	-178%	-507%	-507%
2050						
cobalt high	279%	4867%	81%	872%	56%	491%
cobalt low	-194%	-3267%	-56%	-594%	-39%	-334%
lithium high	142%	2500%	23%	178%	0%	0%
lithium low	-282%	-5033%	-22%	-167%	0%	0%

manganese high	0%	100%	1%	6%	0%	0%
manganese low	-6%	0%	0%	0%	0%	0%
nickel high	1055%	19300%	305%	3506%	214%	1972%
nickel low	-139%	-2533%	-40%	-456%	-28%	-256%

Table J: The pack-level retired material per year in metric tons (t).

Year	*Min (t/year)*	*Max (t/year)*
2020	3,172	9,537
2021	4,782	14,468
2022	6,973	21,838
2023	9,949	33,067
2024	13,924	49,668
2025	19,107	73,148
2026	25,688	104,946
2027	33,989	149,160
2028	44,175	210,526
2029	56,352	293,885
2030	70,575	403,945
2031	87,556	545,092
2032	107,589	721,281
2033	130,885	936,008
2034	157,476	1,191,811
2035	187,572	1,491,295
2036	221,184	1,836,001
2037	258,419	2,223,234
2038	299,599	2,649,346

2039	344,151	3,107,181
2040	393,365	3,594,527
2041	447,151	4,104,903
2042	505,208	4,628,435
2043	567,708	5,159,061
2044	634,769	5,690,856
2045	706,456	6,218,250
2046	782,778	6,736,213
2047	863,892	7,228,929
2048	951,819	7,694,994
2049	1,047,783	8,133,811
2050	1,153,014	8,545,553

Table K: The commercial lithium-ion battery recycling facilities in the US and Canada that are operational or under development. This Table was adapted from the California AB 2832 report (Kendall et al., 2022).

Company	***Location(s)***	***Current capacity (t/year)***	***Planned total capacity (t/year)***
American Battery Technologies (Graham, 2020; Recycling Coordinators, n.d.)	Fernley, Nevada	-	20,000
American Manganese (American Manganese, 2021)	Vancouver, British Columbia	-	182.5
Ascend Elements (PR Newswire, 2022) *(formerly Battery Resourcers)*	Worcester, Massachusetts Novi, Michigan; Covington, Georgia	Unknown	30,000
Interco (Interco, 2022)	Madison, Illinois	Unknown	Unknown
Li-cycle Corporation (Li-Cycle, 2022; Roberts, 2021)	Rochester, N.Y. (spoke) Kingston, Ontario (spoke) Phoenix, Arizona (spoke)	5,000 5,000 -	5,000 5,000 10,000

	Tuscaloosa, Alabama (spoke)	-	5,000
	Rochester, N.Y. (hub)	-	60,000
Lithion (Lithion, 2021)	Ajou, Quebec; Planned locations unknown	200	7,500
Princeton NuEnergy (PR Newswire, 2021)	Dallas, Texas	-	Unknown
Recycling Coordinators (Recycling Coordinators, n.d.)	Akron, Ohio	Unknown	Unknown
Redwood Materials (Carney, 2021)	Carson City, Nevada; Reno, Nevada	18,100	Unknown
Retriev Technologies (Pinegar & Smith, 2019)	Lancaster, Ohio and Trail, British Columbia	4,500	4500
Umicore Canada Inc. (Umicore, n.d.)	Fort Saskatchewan, Alberta	Unknown	Unknown

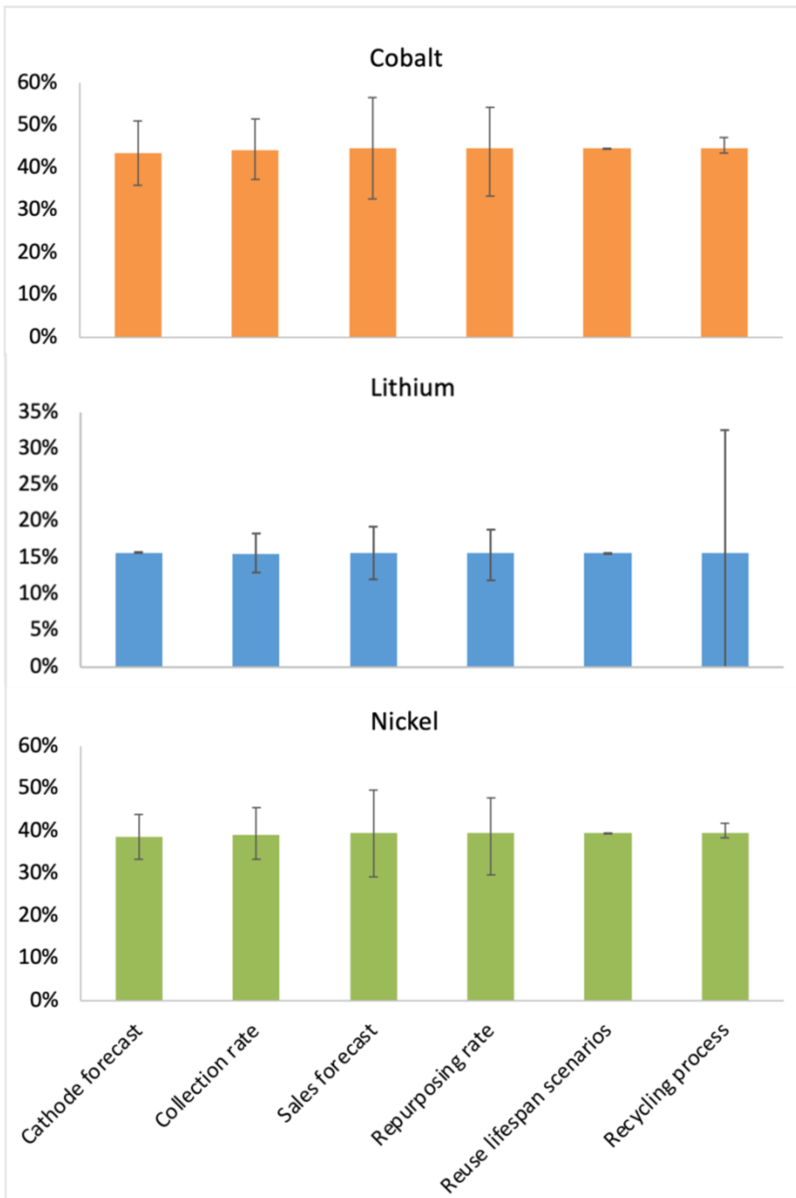
Table L: The data behind Figure 6.6. The environmental impact of recycling lithium-ion batteries in 2020. The avoided emissions represent the environmental impacts if the materials recovered were from virgin ore, the recycling emissions are the emissions resulting from the recycling process, and the net emissions represent the emissions saved because materials were recycled instead of mined. Recycling in China and transporting via truck and ocean tanker is abbreviated to "China- T&T".

	Direct	***Hydro***	***Pyro***
CO2e			
China-T&T			
avoided emissions	-5698.08	-4954.01	-3434.57
net emissions	-3432.62	-3048.70	-968.95
recycling emissions	2265.46	1905.31	2465.61
US-Train			
avoided emissions	-5698.08	-4954.01	-3434.57
net emissions	-4281.26	-3390.22	-1401.26
recycling emissions	1416.83	1563.79	2033.30
US-Truck			
avoided emissions	-5698.08	-4954.01	-3434.57
net emissions	-4075.33	-3184.28	-1195.33
recycling emissions	1622.76	1769.73	2239.24
NOx			
China-T&T			

avoided emissions	-9.86	-10.67	-7.60
net emissions	-4.47	-5.69	-3.43
recycling emissions	5.39	4.97	4.17
US-Train			
avoided emissions	-9.86	-10.67	-7.60
net emissions	-7.42	-8.51	-6.27
recycling emissions	2.44	2.15	1.33
US-Truck			
avoided emissions	-9.86	-10.67	-7.60
net emissions	-7.27	-8.37	-6.12
recycling emissions	2.59	2.30	1.48
SOx			
China-T&T			
avoided emissions	-161.23	-208.33	-191.89
net emissions	-155.84	-183.61	-188.93
recycling emissions	5.38	24.72	2.96
US-Train			
avoided emissions	-161.23	-208.33	-191.89
net emissions	-159.55	-185.51	-191.15
recycling emissions	1.67	22.83	0.74
US-Truck			
avoided emissions	-161.23	-208.33	-191.89

net emissions	-159.54	-185.49	-191.13
recycling emissions	1.69	22.84	0.76

Figure A: The mean RCS for each scenario in 2050. The error bars represent the maximum and minimum RCS in 2050 for each scenario grouping.



C: Supporting Information for Section 7

This section is organized in the following way:

- S1: Capacity calculations
- S2: Updated life cycle inventory from Kallitsis et al. (2020)
- S3: Stoichiometry calculations
- S4: Battery end-of-life life cycle inventory
- S5: Comparing results of NMC111-Gr with Kallitsis et al. (2020) and Ellingsen et al. (2014)
- S6: Supplementary results

S1. Capacity calculations

The full model for calculating the battery capacity was taken from Wentker et al. (2019). To calculate the capacity per pack (Equation S1-1), the volumetric energy [Wh/L] is multiplied by the active material [L].

$$kWh\ [per\ pack] = Volumetric\ energy\ \left[\frac{Wh}{L}\right] * \frac{Active\ material\ [L]}{1000} \qquad \text{Eq. S1-1}$$

The volumetric energy is calculated by the following equation:

$$Volumetric\ energy\ \left[\frac{Wh}{L}\right] = Cell\ voltage * \frac{Useable\ capacity\ [\frac{mAH}{cm^3}]}{Single\ stack\ thickness\ [cm]} \qquad \text{Eq. S1-2}$$

To calculate the volumetric energy, first the cell voltage, useable capacity, and single stack thickness must be found. The following three equations use inputs from Table S1-1 and S1-2.

$$Volumetric\ Capacity\ \left[\frac{mAh}{cm3}\right] = Active\ Material\ Density\ incl.Pores\ \left[\frac{g}{cm^3}\right] * Practical\ Discharge\ Capacity\ [\frac{mAh}{g}]$$

Eq. S1-3

$$Useable\ capacity\ \left[\frac{mAH}{cm^3}\right] = Pos.Elec.[\mu m] * .0001 * Volumetric\ Capacity\ [mAh/cm3]$$

Eq. S1-4

$$Single\ stack\ thickness\ [cm] = \frac{Pos.Elec.[\mu m]}{10000} + \frac{Neg.Elec.\ [\mu m]}{10000} + Al\ [cm] + Cu\ [cm] \qquad \text{Eq. S1-5}$$

$$Cell\ voltage = Voltage_{cathode} - Voltage_{anode} \qquad \text{Eq. S1-6}$$

To find the second part of Equation S1-1, the active material, Equation S1-7 is used with inputs from Table S1-1.

$$Active\ material\ [L] = \frac{Active\ material\ per\ pack\ [g]}{Active\ Material\ Density\ incl.Pores\ [g/cm^3]} * .001 \qquad \text{Eq. S1-7}$$

Table S1-1: Variables used in Equations S1-3 and S1-6.

Variable	***NMC 111***	***NMC 622***	***NMC 811***	***Gr***	***Si***
Active material density including pores [g/cm^3]	2.61	2.61	2.61	1.33	1.36
Practical discharge capacity [mAh/g]	160	170	200	365	1000
Voltage [V]	3.7	3.8	3.8	.1	.4

Table S1-2: Variables used in Equations S1-4, S1-5, and S1-7.

Variable	***NMC111-Gr***	***NMC622-Gr***	***NMC811-Gr***	***NMC111-Si***	***NMC622-Si***	***NMC811-Si***
Positive electrode thickness [µm]	65	65	65	80	80	80
Negative electrode thickness [µm]	61.54	65.38	76.92	29.45	25.42	29.91
Al thickness [cm]	.001	.001	.001	.001	.001	.001
Cu thickness [cm]	.0005	.0005	.0005	.0005	.0005	.0005
Active material per pack [g]	42.55	39.98	40.05	43.37	43.43	43.38

S2. Updated life cycle inventory (LCI) from Kallitsis et al.

The LCI from Kallitsis et al. (2020) is updated to the 3.6 process for NMC111-Gr. The input is then adjusted to match the desired chemistry. The tables below are taken from Kallitsis et al. (2020) and modified for the chemistries NMC622-Gr, NMC811-Gr, NMC111-Si, NMC622-Si, and NMC811-Si. Not all tables are listed, only those from Kallitsis et al. (2020) which required modification for this study.

NMC622-Gr

Table S2-1: Positive active material production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Li[N3/5C1/5M1/5]O2		1	kg	
Lithium carbonate	0.38		kg	GLO: market for lithium carbonate
N3/5C1/5M1/5(OH)2	0.95		kg	
Chemical plant	4.6×10-10		pcs.	RER: chemical factory construction, organics
Electricity	25.2		MJ	RAS: market group for electricity, low voltage

Table S2-2: NCM hydroxide production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
$N_{3/5}C_{1/5}M_{1/5}(OH)_2$		1	kg	
Cobalt sulphate	0.32		kg	
Manganese sulphate	0.31		kg	GLO: market for manganese sulfate
Nickel sulphate	0.96		kg	GLO: market for nickel sulfate
Sodium hydroxide	0.84		kg	RER: soda production, solvay process
Chemical plant	4×10^{-10}		pcs.	RER: chemical factory construction, organics
Heat	42.6		MJ	RoW: market for heat, from steam, in chemical industry
Sodium sulphate		1.6	kg	Sodium sulphate [Inorganic emissions to fresh water]

NMC811-Gr

Table S2-3: Positive active material production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Li[N4/5C1/10M1/10]O2		1	kg	
Lithium hydroxide	0.25		kg	GLO: market for lithium hydroxide
N4/5C1/10M1/10(OH)2	0.95		kg	
Chemical plant	4.6×10-10		pcs.	RER: chemical factory construction, organics
Electricity	25.2		MJ	RAS: market group for electricity, low voltage

Table S2-4: NCM hydroxide production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
$N_{1/3}C_{1/3}M_{1/3}(OH)_2$		1	kg	
Cobalt sulphate	0.16		kg	
Manganese sulphate	0.16		kg	GLO: market for manganese sulfate
Nickel sulphate	1.27		kg	GLO: market for nickel sulfate
Sodium hydroxide	0.85		kg	RER: soda production, solvay process
Chemical plant	4×10^{-10}		pcs.	RER: chemical factory construction, organics
Heat	42.6		MJ	RoW: market for heat, from steam, in chemical industry
Sodium sulphate		1.6	kg	Sodium sulphate [Inorganic emissions to fresh water]

NMC111-Si

Table S2-5: Battery cell production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Battery cell		1	kg	
Anode	0.31		kg	
Cathode	0.50		kg	
Electrolyte	0.16		kg	
Separator	0.022		kg	
Cell container	6.70×10^{-3}		kg	
Decarbonised water	3.80×10^{2}		kg	GLO: market for water, decarbonised, at user
Electricity	1.01×10^{2}		MJ	46%: KR: electricity production, hard coal 33%: KR: electricity production, nuclear, pressure water reactor 15%: KR: electricity production, natural gas, conventional power plant 4.4%: KR: electricity production, oil 1.4%: KR: electricity production, hydro, run-of-river 0.15%: KR: electricity production, wind, 1-3MW turbine, onshore 0.05 % KR: electricity production, photovoltaic, 570kWp open ground installation, multi-Si

Rail transport	0.26		tkm	RER: market group for transport, freight train
Lorry transport	0.1		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3
Waste heat		100	MJ	

Table S2-6: Anode Production

Description	Input	Output	Unit	Ecoinvent 3.5 Process
Anode		1	kg	
Negative electrode paste	0.43		kg	
Negative current collector Cu	0.57		kg	
Laser etching	0.1		h	GLO: market for laser machining, metal, with YAG-laser, 30W power
Rail transport	0.37		tkm	RER: market group for transport, freight train
Lorry transport	0.1		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3

Table S2-7: Mixing of anode active material

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Negative electrode paste		1	kg	
Silicon, electronics grade	0.96		kg	DE: silicon production, electronics grade
Acrylic acid	0.02		kg	RER: acrylic acid production
Carboxymethyl cellulose	0.02		kg	RoW: carboxymethyl cellulose production, powder
Rail transport	1.2		tkm	RER: market group for transport, freight train
Lorry transport	0.19		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3
Chemical plant	4×10^{-10}		pcs.	RER: chemical factory construction, organics
1-Methyl-2-pyrrolidinone		0.94	kg	1-Methyl-2-pyrrolidone [Group NMVOC to air]

Table S2-8: Positive active material production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
$Li[N_{1/3}C_{1/3}M_{1/3}]O_2$		1	kg	
Lithium carbonate	0.32		kg	GLO: market for lithium carbonate
$N_{1/3}C_{1/3}M_{1/3}(OH)_2$	0.96		kg	
Chemical plant	4.6×10^{-10}		pcs.	RER: chemical factory construction, organics
Electricity	25.2		MJ	RAS: market group for electricity, low voltage

Table S2-9: NCM hydroxide production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
$N_{1/3}C_{1/3}M_{1/3}(OH)_2$		1	kg	
Cobalt sulphate	0.50		kg	
Manganese sulphate	0.48		kg	GLO: market for manganese sulfate
Nickel sulphate	0.50		kg	GLO: market for nickel sulfate
Sodium hydroxide	0.89		kg	RER: soda production, solvay process
Chemical plant	4×10^{-10}		pcs.	RER: chemical factory construction, organics
Heat	42.6		MJ	RoW: market for heat, from steam, in chemical industry
Sodium sulphate		1.6	kg	Sodium sulphate [Inorganic emissions to fresh water]

NMC622-Si

Table S2-10: Battery cell production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Battery cell		1	kg	
Anode	0.31		kg	
Cathode	0.50		kg	
Electrolyte	0.16		kg	
Separator	0.022		kg	
Cell container	6.70×10^{-3}		kg	
Decarbonised water	3.80×10^{2}		kg	GLO: market for water, decarbonised, at user
Electricity	1.01×10^{2}		MJ	46%: KR: electricity production, hard coal 33%: KR: electricity production, nuclear, pressure water reactor 15%: KR: electricity production, natural gas, conventional power plant 4.4%: KR: electricity production, oil 1.4%: KR: electricity production, hydro, run-of-river 0.15%: KR: electricity production, wind, 1-3MW turbine, onshore 0.05 % KR: electricity production, photovoltaic, 570kWp open ground installation, multi-Si

Rail transport	0.26		tkm	RER: market group for transport, freight train
Lorry transport	0.1		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3
Waste heat		100	MJ	

Table S2-11: Anode Production

Description	Input	Output	Unit	Ecoinvent 3.5 Process
Anode		1	kg	
Negative electrode paste	0.43		kg	
Negative current collector Cu	0.57		kg	
Laser etching	0.1		h	GLO: market for laser machining, metal, with YAG-laser, 30W power
Rail transport	0.37		tkm	RER: market group for transport, freight train
Lorry transport	0.1		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3

Table S2-12: Mixing of anode active material

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Negative electrode paste		1	kg	
Silicon, electronics grade	0.96		kg	DE: silicon production, electronics grade
Acrylic acid	0.02		kg	RER: acrylic acid production
Carboxymethyl cellulose	0.02		kg	RoW: carboxymethyl cellulose production, powder
Rail transport	1.2		tkm	RER: market group for transport, freight train
Lorry transport	0.19		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3
Chemical plant	4×10^{-10}		pcs.	RER: chemical factory construction, organics
1-Methyl-2-pyrrolidinone		0.94	kg	1-Methyl-2-pyrrolidone [Group NMVOC to air]

Table S2-13: Positive active material production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
$Li[N_{1/3}C_{1/3}M_{1/3}]O_2$		1	kg	
Lithium carbonate	0.32		kg	GLO: market for lithium carbonate
$N_{1/3}C_{1/3}M_{1/3}(OH)_2$	0.96		kg	
Chemical plant	4.6×10^{-10}		pcs.	RER: chemical factory construction, organics
Electricity	25.2		MJ	RAS: market group for electricity, low voltage

Table S2-14: NCM hydroxide production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
$N_{1/3}C_{1/3}M_{1/3}(OH)_2$		1	kg	
Cobalt sulphate	0.30		kg	
Manganese sulphate	0.29		kg	GLO: market for manganese sulfate
Nickel sulphate	0.89		kg	GLO: market for nickel sulfate
Sodium hydroxide	0.89		kg	RER: soda production, solvay process
Chemical plant	4×10^{-10}		pcs.	RER: chemical factory construction, organics
Heat	42.6		MJ	RoW: market for heat, from steam, in chemical industry
Sodium sulphate		1.6	kg	Sodium sulphate [Inorganic emissions to fresh water]

NMC811-Si

Table S2-15: Battery cell production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Battery cell		1	kg	
Anode	0.31		kg	
Cathode	0.50		kg	
Electrolyte	0.16		kg	
Separator	0.022		kg	
Cell container	6.70×10^{-3}		kg	
Decarbonised water	3.80×10^{2}		kg	GLO: market for water, decarbonised, at user
Electricity	1.01×10^{2}		MJ	46%: KR: electricity production, hard coal 33%: KR: electricity production, nuclear, pressure water reactor 15%: KR: electricity production, natural gas, conventional power plant 4.4%: KR: electricity production, oil 1.4%: KR: electricity production, hydro, run-of-river 0.15%: KR: electricity production, wind, 1-3MW turbine, onshore 0.05 % KR: electricity production, photovoltaic, 570kWp open ground installation, multi-Si

Rail transport	0.26		tkm	RER: market group for transport, freight train
Lorry transport	0.1		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3
Waste heat		100	MJ	

Table S2-16: Anode Production

Description	Input	Output	Unit	Ecoinvent 3.5 Process
Anode		1	kg	
Negative electrode paste	0.43		kg	
Negative current collector Cu	0.57		kg	
Laser etching	0.1		h	GLO: market for laser machining, metal, with YAG-laser, 30W power
Rail transport	0.37		tkm	RER: market group for transport, freight train
Lorry transport	0.1		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3

Table S2-17: Mixing of anode active material

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Negative electrode paste		1	kg	
Silicon, electronics grade	0.96		kg	DE: silicon production, electronics grade
Acrylic acid	0.02		kg	RER: acrylic acid production
Carboxymethyl cellulose	0.02		kg	RoW: carboxymethyl cellulose production, powder
Rail transport	1.2		tkm	RER: market group for transport, freight train
Lorry transport	0.19		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3
Chemical plant	4×10^{-10}		pcs.	RER: chemical factory construction, organics
1-Methyl-2-pyrrolidinone		0.94	kg	1-Methyl-2-pyrrolidone [Group NMVOC to air]

Table S2-18: Positive active material production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
$Li[N_{1/3}C_{1/3}M_{1/3}]O_2$		1	kg	
Lithium hydroxide	0.21		kg	GLO: market for lithium hydroxide
$N_{1/3}C_{1/3}M_{1/3}(OH)_2$	0.96		kg	
Chemical plant	4.6×10^{-10}		pcs.	RER: chemical factory construction, organics
Electricity	25.2		MJ	RAS: market group for electricity, low voltage

Table S2-19: NCM hydroxide production

Description	Input	Output	Unit	Ecoinvent 3.6 Process
$N_{1/3}C_{1/3}M_{1/3}(OH)_2$		1	kg	
Cobalt sulphate	0.14		kg	
Manganese sulphate	0.15		kg	GLO: market for manganese sulfate
Nickel sulphate	1.18		kg	GLO: market for nickel sulfate
Sodium hydroxide	0.89		kg	RER: soda production, solvay process
Chemical plant	4×10^{-10}		pcs.	RER: chemical factory construction, organics
Heat	42.6		MJ	RoW: market for heat, from steam, in chemical industry
Sodium sulphate		1.6	kg	Sodium sulphate [Inorganic emissions to fresh water]

S3. Stoichiometry calculations

The life cycle inventory of a lithium-ion battery with a NMC111 cathode with a graphite anode is taken from Ellingsen et. al. This chemistry is altered through adjusting $N_xM_yC_z$ ratio as well as the substituting the anode material (graphite for silicon). The 6 cell cases studied, including the base case, are as follows: NMC111-Gr, NMC622-Gr, NMC811-Gr, NMC111-Si, NMC622-Gr, and NMC811-Si. By changing the ratio of $N_xM_yC_z$ – x=Ni content, y= Mn content, and z= Co content – in the cell's positive electrode paste (cathode), we expect to see a change in environmental impacts, specifically as a function of Ni and Co content increase.

N/P Ratio

N/P ration is the ratio of the capacity of the negative electrode (anode) to the positive electrode (cathode). The N/P ratio used is set to reflect values commonly used in literature which are 1.1 and 1.2 for graphite and silicon, respectively. To verify these ratios are maintained in our cells, theoretic areal capacity was calculated for both anode (a) and cathode (c) using their theoretical capacity (TC), Mass loading (ML), and % active material (AM) for each cell architecture. according to the following equation:

$$\text{Nominal Capacity } (\text{mAh cm}^{-2}) = \frac{\text{TC}_\text{a}(\text{mAh g}^{-1})\times \text{ML}_\text{a}(\text{g cm}^{-2})\times \text{AM}_\text{a}}{\text{TC}_\text{c}(\text{mAh g}^{-1})\times \text{ML}_\text{c}(\text{g cm}^{-2})\times \text{AM}_\text{c}} \times \text{ML}_{\text{a+b}} \times \text{AM}_{\text{a+b}} \quad \text{Eq. S3-1}$$

Cell Chemistry Calculations

It is important to note, while Al and Cu current collectors are known to have disproportionate environmental impacts in NMC battery lifecycles, in this LCA study their impacts were treated as a constant similar to Kallitsis et al. (2020) This was achieved by keeping the Cu and Al current collector mass and area constant across cell architectures. Essentially, constraining their impacts by keeping dimensions and mass of the cell constant and assuming only changes in the active electrode material changed mass. This simplifying assumption allows us to only consider the effects that changing NMC cell chemistry (both anode and cathode) has on environmental impacts in the LCA. Kallitsis et al. use a similar process to modify the base case to construct a mathematical constant representing this simplifying assumption, where only changes in mass of the active material –in our case both NMC chemistry and anode material from graphite to silicon– are considered in calculation for cell capacity.

Due to compositional differences each mass will vary slightly for NMC-111, NMC-622, and NMC-811 chemistry based on the NMC mass calculations below:

$$\mathrm{N_xM_yC_z} = \mathrm{LiNi_xMn_yCo_z(O_2)} \quad \text{Eq. S3-2}$$

Table S3-1: The molar mass of materials used in equation S2-3 to S2-5.

Element	**Molar Mass (g/mol)**
Li	6.94
Ni	58.693
Mn	54.938
Co	58.933
O	16

$$\mathrm{NMC111} = 1 \times 6.94 + .33 \times 58.693 + .33 \times 54.938 + .33 \times 58.933 + 2 \times 16 \quad \text{Eq. S3-3}$$

$$\mathrm{NMC111} = 95.886\ \mathrm{g/mol}$$

$$\mathrm{NMC622} = 1 \times 6.94 + .6 \times 58.693 + .2 \times 54.938 + .2 \times 58.933 + 2 \times 16 \quad \text{Eq. S3-4}$$

$$\mathrm{NMC622} = 96.93\ \mathrm{g/mol}$$

$$\mathrm{NMC111} = 1 \times 6.94 + .8 \times 58.693 + .1 \times 54.938 + .1 \times 58.933 + 2 \times 16 \quad \text{Eq. S3-5}$$

$$\text{NMC811} = 97.93\ \text{g/mol}$$

Since the source of the metal cations each originate from one source throughout the synthesis of NMC, there is direct correlation between the molar fraction composition of the metals in NMC and the amounts of metal sulfate precursors used in the co-precipitation (i.e. Eq. S3-6, the "x" in the coefficient of $NiSO_4$ is equal to the "x" subscript in $Ni_xCo_yMn_z(OH)_2$). This reaction is based on the mass control over x, y, and z, in the metal sulfate precursors and subsequent control over Ni, Mn, Co content in the co-precipitation step in the reaction for the NMC hydroxide that is used to synthesize the NMC. Using these values to accurately assess the required mass of precursors, the inventories of the unique NMC chemistries were calculated based on the well-studied co-precipitation reaction below.

$$x\ \text{NiSO}_4 + y\ \text{CoSO}_4 + z\ \text{MnSO}_4 + 2\ \text{N}aOH \rightarrow \text{Ni}x\text{Co}y\text{Mn}z(\text{OH})_2 + \text{Na}_2\text{SO}_4 \qquad \text{Eq. S3-6}$$

Since Na_2SO_4 is a waste product, any loss during the synthesis of the $N_xM_yC_z(OH)_2$ is contained in its mass. This aligns well with other LCA's subpack inventories and literature (Ellingsen et al., 2014; Kallitsis et al., 2020)

Anode

In calculating the mass changes of the anode paste, the thickness of the coating was kept at a constant range of 45-75 µm based on literature (Ellingsen et al. (2014) SI 2.1). Due to the manufacturing process, the coating of the anode paste is constrained to the same area dimensions as the negative current collector (the current collectors are 10-15 cm by 15-25 cm and are between 10-30 µm thick), in this case Cu. Thus, anode mass active material of the anode (AM_a) was calculated as a range using the area (A) and coating thickness (CT) of the base case NMC111-Gr cell using the equation below:

$$\mathrm{AM_a(g) = CT_a(\mu m) \times A_a(cm^{2-}) \times \ TC_{a+b}(g\ cm^{3-})} \qquad \text{Eq. S3-7}$$

It is noteworthy that the theoretical density of the active material changes as we change our anode active material from graphite to silicon in our studies. Units are adjusted to pack level as the final step in the calculations

Cathode

Similar calculations to the anode active materials were made for the cathode since both anode and cathode active material coatings are constrained to the dimensions of the cell. The coating was kept at a constant range of 45-75 μm based on literature (Ellingsen SI 2.1).(Ellingsen et al., 2014) The mass of active material for the cathode (AM_b) was calculated using the equations below:

$$\mathrm{AM_c(g) = CT_c(\mu m) \times A_c(cm^{2-}) \times \ TC_{a+b}(g\ cm^{3-})} \qquad \text{Eq. S3-8}$$

Mass adjustments for each $N_xM_yC_z$ chemistry (e.g. NMC111, NMC622, and NMC811) were made as a function of theoretical density changes due to the varying compositions of Ni, Mn, and Co.

S4. Battery end-of-life life cycle inventory (LCI)

Recycling of the battery requires dismantling from the EV, mechanical crushing, and then a hydrometallurgical process to recover constituent materials. The required energy for dismantling is taken from Hawkins et al. (2013).

Table S4-1: Recycling process

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Recycling of lithium-ion battery		1	kg	
Hydrometallurgical process	1		kg	GLO: treatment of used Li-ion battery, hydrometallurgical treatment
Dismantling from EV	.023		kg	US: Market for electricity
Mechanical treatment	1		kg	GLO: treatment of waste electric and electronic equipment, shredding
Lorry transport	.004		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3

The repurposing of the battery requires dismantling from the EV and testing the state of health. The amount of energy (E) used for state of health testing is dependent on the capacity (C) of the lithium-ion battery chemistry (b). It is assumed the battery is received at 80% capacity and 80% efficiency. Thus, the calculation for a full charge and discharge for cycling is based on Equation S3-9.

$$E = \frac{.8 * .8 * C_b}{253} \qquad \text{Eq. S4-9}$$

Table S4-2: Repurposing process

Description	Input	Output	Unit	Ecoinvent 3.6 Process
Repurposing of lithium-ion battery		1	kg	
Dismantling from EV	.023		kg	US: Market for electricity
State of health testing	E		kg	US: Market for electricity
Lorry transport	.004		tkm	RER: market for transport, freight, lorry >32 metric ton, EURO3

S5. Comparing results of NMC111-Gr with Kallitsis et al. (2020) and Ellingsen et al. (2014)

The results from this study were also calculated for the ReCiPe 2008 indicators to do a direct comparison with Ellingsen et al. and Kallitsis et al. The results in Table S1 are the results of the three studies for NMC111-Gr battery, with only the EcoInvent inventory differing. There are a few large variances in our results compared to those of the others for FEP, TETP, and HTP. After an in-depth review, it has been concluded that the updated LCI of copper is resulting in this increase. To confirm, copper in our analysis was replaced with the 3.5 EcoInvent inventory, which resulting in a closer range to the findings in the other studies. This micro analysis was not used in the results of the study; EcoInvent 3.6 (Burhan et al., n.d.) is used for all inventories and ReCiPe 2016 (Huijbregts et al., 2017) for all indicators.

Table S5-1: The midpoint impacts of a NMC111-Gr battery.

ReCiPe 2008 midpoint indicator	Ellingsen et al 2014	Kallitsis et al 2020	Dunn et al 2022	Dunn et al 2022
	EcoInvent 3.1	EcoInvent 3.5	EcoInvent 3.6	EcoInvent 3.6 & copper replaced with 3.5 value
GWP (CO2 eq.)	4,580.00	4,990.00	3,246.99	3,313.20
FDP (m3)	1,320.00	1,390.00	1,067.98	1,088.20
ODP (CFC-11 eq.)	3.00E-04	4.00E-04	1.76E-04	1.81E-04
POFP (NOx eq.)	18.00	22.50	19.12	20.12
PMFP (PM2.5 eq.)	16.00	18.30	20.67	20.83
TAP (SO2 eq.)	51.00	61.00	64.53	65.38
FEP (P-eq)	8.00	8.40	4.03	7.92
MEP (N eq.)	6.40	2.10	1.28	1.58
FETPinf (1,4 DB eq.)	256.00	478.00	613.88	716.42
METPinf (1,4 DB eq.)	276.00	445.00	577.42	665.87
TETPinf (1,4 DB eq.)	1.30	2.60	154.43	6.58
HTPinf (1,4 DB eq.)	15,900.00	15,700.00	9,118.57	16,272.75
MDP (Cu-eq)	4,100.00	3,820.00	4,935.68	5,294.10

S6. Supplementary results

Figure S6-1: The ReCiPe 2016 endpoint impacts of an LIB battery with a NMC111, and NMC811 cathode and with a graphite and silicon anode.

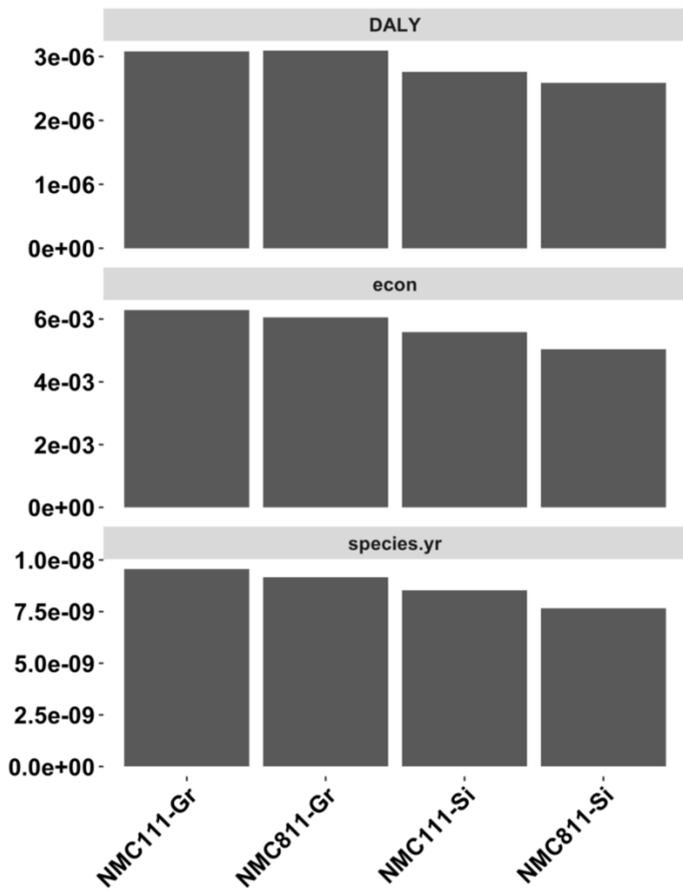


Figure S6-2: The ReCiPe 2016 midpoint impacts of an LIB battery with NMC111and NMC811 cathode and with a graphite and silicon anode.

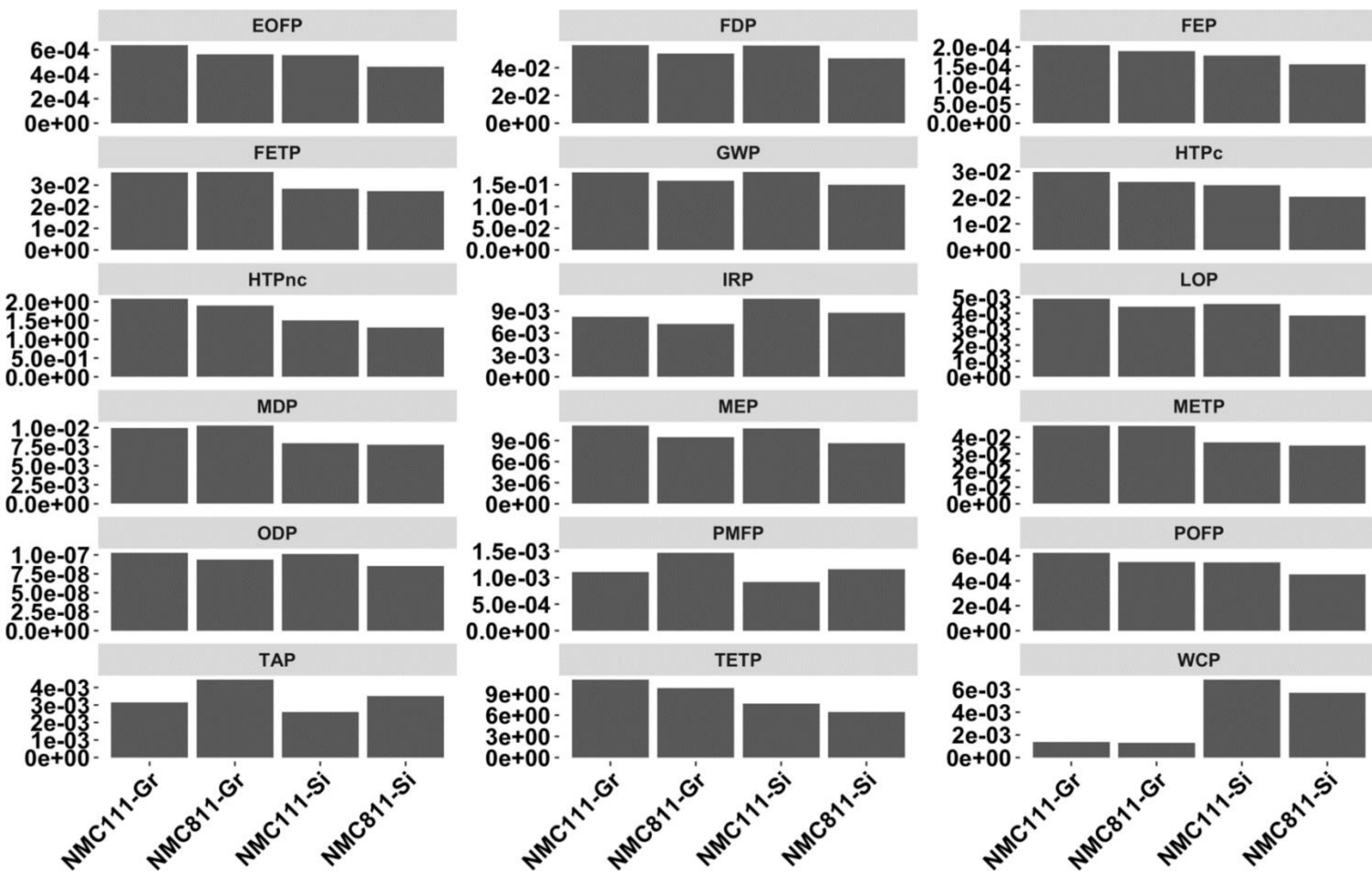


Figure S6-3: The environmental impacts of each scenario for the ReCiPe 2016 endpoint indicators.

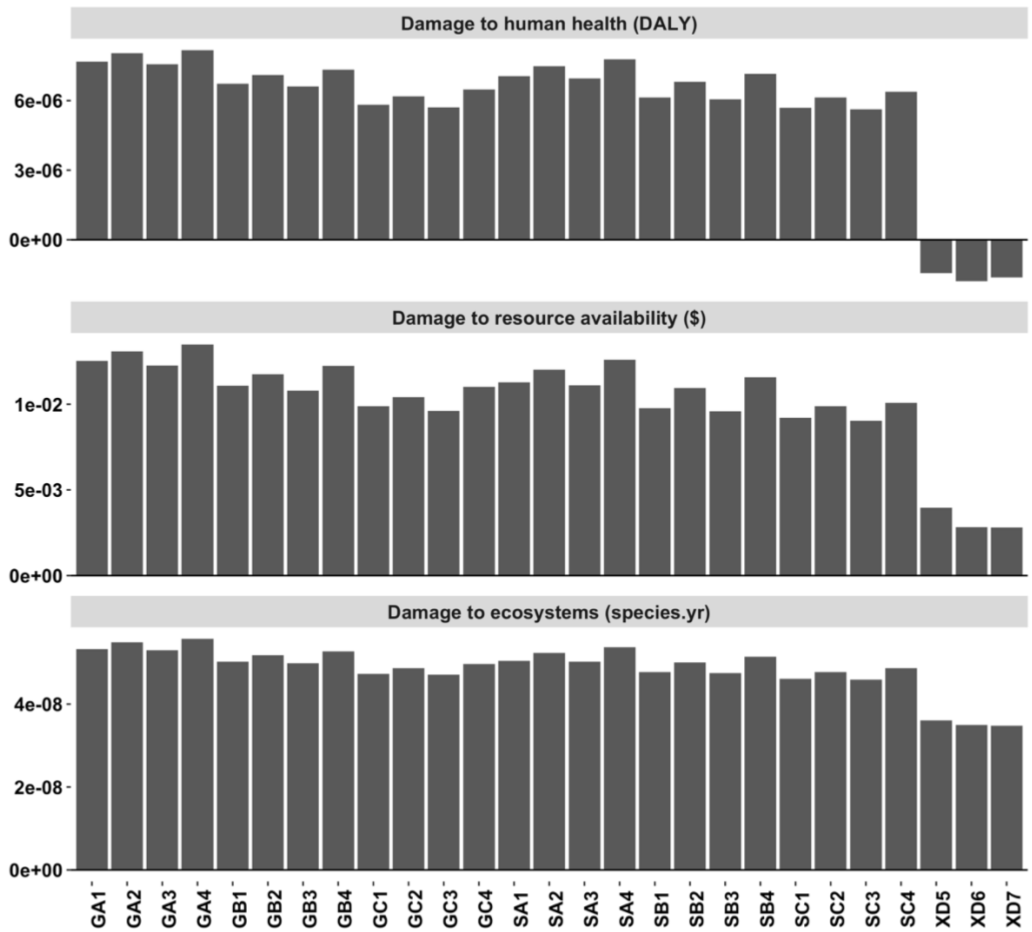


Figure S6-4: The net environmental impacts of all scenarios using ReCiPe 2016 midpoint indicators.

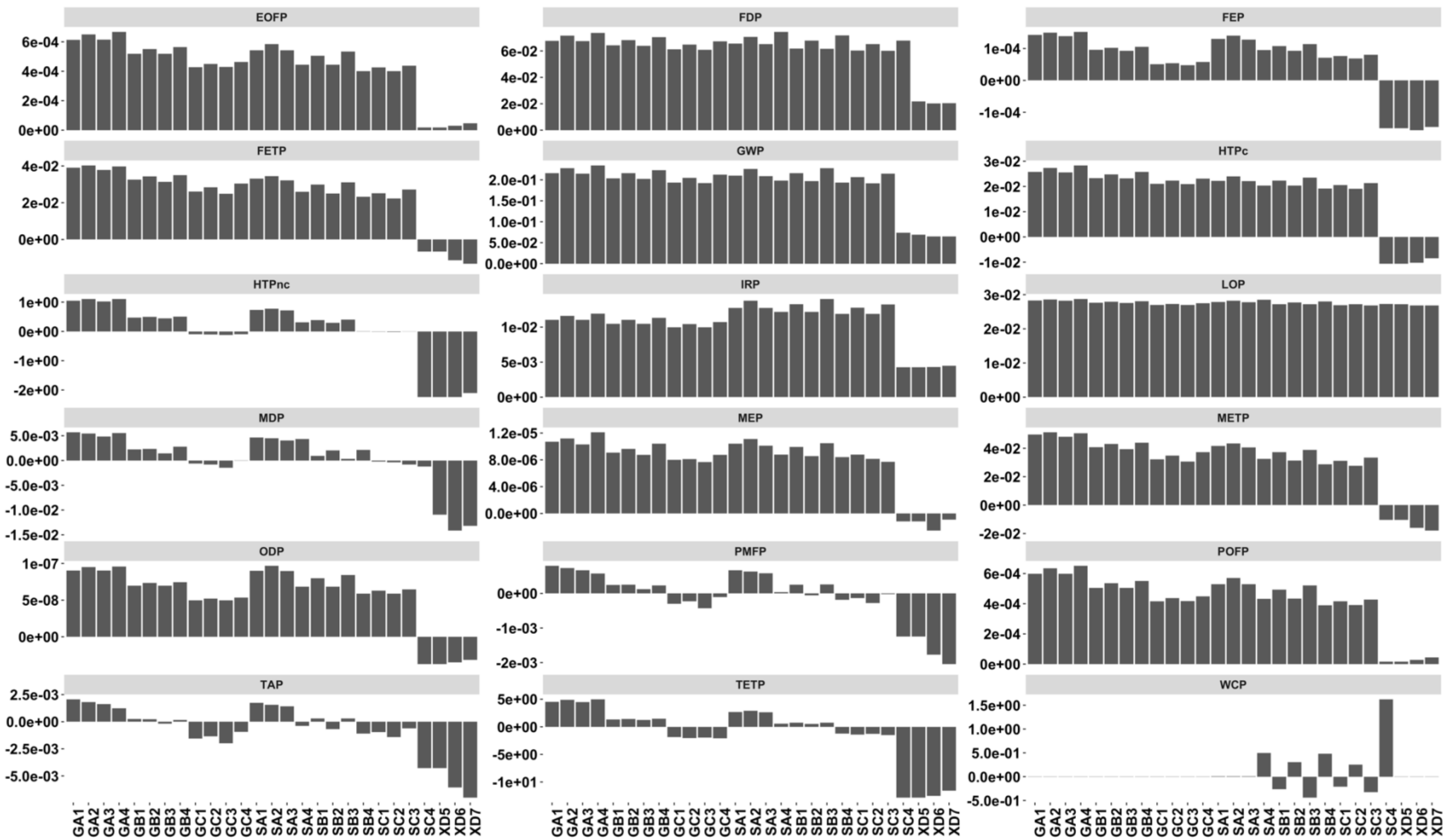


Figure S6-5: The scenarios and the environmental impacts of the graphite battery, displayed by battery component, remanufacturing, and recycling. The impacts are represented by ReCiPe 2016 midpoint indicators.