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GREEN CHEMISTRY AND THE FUTURE OF LITHIUM BATTERY RECYCLING

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Abstract

Spent batteries, which are omnipresent in modern life, have a dual challenge: disposal causes environmental damage, but recycling offers a transformational alternative. Battery recycling is a critical step towards more sustainable resource management. Recycling not only conserves resources, but also reduces the need for dangerous mining operations by preserving important elements like lithium, cobalt, and nickel. This strict recycling strategy dramatically decreases carbon footprint, promoting a circular economy in which materials are used rather than discarded. Nonetheless, life cycle assessment (LCA) is an effective method for guiding the development of higher-performing batteries with a lower environmental impact. This study investigates typical procedures in lithium-ion battery life cycle assessments and gives recommendations for future studies that are more interpretable, representative, and effective. Proper disposal keeps hazardous compounds from seeping into the environment, decreasing soil and water contamination. However, challenges persist, including technological limitations, cost-effectiveness, and the global demand for standard recycling methods. Increased public awareness and concerted efforts among businesses, governments, and consumers are necessary to optimize the environmental benefits of battery recycling, assuring a brighter future for future generations.

Keywords: Spent Batteries, Recycling, Sustainable Resource Management, Conservation Of Resources, Dangerous Mining Operations, Carbon Footprint, Life Cycle Assessment (LCA), Lower Environmental

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Impact, Hazardous Compounds, Soil Contamination, Water Contamination, Cost-Effectiveness, Global Demand, Public Awareness, Environmental Benefits.

1. INTRODUCTION

Information technology (IT) advanced substantially in the 1980s with the introduction of portable electronic devices such as video cameras, mobile phones, and computers. This technological revolution created an increasing need for rechargeable batteries with higher capacity or less size and weight for the same capacity. Conventional rechargeable batteries available or under development at the time, such as lead-acid, nickel-cadmium, and nickel-metal hydride batteries, employed aqueous electrolytes, which limited their ability to increase energy density while decreasing size and weight. Thus, there was still an unmet demand for a novel, tiny, and lightweight rechargeable battery to be put into practical use. The initial commercialization of the lithium-ion battery (LIB) occurred in 1991, after research began in the early 1980s. Since then, LIBs have developed to be the primary power storage solution for portable IT devices. Energy storage is critical to the fast decarbonization of the electric grid and transportation sector, addressing the requirement for short-term power storage on the grid while also allowing electric vehicles (EVs) to store and consume energy on demand.

In recent years, China has become a major producer of lithium-ion batteries (LIBs), which require critical components such as lithium, cobalt, and graphite. The number of battery electric cars (BEVs) on the road is expected to exceed 130 million worldwide by 2030 (Grushevenko, 2020). The need for LIBs is expected to grow dramatically, with a global demand of 9300 GWh by 2030. This expansion in the BEV industry is predicted to be driven mostly by battery technology, which will have a substantial influence on LIB production and disposal in the near future. New energy cars, particularly pure electric vehicles, use less energy and emit less pollutants into the atmosphere than gasoline-powered vehicles. However, essential material usage and upstream environmental implications from manufacture are frequently noted as disadvantages of the widespread use of rechargeable batteries. Life-cycle assessment (LCA) is a popular method for determining the possible effects of large-scale battery manufacture, usage, disposal, and/or recycling. A thorough LCA includes all of the product's life cycle stages. Many studies have examined the effects of the NMC battery manufacture stage, but they have not addressed the other life cycle stages, frequently ignoring the End-of-Life (EoL) and Use stages.

Too far, there has been no agreement in the field of LCA on how to assess the environmental effect of batteries or how to present the findings. Studies employ a broad range of system boundaries, functional units, main data sources (which give data at various degrees of granularity), and life-cycle inventory, midpoint, and effect classifications. This complicates cross-technology comparisons and hinders LCA's potential to serve as a feedback loop for early scientific research and technological development. It can also limit our capacity to find and remedy inaccuracies in the literature; life-cycle inventory results frequently differ by one or more orders of magnitude

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throughout the literature, and most evaluations are unable to explain the underlying reason of the disparities.

This work contributes to the field of life-cycle assessment research by doing a complete cradle-to-grave lifecycle analysis on a lithium-ion battery system that is presently in the design phase and is intended for ESS applications. This study gives a more in-depth assessment of the battery system's overall environmental effect and identifies hotspots along the production chain. The findings can assist battery producers build more sustainable products and identify crucial elements that influence the environmental performance of battery packs.

2. LITHIUM-ION BATTERY TECHNOLOGIES

LIBs are the most often used battery chemistry, and while this study will not get into the specifics of the technologies, it is useful to quickly outline the most prevalent forms of LIBs investigated in the present literature. Non-LIB battery technologies, such as sodium-ion batteries, potassium-ion batteries, solid-state batteries (Li-metal, Li-sulfur, and rechargeable zinc alkaline), flow batteries, and multivalent batteries, have been researched, but LIBs are expected to continue to dominate the market in the near future. LIBs are commonly classified according to the cathode material: lithium manganese oxide (LMO), lithium nickel manganese cobalt oxide (NMC), lithium iron phosphate (LFP), and lithium nickel cobalt aluminum oxide (NCA). Most batteries investigated in previous LCA studies include a graphite carbon anode. The specific energy of these batteries at the cell level ranges from 90 to 250 Wh kg^{-1} (Jason Porzio, 2021).

BESS are often built-up of modular battery packs that can be added in series and parallel to reach specific grid requirements. BESS are often evaluated based on their energy density, round-trip efficiency, and cycle life. The energy density can be expressed as the volumetric or gravimetric energy density, which is defined as the amount of energy that can be stored in a unit volume or weight. The LIB cell manufactured at the current gigafactory is a cylindrical cell of the 21,700-type designed for automotive applications. This cell type has an outside diameter of 21 mm, measures 70 mm in length, and weighs between 67 and 69 g. The investigated cell chemistry is NMC-8:1:1, with the ratio 8-1-1 indicating the active cathode material composition of 80% nickel, 10% manganese, and 10% cobalt. The model is applicable for cells with an energy density of 210-240 Wh/kg. The active anode material is composed of synthetic graphite. The positive and negative current collectors are comprised of aluminum and copper foil, respectively. The mass and chemical composition of the active cathode material are critical factors of the cell's storage capacity and power, respectively.

2.1 Background:

The growth of lithium-ion battery (LIB) manufacture in recent years has been fueled mostly by the transition away from combustion engines and toward electric cars. The electrification of road transportation, particularly in Europe and China, has pushed both technological breakthroughs in LIB and cost reductions in production, making LIB more

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cost-competitive and scaleable. Some of the elements used in LIBs, such as cobalt and lithium, are associated with human toxicity and geopolitical danger. The detrimental impacts of mineral mining vary by area. As a result, policymakers and battery producers are both interested in LIB's long-term viability.

3. LIFE-CYCLE ASSESSMENT OVERVIEW

When comparing different types of batteries in terms of their environmental impact, or simply trying to understand how making and using more batteries affects the environment, we use a method called Life Cycle Assessment (LCA). This method has four main phases that are crucial for a meaningful study: a) Goal and Scope Definition: This is where researchers decide what question they want to answer with their study. They figure out things like what parts of the battery's life cycle to look at, what environmental factors to measure, and what units of measurement to use, b) Inventory Analysis: Here, researchers gather data on everything involved in making and using the batteries, from raw materials to energy use, c) Impact Assessment: This phase looks at the data from the inventory analysis to see how different parts of the battery's life cycle impact the environment, d) Interpretation: Finally, researchers interpret all the data they've collected to draw conclusions and make recommendations based on their findings. For batteries, LCA results can help improve battery technology to reduce environmental harm, compare different types of batteries for specific uses, or predict the environmental effects of using lots of batteries in things like electric cars or power grids (contributors, 2024).

One tricky thing about using LCA for batteries is that they store energy, and how they're used affects their lifespan and environmental impact in ways that aren't always easy to predict. So, when comparing different battery types for the same use, it's best to focus on the service they provide rather than just looking at the raw materials or production process.

There are different ways to define the scope of an LCA study. Some studies only look at the production phase (from raw materials to when the battery is made), while others also include how the battery is used and what happens to it at the end of its life (like recycling or disposal). The latter is called a 'cradle-to-grave' study, while the former is 'cradle-to-gate.' There's also a term called 'cradle-to-cradle,' which refers to systems that aim for zero waste through recycling, but it's not commonly used for battery life cycles, even if recycling is part of the process.

3.1 The Life Cycle of Stationary and Vehicle Li-Ion Batteries:

The standard lifecycle for LIBs starts with extracting raw materials and then processes them into materials suitable for components, manufacturing cells which are used for module assemblies before being assembled into packs that can be placed inside cars or trucks etcetera. It can also involve putting them together into racks within the site itself before being transported outwards by ship to other places where power is stored docks. So, after laying out the basics, researchers have to think about whether they want to focus on a specific situation and, if they do, whether to look at everything from when the battery

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is first used to when it's no longer usable (its end-of-life). Even if they're only studying up to the point when the battery is made (a 'cradle-to-gate' approach), researchers need to be clear about whether they're talking about batteries in their simplest form, like a module or pack, or if they mean the whole assembled setup, like a rack for stationary storage (Jason Porzio, 2021).

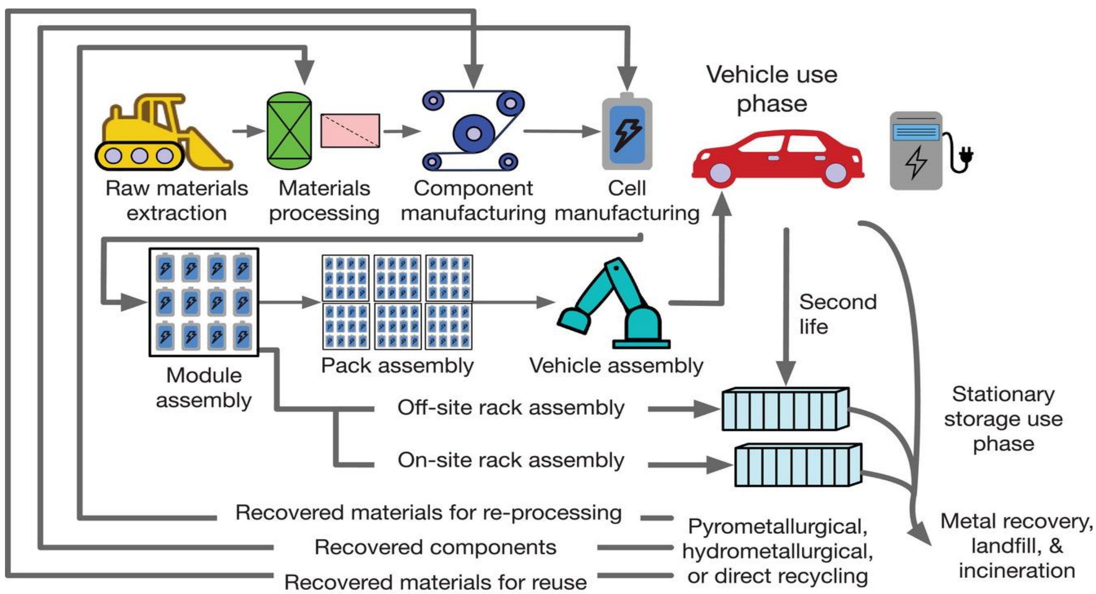

Figure 1: Major life-cycle Stages for Vehicles and Stationary Batteries

However, not all studies include battery use phase for a specific application, nor is this always feasible for more advanced, pre-commercialization battery technologies. For a use-agnostic cradle-to-gate analysis of a LIB, researchers must still select a pack or rack configuration that is tied to a stationary or EV application. The system boundary may need to be specified at the module assembly stage in a genuinely use-agnostic LCA, because the construction of the pack or rack (containing components such as thermal management and electrical control) varies significantly depending on how the battery will be used (see **Figure 1**). Cradle-to-grave life cycle assessments address how batteries will be used and processed at the end of their life, including collection, recycling, and/or disposal.

4. WHAT IS PRODUCT ENVIRONMENT FOOTPRINT

The Product Environmental Footprint (PEF) is like a detailed guide that works alongside the Life Cycle Assessment (LCA) method, which is part of the ISO standards. The PEF method is focused on measuring how products impact the environment, and it's especially important in the European Union (EU). For batteries, there's a specific set of guidelines called the Batteries Product Environmental Footprint Category Rules (PEFCR). These rules make LCA studies about batteries more consistent and easier to compare because they provide clear details and guidelines (Liu, 2020).

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The Batteries PEFCR are designed for high-specific energy mobility applications and were created in partnership with industry partners and the worldwide non-profit organization Recharge. However, the Batteries PEFCR presently solely addresses high-specific energy mobile applications, with no approach for BESS applications. This study will consequently follow The Battery PEFCR for High Specific Energy Mobile Applications to the greatest degree feasible for BESS (European Commission 2018). Furthermore, the Batteries PEFCR method only includes thorough details of battery manufacturing, beginning with electrode fabrication. The approach has not been fully developed to describe battery cell manufacture with upstream operations, such as Northolt's, which includes chemical preparation and precursor synthesis. However, the Batteries PEFCR approach is now being developed, and a technical secretariat led by RECHARGE will examine and update the present methodology.

The Product Environmental Footprint (PEF) is a multi-criteria assessment of a product or service's environmental performance across its entire life cycle (Advanced Rechargeable & Lithium Batteries Association, 2018). The goal of PEF information is to reduce the environmental effect of goods and services while considering end-to-end supply chain activities. However, it is vital to highlight that normalization and weighting factors have intrinsic subjective values that are determined by policy and the developers' preferences.

5. OPENLCA OVERVIEW

Green-Delta created OpenLCA, an open-source program for life cycle assessment (LCA) and sustainability assessment, in 2007. It is free and open-source software. The world's most powerful open source Life Cycle Assessment program. openLCA is an open source and free software for sustainability and life cycle assessment that includes calculation of your sustainability assessment and/or life cycle assessment (LCA), detailed insights into calculation and analysis results, best-in-class import and export capabilities, easy model sharing, life cycle costing and social assessment seamlessly integrated into the life cycle model, and more.

5.1 Steps for Working:

- OpenLCA is available in both 64-bit and 32-bit versions for Windows installation. This article focuses on the 64-bit version. You will need administrator access to complete the installation.
- After installation, launch openLCA. Because openLCA starts with no data, the 'navigation' area on the left is empty.
- To create a new database, right-click in the navigation box. To begin, it is advised to construct the database using the following settings: 'local database' and 'full reference data'. After a few seconds, you may view the freshly generated database (Andreas Ciroth, 2014).

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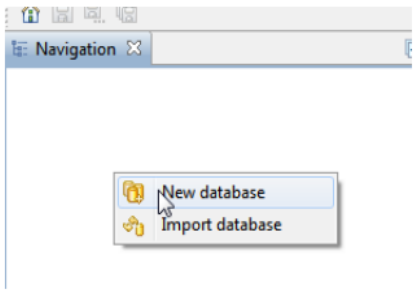
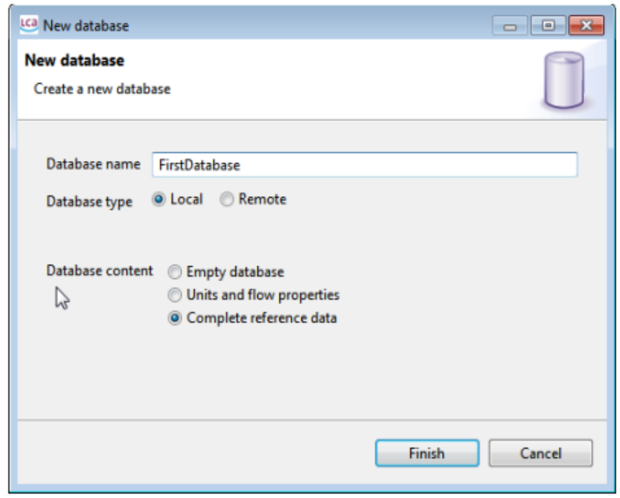
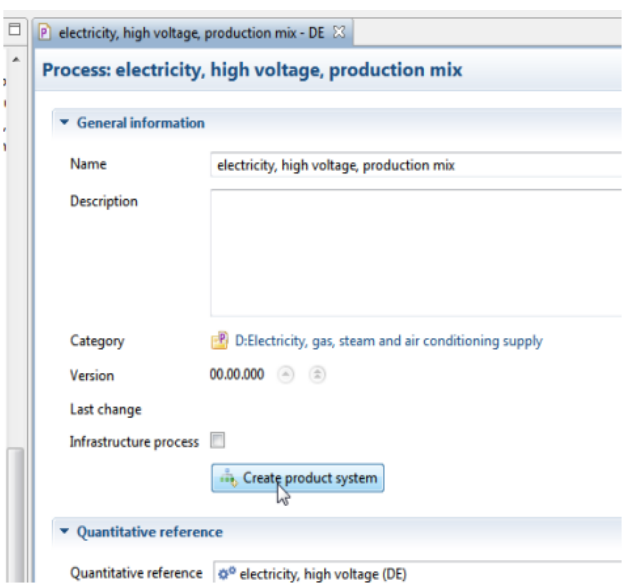
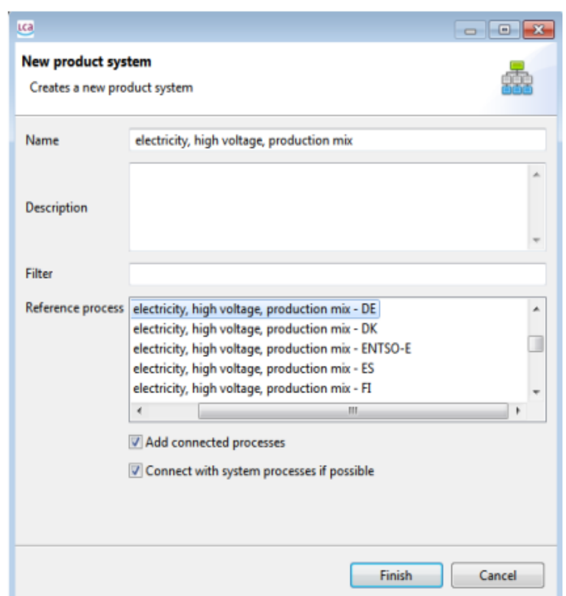




- You may also import an existing database. The database can be a converted openLCA 1.3 database, or you can import a database from the openLCA nexus website.
- Nexus datasets do not contain LCIA techniques ('impact methods' in the category tree). To include them, get the LCIA method bundle from the openLCA download page.
- We will now import the following into the ecoinvent database: We choose 'database import' from File / Import menu. This import may take a few minutes. Once completed, the LCIA procedures will be available in the database.
- The first possibility is to create the product system from the process. Open a process data set, go to the page 'general information', and click on 'Create product system'.





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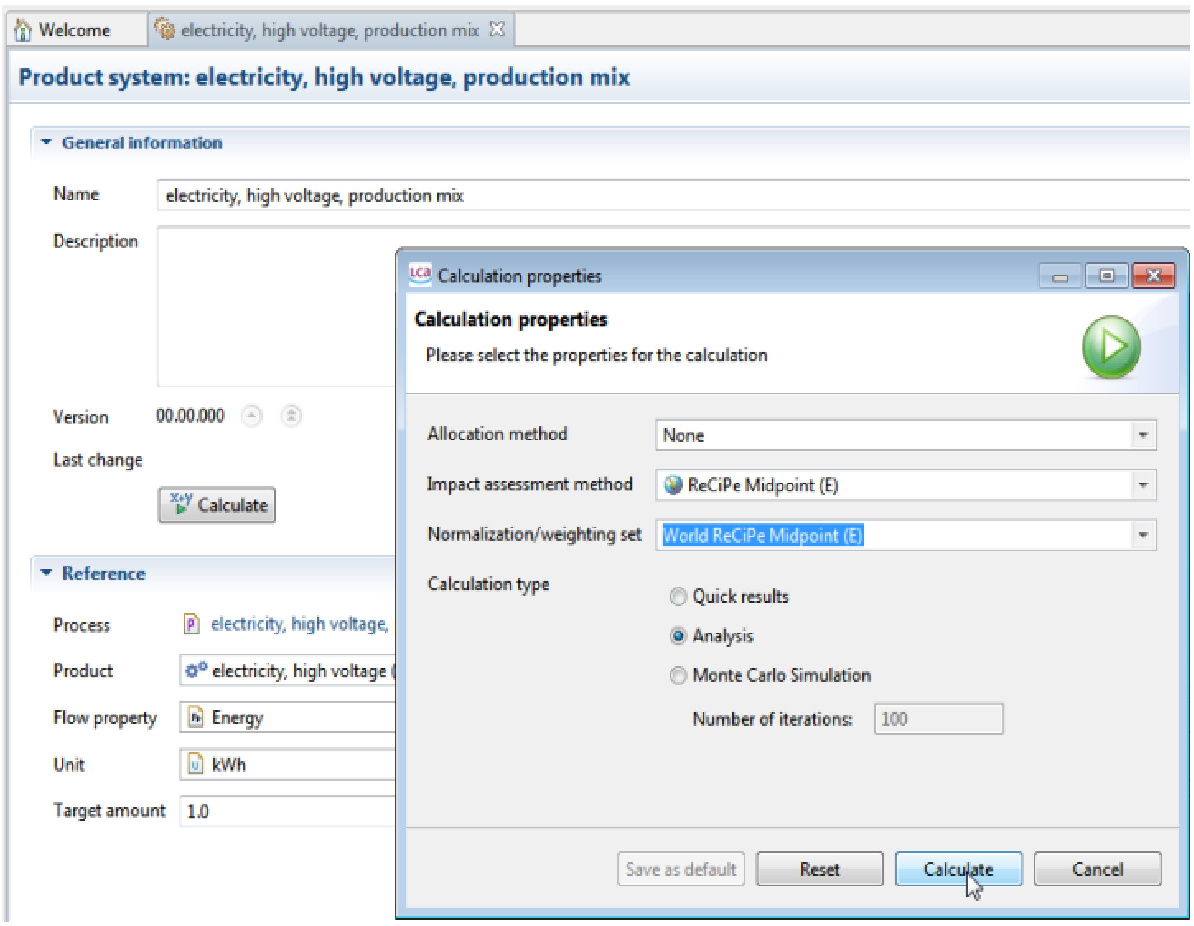
- The product system's model graph displays related processes. These links may be changed (deleted or increased for new product suppliers), and processes can be completely removed from the product system if they no longer have any life cycle connections.
- To calculate the life cycle, click 'compute' on the product system's general information tab. You can pick an LCIA technique and normalization set if applicable. You may also pick between rapid findings and in-depth analysis.



- Results are presented on many pages that are generally self-explanatory. Calculation in which just primary contributors, inventory, and effect assessment tables are generated; this short calculation is approximately twice as fast as the analytical calculation.

6. PAST, PRESENT & FUTURE OF LI-ION BATTERIES

6.1 How lib was born?

In the 1980s, the design of numerous types of audio/visual devices for outdoor usage began, and they were widely available in the market. Furthermore, the popularity of so-

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called information technology equipment, such as cellular phones, laptop computers, digital cameras, and so on, has increased since then. Although primary batteries dominated until the 1970s, secondary batteries like lead-acid and nickel-cadmium (Ni-Cd) gradually replaced them.

Ni-Cd, a popular small-sized secondary battery, has various shortcomings as a power source for portable electronics, including low energy density and environmental concerns. Ni-Cd performance improved significantly, but its energy density was limited by the end of the 1980s. LIB offers superior qualities as compared to typical secondary batteries such as Ni-Cd, nickel-metal hydride, and lead-acid batteries.

The characteristics of LIBs are as follows: a) High working voltage (3.7 V on average), b) High gravimetric and volumetric energy densities, c) No memory effect, d) Low self-discharge rate (less than 20% per year), and e) Operation throughout a wide temperature range (Pistoia, 2014).

6.2 Performance that Users Expect from LIB:

Based on the results using hard carbon negative electrodes, the performance was improved, and a realistic discharge capacity of 550 mAh/g was achieved. Graphite has a theoretical discharge capacity of 372 mAh/g and a real capacity of around 350 mAh/g. As a result, hard carbon is thought to be a very desirable anode material.

Graphite has a density of around 2.15-2.25 g/cm3, while hard carbon has a density of about 1.45-1.55 g/cm3. Graphite and hard carbon have volumetric discharge capacities of around 750-790 and 800-850 mAh/cm3, respectively, with a negligible variation in energy density.

When LCO is employed as the active material for a positive electrode, the average voltage of a graphite cell is 3.7 V and that of a hard carbon cell is 3.6 V, implying that the energy density of the former is 2.8-2.9 and that of the latter is 2.9-3.1 Wh/cm3.

Graphite batteries have an initial charge and discharge efficiency of around 95%, while hard carbon batteries are at about 85%. Also, graphite batteries tend to have a flat discharge curve, whereas hard carbon batteries show a more sloping curve (Liu, 2020).

1. Because cell size is often fixed, both volumetric and gravimetric energy density are relevant. From this perspective, the material with the lowest specific density is not preferred.
2. Cutoff voltage affects discharge capacity. A cell having a slanted discharge profile is unfavorable.
3. Ensure optimal initial charge and discharge efficiency.

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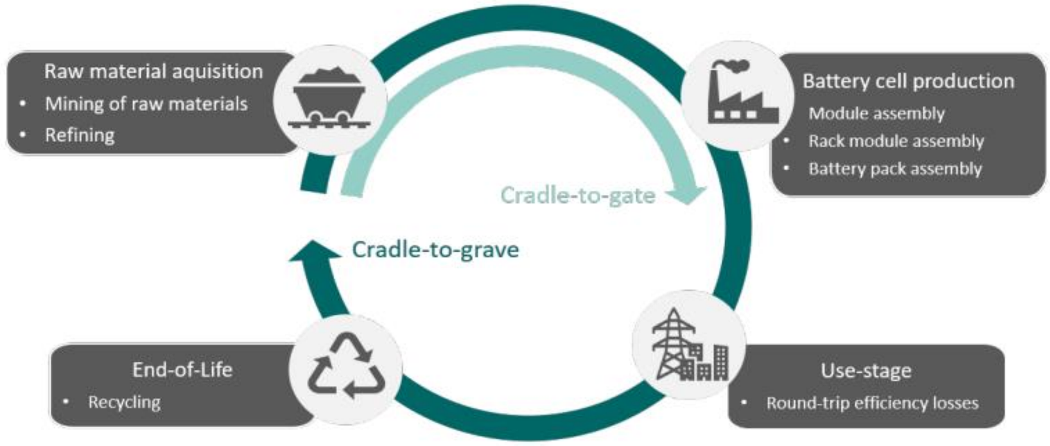

Figure 2: Product life-cycle Phase Included in Study

6.3 Improvement of LIB:

To meet the needed volumetric energy density, we added a graphite negative electrode to our third-generation LIBS. The newest 18650 cells have energy densities of 230 Wh/kg and 620 Wh/dm3. To increase energy density, innovative active materials for both negative and positive electrodes must be researched. Since the first release of lithium-ion batteries to the market in 1991, significant advances have been made. The earliest commercial LIBs had energy densities of 80 Wh/kg and 200 Wh/dm3. The current iteration of LIBS has energy density of more than 230 Wh/kg and 620 Wh/dm3. It is acknowledged, however, that new technologies are required to increase LIB performance, including energy density and safety properties. As a result, several new technologies have been shown that are scientifically appealing. Some of them, however, do not satisfy the specifications (Chengetai Portia Makwarimba, 2022).

Active materials for positive electrodes with excess lithium are now the focus of our research since they are predicted to have a high discharge capacity. Liz MnO3 is one of the options, and a solid solution of LiMO2 (M = Co, Ni, Mn) and Li2MnO3 has recently been studied. It was stated that this material has a discharge capacity of around 300 mAh/g, nearly double that of LiCoO2. However, this solid solution demonstrated low cycle performance. The charge voltage was gradually increased to 4.5, 4.6, 4.7, and 4.8 V, with each voltage resulting in two charge/discharge cycles, for a total of eight charge/discharge cycles. The electrochemical pretreatment increased the cycle performance of the solid-solution cathode. However, this preparation is not appropriate for mass production due to its complexity and time consumption.

6.4 Conclusion:

New uses for lithium-ion batteries have been proposed, including power supplies for electric vehicles, hybrid electric vehicles, plug-in hybrid electric vehicles, and stationary power sources. To fulfill the needs of these applications, new lithium-ion battery technologies have been described. Some publications, however, appear to neglect the

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viability of suggested new technologies, particularly their usage and mass manufacturing feasibility.

7. RAW MATERIALS EXTRACTION & PRODUCTION

LIBs' reliance on finite resources, combined with dramatic growth in production (approximately doubling every 5 years) and uncertain future recycling practices has generated concern over material constraints. Explored the potential bottlenecks in critical material supplies for LIB manufacturing. The breakdown of material comprising batteries, from active material through individual cells, modules, and packs, is well documented in the literature; breakdowns of elements present in each type of cathode active material are shown in **Table 1**. Although the usage of crucial resources is sometimes regarded as a single concern, three distinct topics merit discussion. First, there is the question of resource availability in relation to use, and if expanding battery manufacture would deplete crucial material sources and/or raise costs. Second, there is a geopolitical risk associated with highly concentrated manufacturing, which can result in war, price volatility, and artificial shortages. We define this worry as supply chain risk and suggest that academics frequently confuse it with resource depletion. Traditional life cycle assessment methodologies are inadequate for capturing supply chain risk. Third, there are environmental and social implications connected with mining activities, which are well within the scope of LCA (Jason Porzio, 2021).

Table 1: Elements Mass Ratio Per Cathode Active Material

Elements	NMC-111 (% mass)	NMC-532 (% mass)	NMC-622 (% mass)	NMC-811 (% mass)	NCA(% mass)	LFP(% mass)	LMO(% mass)
Li	0.078	0.022	0.077	0.077	0.072	0.044	0.038
Ni	0.197	0.083	0.354	0.471	0.489	-	-
Mn	0.184	0.466	0.111	0.055	-	-	0.608
Co	0.198	0.334	0.119	0.059	0.092	-	-
Al	-	-	-	-	0.014	-	-
Fe	-	-	-	-	-	0.354	-
P	-	-	-	-	-	0.196	-
O	0.343	0.095	0.339	0.338	0.333	0.406	0.354

When we talk about the energy used to get raw materials, it usually involves using diesel for mining machines and transportation, electricity for running machines, and natural gas for heat during processing. The processing part, where they refine the materials, is usually where you see big differences in how much energy is needed.

This means that when we do Life Cycle Assessment (LCA) studies, we need to really look into where and how we get lithium (Li) from, including average, marginal, and incremental sources. It's also important to check different ways of mining and processing to understand their impacts better.

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7.1 Production of Battery:

Figure 3 depicts the five processes in the production process: manufacturing battery cells, assembly of battery modules, battery rack module, battery subpack, and ultimately creation of the battery pack.

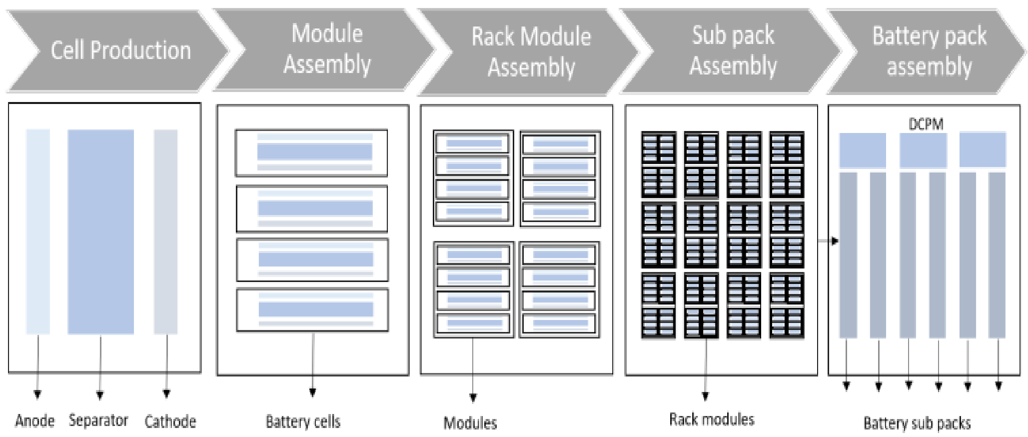

Figure 3: Production Stages from Battery Cell to Battery Pack

The creation of battery cells consists of three basic processes: electrode fabrication, cell assembling, and cell finishing. The cathode is made of an aluminum current collector that links the active materials. The most typical anode configuration is graphite coupled to a copper current collector. The electrolyte is typically composed of lithium combined with organic solvents. The separators, which are constructed of polyethylene or polypropylene, provide electrical isolation between the electrodes.

First, they mix the active materials for the anode and cathode to make a slurry. Then, they spread this slurry onto copper and aluminum sheets, which become the anode and cathode. After drying, they assemble these sheets into finished cathodes. Next, they roll up the anode and cathode with separator foils to make a jellyroll. This jellyroll goes into an aluminum housing, which is sealed shut with laser welding. Then, they fill the cells with electrolyte and seal them with a valve, also using laser welding. Finally, they go through a process of charging and discharging the cells to check their quality and performance.

7.2 Battery Pack Production

The battery pack include the mechanical structure, battery sub-packs and modules that holds the battery cells. **Figure 4** shows one battery pack including multiple strings of battery sub-packs. There are several passive components within the battery pack. The passive components include the mechanical structure, cooling system, internal conductors, wiring, exterior connectors, sensor boards, battery control unit, and a protective mechanical container. The battery subpacks are

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Figure 4: One Battery Pack with Multiple Sub-packs

inserted into a mechanical frame that is made of 500 kg steel. The mechanical frame also includes cooling manifolds manufactured by the ESS client and placed directly into the frame (Liu, 2020). When the cells arrive at the production site, they are first cleansed. After that, they are squeezed and layered together using glue. First, they use an adhesive made of polyurethane to both insulate and protect the battery from electrical and thermal issues. Depending on the adhesive used, they might need to remove any solvent vapors. The stacked cells are then pressed together and covered with plastic plates for extra protection. The battery module's enclosure, made up of side and bottom plates, is then placed on top and secured with more adhesive films.

Next, they connect the electrical components by laser welding tin-plated copper bus bars to the cells using high-powered laser welding machines. This process ensures a strong connection by melting the materials together. After welding, they conduct a thorough quality check, and if everything looks good, they attach the cell sensing board and connect it to a flexible printed circuit (FPC). The module then goes through various tests to check for any issues like external damage, software functionality, charge status, resistance, and gas leaks. If it passes all these tests, they seal the housing with a top cover using welding techniques and add labels to the finished product.

8. BATTERY END-OF-LIFE AND RECYCLING

8.1 Use Phase:

Many articles fail to incorporate the usage phase of a LIB when doing an LCA, citing the unpredictability and complexity of battery performance and lifespan. When comparing different battery technologies, it is critical to take into consideration differences in

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roundtrip efficiency and lives. Other features may be more or less important, depending on the application. For example, pack weight has an influence on vehicle economy in an electric car, truck, or aircraft, although weight is significantly less relevant in stationary applications. Cycle life is the number of charge/discharge cycles a battery can do under specific conditions before its storage capacity decreases to a predetermined level, which is generally 80% of its original capacity for EVs and 60% for stationary storage.

Table 2: Use-phase Requirements

Scenario	Applications	Installed Energy capa-city(MWh)	# cycles per day	Avg DOD (%)	Application service (MWh)
1	Electric time shift	81.6	1	90%	402,084
2	Renewable inte-gration	40.8	1	90%	201,042
3	Primary regulation	10.2	1	60%	33,507
4	Peak-shaving	40.8	0.5	90%	100,521

A battery's actual cycle life is influenced by its working circumstances, and when data is available, it should be modified depending on the predicted use case before determining lifetime energy throughput. Battery activities at high or low state of charge (SOC) contribute to faster battery aging. When comparing two theoretical EV batteries, one has a cycle life of 3000 cycles and a cycling frequency of 2 cycles per day, while the other has a cycle life of 3500 cycles and a cycling frequency of 1.6 cycles per day. As a result, there was a 5% difference in global warming potential between the two battery scenarios during their use. Although a more detailed examination of use-phase cycling and its influence on lifetime and efficiency would be ideal, credible data for LCAs is scarce.

8.2 End-Of-Life & Recycling Phase:

At the end of life, the batteries are recycled. The recycling process is separated into five steps: collection, discharge, dismantling, mechanical pre-treatment, and chemical treatment. The hydrometallurgical technique is used to treat the chemical.

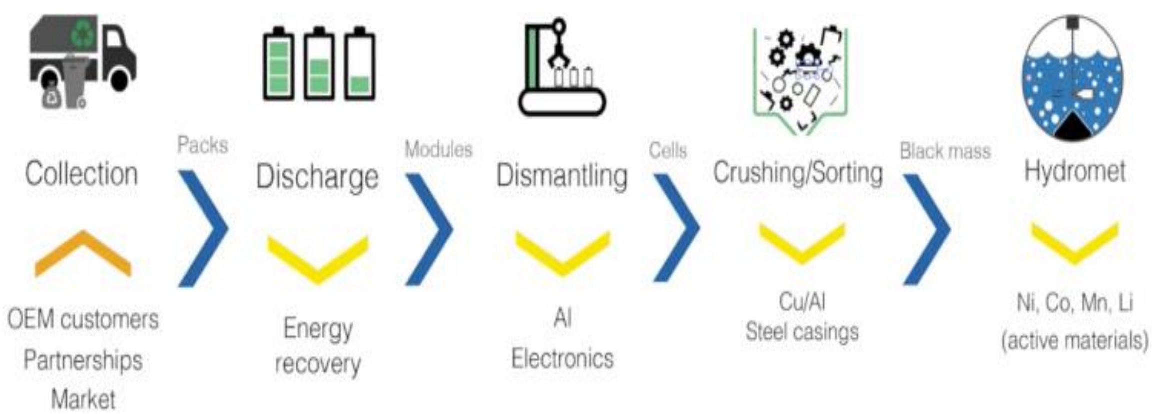

Figure 5: Recycling Process

Incorporating reuse and recycling into LCA has long been a methodological difficulty, presenting problems about how credits for recovered materials, as well as the avoided consequences of virgin material creation, should be distributed.

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Recycling is classified as closed-loop, which means that materials are recycled within the same production system (e.g., cathode materials recovered for use in new cathodes), or open-loop, which means that materials are recovered for use in different production systems. Most research on batteries use a closed-loop approach to recycling and investigate one or more of the three primary recycling approaches: pyrometallurgical, hydrometallurgical, and direct.

Pyrometallurgical recycling is a smelting technique that may recover transition metals such as Co, Ni, and Cu. It is used to recycle both LIBs and Ni Metal-Hydride (NiMH) batteries. Other materials, including as Li and Al, are oxidized throughout the process to create process heat but are seldom recovered.

Hydrometallurgical and direct recycling, which use leaching and physical separation techniques, respectively, recover a higher percentage of battery materials by mass. Both procedures are intended to recover the cathode (containing Li as well as metals such as Co or Mn), Al, and anode, however only direct recycling can recover the electrolyte (by cell flushing). With the exception of water usage, hydrometallurgical recycling saves more money on a variety of life-cycle inventory measures than pyrometallurgical recycling. Direct recycling is more difficult to compare since it is less widely employed and the process configuration and materials recovered differ.

However, as Gaines points out, there is more of a continuum than a clear separation between hydrometallurgical and direct recycling; as the Co content of LIBs decreases, a hybrid direct/hydrometallurgical strategy may be preferred over a pyrometallurgical process. Previous LCA studies on battery recycling have assumed that recovered materials are functionally comparable to fresh materials. This is understandable given the scarcity or absence of empirical facts to back up any alternative assumption. An additional problem is developing a clear business-as-usual scenario to provide as a baseline for comparison.

The number of stationary and EV LIBs nearing end-of-life remains modest, and recycling and disposal policies vary by nation. Globally, around 95% of LIBs are not recycled. As demand for energy storage in EV and stationary energy storage applications grows and batteries approach their end-of-life, more research will be required to track the date of these batteries and gain a better understanding of what processes are used and what materials are ultimately recovered.

8.3 Recycling Phase:

While thorough and high-quality inventory data for recycling procedures is already uncommon for lithium-ion batteries, information for future battery systems such as SIB is even more limited. The majority of existing research in this area uses data determined for processing a certain cell type (mainly NMC) and assumes that these inputs would remain constant regardless of the actual feed mix.

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This restricts their application to diverse cell chemistries, since the needed amount of chemicals and process inputs varies depending on the processed materials, even when the same process chain is utilized. Furthermore, hydrometallurgical recycling facilities currently achieve significant recovery efficiency for automotive-type LiNMC batteries, although this is not always the case with lower-value containing batteries such as SIB. In fact, even for current LiFP batteries, recycling is usually limited to recovering the aluminum, copper, and steel components obtained from mechanical recycling steps (crushing, shredding, and mechanical separation), while the active material fraction, the so-called black mass (which contains primarily lithium, carbon, iron, and phosphorous in the case of LiFP), is typically discarded rather than further processed. To evaluate the individual recycling performance of the considered SIB cells, a cell-specific recycling model is therefore required.

The recycling process model is based on prior work, which supplied inventory data for several recycling processes. Inventory data were presented in aggregated form, and consumables were simply scaled based on the mass of the supplied battery cells, resulting in significant simplicity. As a consequence, the procedure was discovered to boost loads by deeper hydrometallurgical processing of LiFP and SIB batteries.

The underlying model has been updated and integrated into the excel-calculation tool, with the amount of essentials estimated for the specific cell composition using stoichiometric calculations and additional information obtained from recycling patents and secondary publications. To recycle batteries effectively, the best method we have is a fancy process called hydrometallurgical treatment. Here's how it works: first, they crush and grind the battery cells to get the metal parts like housings and collectors.

They use machines to separate out plastic parts like the housing, seals, and separators, which are then thrown away as plastic waste. They also find a way to recycle the electrolyte during this process. The leftover black matter is then processed via a thorough hydrometallurgical recycling step, which recovers all important metals as well as the carbonaceous anode active material.

9. CRADLE-TO-GRAVE RESULTS

9.1 Results:

This section presents the impact categories that contribute more than 80% of the weighted aggregated single score. According to the Batteries PEFCR, these impact categories are the most relevant for future investigation. As a result, a more detailed breakdown of the contribution of each life-cycle stage, component, and material to the most significant impact categories will be provided. Finally, the sensitivity analysis findings are shown to demonstrate how crucial characteristics such as the carbon intensity in the power grid, roundtrip efficiency, and battery pack cycle life impact the results (Jiang, 2023).

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9.2 Cradle-To-Grave Results:

Table 3 shows the effect categories that contributed the most to the cradle-to-grave aggregated single score, and water scarcity was shown to be the most significant impact driver, accounting for 89-93% in each usage scenario. However, as shown later in the section, this is due to errors in the openLCA program. As a result, this outcome should not be considered genuine. As a result, three additional effect categories were examined to provide a more complete picture: acidification, climate change, and resource consumption (fossil). These three impact categories each contributed 2-4% of the overall aggregated score, accounting for the second biggest proportion and possibly the categories with the greatest impact if the water scarcity conclusion had not been wrong (Liu, 2020).

Table 3: Contribution of impact categories to the cradle-to-grave aggregated single score based on characterized, normalized and weighted values

Impact category	Time shift	Renewable Integration	Primary regulation	Peak shaving
Acidification	2%	2%	2%	2%
Climate change	2%	4%	2%	2%
Eutrophication marine	0%	0%	0%	0%
Eutrophication, freshwater	0%	0%	0%	0%
Eutrophication, terrestrial	0%	0%	0%	0%
Ionising radiation, human health	0%	1%	0%	0%
Land use	0%	0%	0%	0%
Ozone depletion	0%	0%	0%	0%
Particulate matter	1%	1%	1%	1%
Photochemical ozone formation-human health	0%	0%	0%	0%
Resource use, fossils	2%	3%	2%	2%
Resurce use, minerals & metals	1%	1%	1%	1%
Water Scarcity	91%	89%	93%	93%

9.2.1 Water Scarcity:

The battery pack's influence on water shortage contributed the most to the total aggregated impact. This was mostly owing to the employment of acids in the hydrometallurgical process, accounting for 99.5% of the impact. Thus, the EOL recycling process was the primary contributor to water shortage. The second highest contributor to the category was power losses during the consumption stage, accounting for just 0.31% of the total effect.

9.2.2 Climate Change:

Climate change was the second most significant effect driver to the aggregated single score, and the use-stage was determined to be the primary contributor within this area in all four usage scenarios. The usage stage accounted for 62-80%, as shown in **Figure 6**. Battery production was the second biggest life-cycle stage, accounting for 22-41% and 8-15% of overall climate change, respectively. The distribution and collecting stages made a relatively tiny contribution, accounting for less than 1% of the total impact. If all

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power provided throughout the usage stage is included, the contribution dominates climate change even more, accounting for approximately 98-99% of the entire effect.

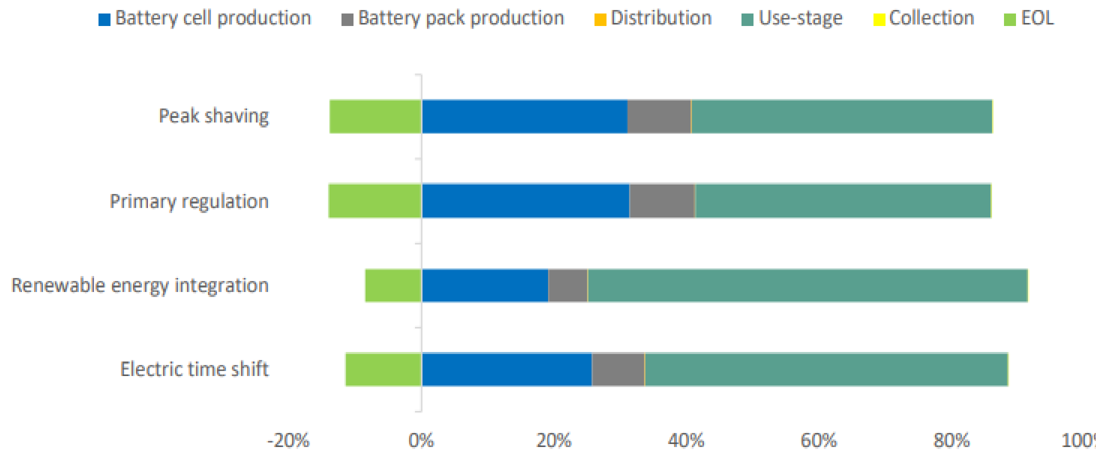

Figure 6: Life-cycle-stage Contribution to Total Cradle-To-Grave Climate Change

9.2.3 Acidification:

The largest contributor to the acidification impact was from the battery pack production stage, where the nickel used in the cathode manufacturing was the main driver of impact, as presented in **Figure 7.** Nickel consumption accounted for 73-81% of overall acidification impact across the four use scenarios. Recycling at the end of life gave between 24-26% credits and 3-4% loads, resulting in a net decrease in overall acidification impact of 20-23%.

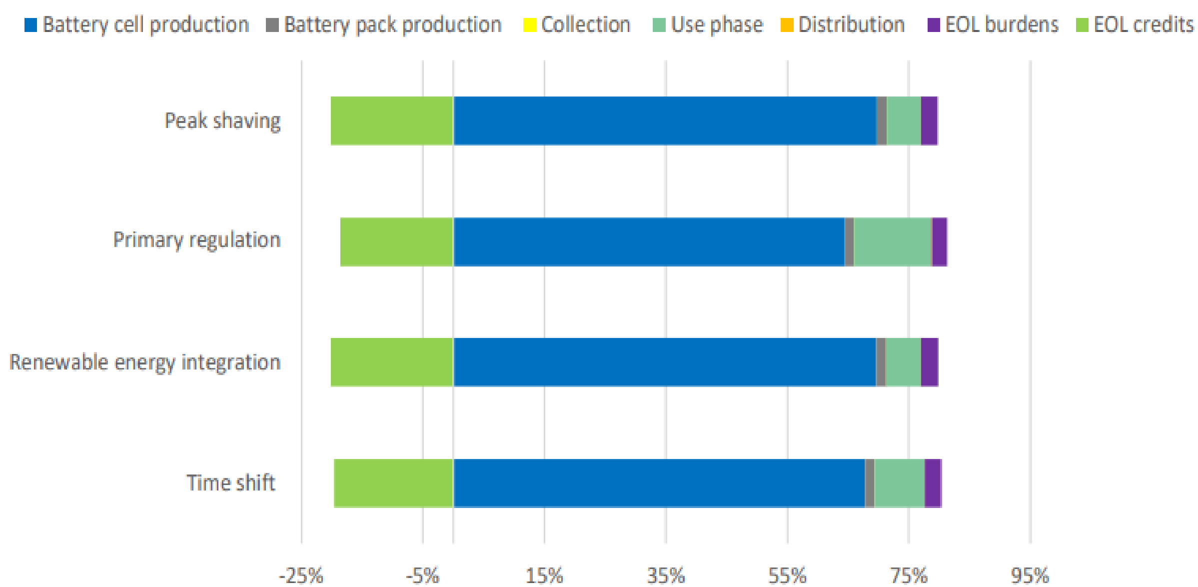

Figure 7: Life-cycle-stage Contribution to Acidification

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9.2.4 Fossil Resource Use:

Figure 8 depicts the usage-stage as the primary driver of effect in the category of fossil resource use. Battery pack manufacturing was the second highest effect driver, accounting for 16-30% of overall impact and 6-11% for battery cell production. The recycling procedure at EOL contributed 3-6% loads and 12-23% credits, resulting in a net effect decrease of 9 to 18%.

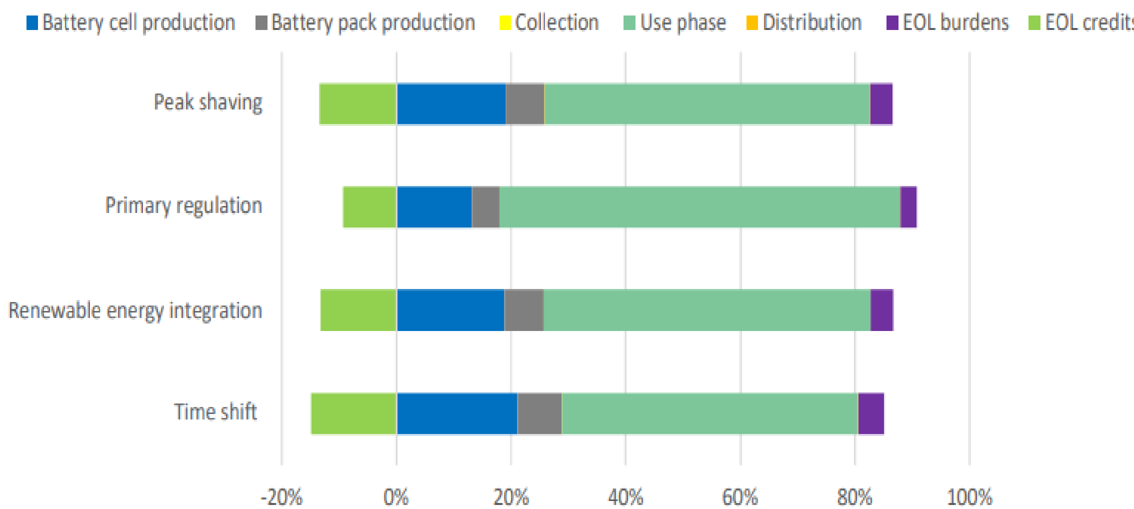

Figure 8: Life-cycle-stage Contribution to Fossil Resource Use

9.3 Comparision OF GWP B/W Al-ION & Li-ION:

The Li-ion recycling steps and their impact on the GWP indicator. Similar to the environmental profile achieved during the production phase, the Al-ion cell has a higher environmental performance per cell, although the Li-ion technology outperforms the innovative technology in terms of energy stored per Wh. The effect breakdown for the Al-ion cell indicates that the VS + LHTP step is the largest contributor to GHG emissions, accounting for 43% of total carbon emissions. Its considerable contribution can be due to the use of a vacuum chamber, which is extremely energy-intensive. The off-gas cleaning process is the second most major source, accounting for 34% of total carbon emissions. Its effects are attributed to upstream activities such as the creation of active carbon, which is utilized to filter organic molecules from the thermal process. Pyrometallurgical activities are the largest contributors to the Li-ion recycling process, accounting for 61% of total emissions due to high energy needs (Mario Amin Salgado Delgado, 2019).

The second most intense stage of this chemistry is the discharging stage, which accounts for 20% of total carbon emissions. Its carbon burdens are attributed to the huge volume of brine utilized in the process. Essentially, the unique recycling process devised by ACCUREC demonstrates that pyrometallurgical processes may be substituted for a vacuum shredding + LHT process, resulting in savings of roughly 6%.

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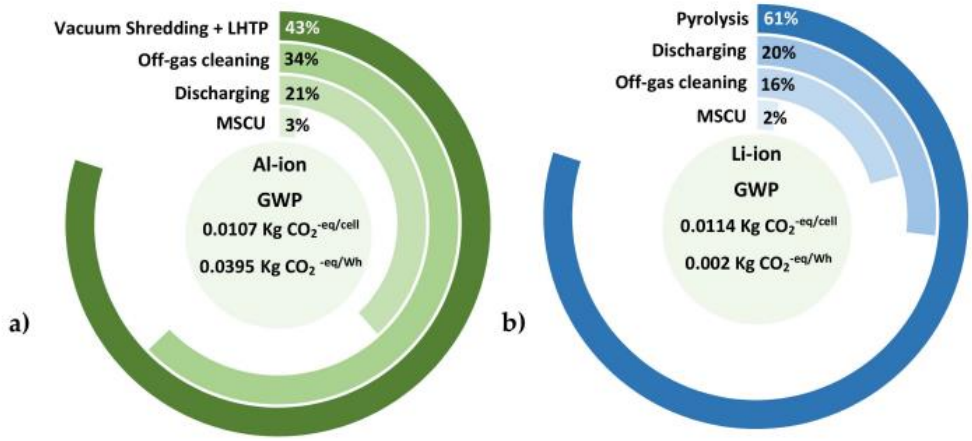

Figure 9: Comparison of the GWP Contribution by the Al-ion's (a) & Li-ion's (b) Recycling Processes

10. SENSITIVITY ANALYSIS

Previous research has shown power use in cell manufacture as a significant contributor to overall climate change. However, power utilized in cell manufacture accounted for just 2% of the study's cradle-to-gate climate change effect. This is primarily because the electricity mix utilized in production was based entirely on Swedish hydropower, a green energy source. This greatly lowered the environmental effect of power use. Therefore, a sensitivity analysis on how the five most relevant cradle-to-gate impact categories; climate change, resource use- fossil, resource use-minerals and metals, acidification and particulate matter are affected by the choice of electricity mix used in the cell production was conducted (Liu, 2020). The electricity mixes used in the sensitivity analysis was chosen with increasing carbon intensity: 100 % Swedish hydropower, Swedish residual electricity mix, EU consumer mix, US consumer mix and Chinese consumer mix, as presented in **Figure 10**.

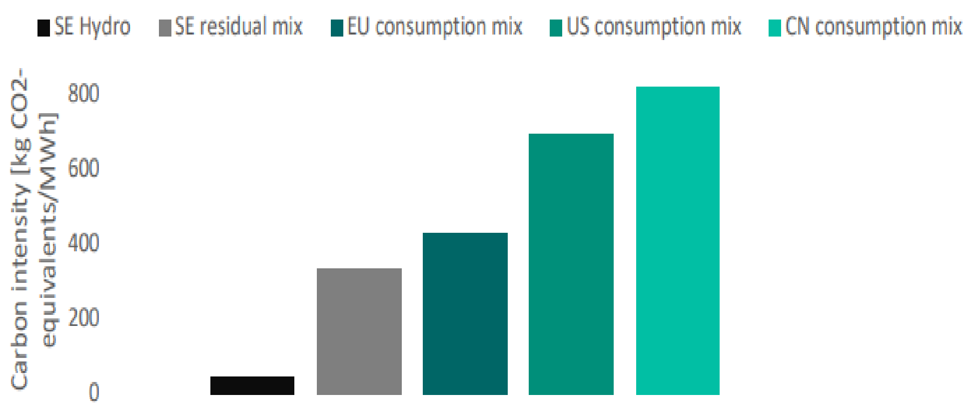

Figure 10: Carbon Intensity of Different Electricity Mixes used in the Sensitivity Analysis

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The results of the sensitivity analysis are presented in **Figure 11** and it showed that the cradle-to-gate climate change was highly dependent on the carbon intensity in the electricity mix. The climate change increased by 30 % when using the Swedish residual mix instead of Swedish hydropower. When employing the EU consumption mix, climate change grew by 73%, whereas the Chinese consumption mix climbed by 154%. Within fossil resource consumption, all other grid mixes increased by over 100% as compared to Swedish hydropower. However, resource utilization (minerals and metals), acidification, and particulate pollution showed only a slight rise.

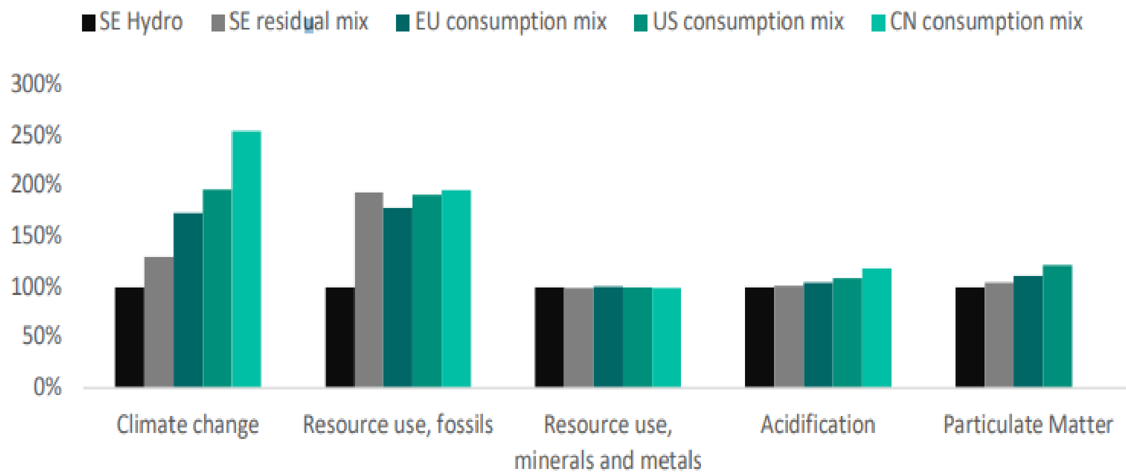

Figure 11: Sensitivity analysis for the carbon intensity in the electricity mix used in battery cell production impact on the characterised result of climate change, fossil resource use and water scarcity results

10.1 Energy Density:

Energy density is an essential quantity in environmental terms. A high energy density minimizes the quantity of battery required to provide a specific storage capacity, lowering the environmental effect. This explains the LiNMC cell's strong GWP findings as well as the NaPBA and NaNMC's relatively high effects in this area. It should be noted that these findings do not rely on individual re-dimensioning of the battery cell by modifying the electrochemical parameters of the active materials, but rather employ a simple linear scaling technique to demonstrate the impact of energy density. It so ignores electrochemical constraints, such as the fact that sodium has an inherently larger molar mass than lithium and a slightly lower potential (lower cell voltage), preventing them from reaching the same maximum energy densities as LiNMC.

The NaPBA would require a 33% increase (120 to 160 W h kg1) to match the LiNMC, but the NaMMO currently outperforms the LiNMC, even with a 10% lower energy density. However, the NaNMMT and NaNMC appear to be unable to outperform the LiNMC. Under HTP conditions, the SIB (with the exception of the NaNMC and NaMVP) are already located below the LIB at current energy densities. The two best-scoring SIBs (NaMMO and NaNMMT) would surpass the LiNMC in this area, although having energy densities that are approximately 25% lower. The NaNMC cell would need (like the LiFP)

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unreasonably high energy densities comparable to the LiNMC, whilst the NaMVP would not even come close to the benchmark.

10.2 Efficiency And Cycle Lifetime:

Depending on the power used for charging, the usage phase (and hence efficiency and longevity) can have a significant influence on the overall performance of the battery system. Both parameters are highly dependent on operating circumstances such as C-rate or temperature, but they can also vary greatly amongst cell manufacturers. In terms of cycle life, 4000 cycles have been taken as the baseline for all cell chemistries except LiFP and NaPBA, which have much longer cycle lifetimes (7000). As a result, despite the larger net impacts from manufacturing, the LiFP outperforms the others in three of the impact categories. Given the technical similarities and the existing high efficiency of contemporary LIBs, it is doubtful that significant improvements can be made to the SIB, making notable changes in battery rankings unlikely. However, efficiency is tied to the usage phase and hence heavily reliant on the source of the charged power.

10.3 Cradle-To-Grave Environmental Impact:

Furthermore, to assess how the cradle-to-grave environmental effect was altered by varied battery lifetimes, another sensitivity analysis was performed by altering the number of cycles delivered within a single battery lifespan (Mudit, 2021). In the electric time-shift scenario, the cycles ranged from an average of 4,000 to 1,500 and 6,500. This amounts to a 63% increase in cycle life. The findings are shown in **Figure 12**. The findings indicate that a 63% increase in cycle life resulted in a significant reduction of 34% in acidity and 29% in water shortage. However, it only lowered the impact of climate change by 14% and 13% for fossil resource consumption.

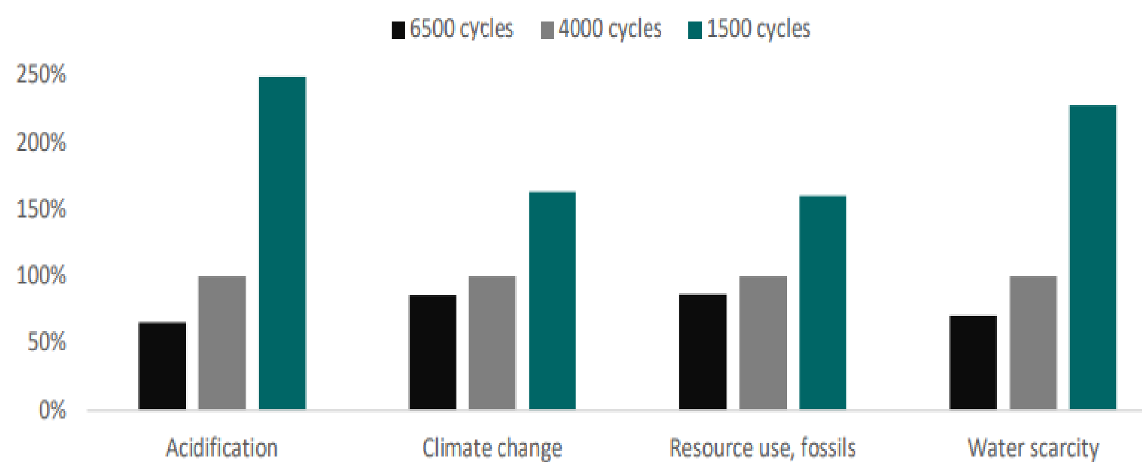

Figure 12: Sensitivity analysis on number of cycles provided during battery lifetime influence on characterised acidification, climate change, fossil resource use and water scarcity results

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Lastly, a sensitivity analysis of round-trip efficiency was performed to see how the cradle-to-grave environmental effect was influenced. The average roundtrip efficiency of 96% was reduced to 94% in a low efficiency scenario and 98% in a high efficiency case. The results are presented in **Figure 13**. The findings indicate that roundtrip efficiency has a significant impact on climate change and the usage of fossil resources. The increased round-trip efficiency resulted in a 51% decrease in climate change and a 49% reduction in fossil resource use. However, the effect on acidification was minimal, with just a 9% decrease. Water scarcity was alleviated by 19%.

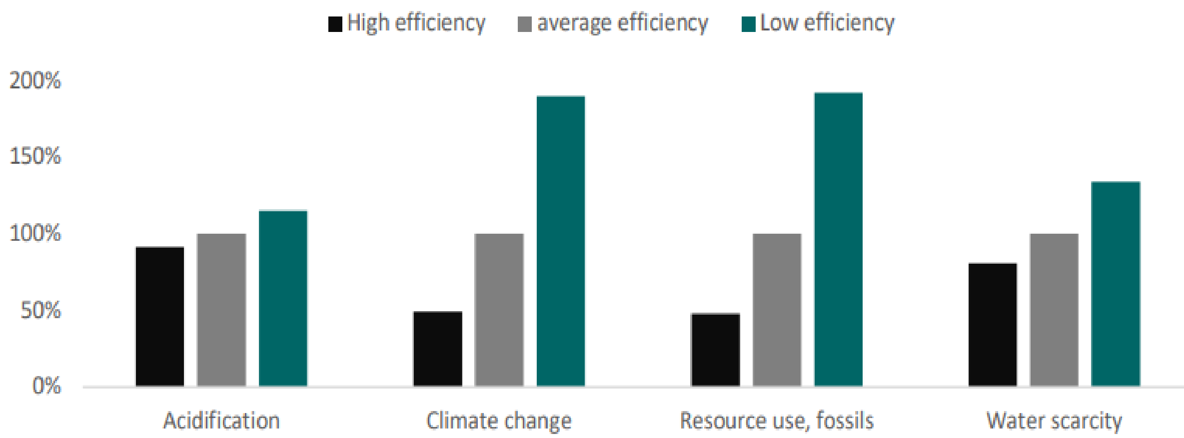

Figure 13: Sensitivity analysis on the round-trip efficiency of the battery pack influence on characterised acidification, climate change, fossil resource use and water scarcity results

11. RECOMMENDATIONS FOR FUTURE WORK

Although we are not the first to point out the problems in reaching agreement on techniques for performing battery LCAs, we think that this analysis provides the most thorough examination of the underlying causes of inconsistent battery LCA findings. Quantifying the environmental implications of battery manufacture may be quite difficult, and we urge that future research simplify and prioritize their efforts based on the processes and materials that make the most contribution. Disaggregating environmental impacts by location and type of operation can improve transparency and accuracy while also creating a framework for companies that carefully manage their supply chains to avoid such suppliers to be recognized in their estimated environmental footprints (Jason Porzio, 2021).

To improve the interpretability and effect of future battery LCAs, each research must include a sensitivity analysis at various manufacturing plant sizes. Our examination of the literature shows that this point, and the resultant variances in anticipated industrial energy usage, produce the most misunderstanding of any parameter. An LCA that uses market reports to estimate global-average energy usage for battery manufacture and, ideally, anticipates possible trends is also urgently required to demonstrate the gap between present literature and current/future industry practices.

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A final conclusion from this review is that a rigorous, complete cradle-to-grave LCA of multiple battery technologies can be made more tractable by the production of consensus-based scenarios to address some of the major sources of uncertainty for these analyses. Ambitious harmonization initiatives are not uncommon, and by collaborating with systems analysis and technology specialists, the community can ensure that future evaluations of battery technologies improve our understanding of their environmental consequences (Christian Aichberger, 2020).

11.1 Limitations and Future Work:

The present assessment relies on an improved cell-specific recycling model that estimates the input of process chemicals and the output of recycled materials in a cell-specific manner, significantly improving previous modelling approaches. However, even after being modified for each cell composition, it still assumes the same hydrometallurgical process route (which is primarily developed for LiNMC cells) for all cell chemistry. There are currently no documented alternate methods for processing black mass from, say, LiFP cells, but it is envisaged that these would necessitate quite different procedures that are better suited to them. This would also necessitate a reliable separation of cell chemistries prior to recycling.

Second, the results for the ADP category (resource depletion) should be interpreted with caution. When examining the contribution of particular constituent flows to total ADP, sulphur, arsenic, and copper are among the most significant. This may be consistent with the real availability of the respective elements in the earth's crust or given reserves, but it appears unusual when analyzing the impact of batteries, where cobalt, nickel, lithium, and copper are the most significant ingredients. This is a direct result of the ecoinvent database's modelling methodology, in which not only process inputs and emissions, but also elementary resource flows, are assigned to mining co-products using economic criteria rather than physical links.

Third, charge-discharge efficiency appears to be a significant characteristic that is not addressed by the cell dimensioning tool established in this study. Rather, round trip efficiency estimates are derived from the literature and may not correlate directly to the tested battery cells. Cells built for maximum energy density will increase electrode thickness (and active material) while decreasing current collector mass, which has the reverse impact on efficiency, resulting in a tradeoff between energy density and efficiency. A thorough study of these trade-offs, as well as an improved cell dimensioning tool that accounts for ohmic losses, would be quite beneficial in this area.

Finally, it should be mentioned that the recycling model presented in this study is based on process modeling, stoichiometric calculations, and a single set of past data from business visits. The advanced hydrometallurgical treatment is the sole model that is thought to be sufficiently trustworthy for assessment, whereas the current data for traditional pyrometallurgical and hydrometallurgical procedures are insufficient for meaningful evaluation. However, the recycling methods are primarily built for existing LiNMC vehicle batteries, whereas some of the evaluated cell chemistries will need

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custom-tailored techniques. More cell-specific recycling methods would be necessary to maximize the recovery potential of these new SIB cell chemistries. The process models for these processes are intended for future usage, but they need be improved with firsthand industry data before being implemented. This would greatly enhance the existing state of the art, as well as allow for comparisons of various recycling procedures.

12. CONCLUSION

To conclude, if excluding the water scarcity that was likely overestimated in this study due to software inconsistencies, the climate change, acidification and fossil resource use were the three impact categories with the most relevant impact to the cradle-to-grave result. Furthermore, the most efficient way to reduce the climate change and fossil resource use is to reduce the electricity losses coupled to the use-stage. This can be achieved by using renewable energy in the electricity grid or by increasing the round-trip efficiency of the battery system. However, to reduce the acidification impact, which is mainly caused by the battery production stage, it is more important to source nickel and cobalt from sustainable suppliers or to increase number of cycles provided by the battery system during one lifetime (Jens F. Peters, 2021). Therefore, it will be crucial for battery manufacturers to incorporate a sustainable energy sourcing strategy, continue investing in research and development and to integrate sustainable supply chains to reduce the overall environmental impact from the battery packs from a cradle-to-grave perspective. Second, in the cradle-to-gate assessment, five impact categories were identified as accounting for more than 80% of the overall aggregated single score effect: climate change, acidification, fossil resource usage, resource use (minerals and metals), and particle matter. Within these impact categories, four main materials accounted for more than 65% of the total impact.

These elements included nickel, aluminum, cobalt, and carbon black (which represented graphite). This conclusion emphasizes the necessity of integrating sustainable supply chains, particularly in relation to these four critical commodities. Third, in the cradle-to-gate evaluation, battery cell production was identified as the highest impact generator, independent of effect type. However, the battery pack components still had a major impact on climate change and fossil resource usage. As a result, battery pack manufacture has proven to be critical in undertaking LIB environmental studies. We believe that the Li-ion batteries can represent a key technology for the decarbonization of the energy system, in particular in the transport sector. Our review highlights five main aspects that will be crucial in the development of Li-ion batteries:

- The evolution and choice of future battery chemistries.
- Potential concerns for raw materials availability.
- The importance of battery end-of-life and recycling processes.
- The need of more detailed data on environmental impacts.
- The potential shift of the energy geopolitical equilibrium.

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This suggests that recovering these elements using the proposed process technology may not be environmentally preferable to mining them from virgin sources. However, because the recycling model used is based on a high-end process built for LiNMC cells, it may not be suitable for processing these low-value chemicals. The worldwide expansion of the lithium battery supply chain makes it harder to measure the production chain's environmental effect. Furthermore, there are limited data points accessible from production locations. The differences in manufacturing sites, chemical processes, energy mix, and cell chemistries and layouts result in a broad range of study outcomes. However, our review demonstrates how, when compared to older articles, more recent studies show a decrease in GHG emissions associated with battery pack production: this is primarily due to a better understanding of the characteristics of commercial-scale factories and an improvement in production performance.

Finally, including battery pack recycling at the end-of-life stage resulted in a net decrease of 9-20% in cradle-to-grave climate change, acidification, and fossil resource usage. This figure is expected to be substantially higher in the future if measured data from recycling plants is used instead of laboratory-scale data, as in this study. The findings revealed that Northolt's approach of increasing green energy source, vertical integration of the supply chain, and including higher recycling rates at EOL is a step in the right way for supplying sustainable LIB for ESS. Additionally, the values provided in this study is based on estimated production data, which are not yet measured and validated. Hence, the result provided is a forecast of the environmental impact from the battery packs life cycle.

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