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Life Cycle Assessment of Recycling Lithium-Ion Cells

Master's thesis in Circular Economy
Supervisor: Anders Hammer Strømman
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Abstract

The continuous rise in global surface temperature has motivated industries and nations to take measures for carbon neutrality and net zero greenhouse gas emissions. A big chunk of these emissions comes from burning fossil fuels in the pursuit of energy and mobility. Transformation of transportation from fossil-based Internal Combustion Engine Vehicles (ICEVs) to Electric Vehicles (EVs) is crucial to achieving carbon neutrality. However, EV production, especially battery manufacturing, is environmentally intensive compared to ICEV production. Besides end-of-life cells, a considerable proportion of secondary supply from the Quality Control (QC) reject cells is also expected, as the giga-cell production setups commence operation. Therefore, understanding the environmental performance of battery recycling technologies is essential, reducing, simultaneously need for new materials and the environmental impact of virgin resource mining.

This study has combined the conventional production and recycling processes and performed a Life Cycle Impact Assessment (LCIA) of producing 1kg of NMC111 lithium-ion cell under varying proportions of virgin and recycled content. The production and recycling models are combined by using a mass allocation-based output from the recycling model by attributing the input requirements of every recycling process step with respect to the amount of recovered cathode active materials. Thoroughly using the data points from the relevant studies, Life Cycle Inventory (LCI) is compiled, and the combined model is tested individually for direct, hydrometallurgy, and pyrometallurgy recycling technologies

The results demonstrated that the impact followed the condition $d_{direct} < d_{hydro} < d_{pyro} < d_{virgin}$. Among all recycling technologies, pyrometallurgy appeared to be the most impact intensive due to high resource and energy consumption and low recovery rates of the constituents of positive electrode paste. Another interesting finding is that the impact is correlated highly with the carbon intensity of the electricity mix indicating recycling while being worthwhile, yet not the sole solution towards carbon neutrality. Also, to realize the benefits of recycling, industrial as well as government policies are instrumental.

Keywords

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1 Introduction

1.1 Motivation

The global surface temperature has risen between 2011-2020 compared to 1850-1900, bringing about climate change (*IPCC, 2023*). Climate change is defined as long-term changes in weather and temperature patterns. These changes and their ongoing effects are brought on by unsustainable use of energy, land, changes in land use, production and consumption patterns, and lifestyles regionally as well as globally. Due to these changes, several extreme weather and climatic effects are already being impacted on every continent having broad negative effects on both humans and ecosystems. These effects range from low life expectancy, poor health, food security and nutrition, water scarcity, and reduced economic and social development.

The pursuit of energy is one of the major causes of these effects. Since the 1800s, humans started burning fuels for cooking, transport, operating machinery, and equipment, and producing electricity (*IPCC, 2023*). Fossil fuels are non-renewable resources that originated from the remains of animals and plants from the prehistoric era. These organic materials underwent a natural process of decomposition and were subsequently buried under layers of rock and sand over millions of years. The immense heat and pressure during this extended period transformed these organic remains into coal, oil, and gas, earning them the label of fossil fuels. The combustion of these fossil fuels produces greenhouse gases like carbon dioxide, methane, and water vapor that trap incoming solar energy but emit lesser. This net gain of energy is called radiative forcing which in turn warms the earth's surface and increases the global temperature. The increase in global temperature severely affects the economic, ecological, and social systems. With the glaciers melting and the sea level rising, the global temperature has already surpassed 1.1°C over preindustrial levels (*WMO Update*, 2022). Consequently, millions of people suffer from poverty and hunger due to climate effects such as floods and droughts. Not only these direct effects but also indirect effects of losing access to essential services and resources, like education and health. Furthermore, it stifles economic growth thus widening inequality and social injustice. About 700 million people are projected to be in danger of being displaced by droughts by 2030 (WHO, 2018).

Therefore, to prevent the catastrophic effects of climate change and to save lives, livelihoods, and nature, it is crucial to act timely. For this cause Paris Agreement sets out a global framework to bring nations together in combating climate change and/or adapting to its effects (*The Paris Agreement | UNFCCC*, 2015). This agreement not only aims to limit the global temperature rise to well below 2°C but also motivates the global partners to limit the global temperature rise to even below 1.5°C. Under this framework, every country outlines its ambitions to lower GHG emissions by committing to Nationally Determined Contributions (NDCs), its roadmap toward capacity building, and climate action.

To achieve the goals of the Paris Agreement, it is necessary to understand the contributions of the key areas that drive climate change in order to establish an appropriate action plan. According to the United Nations Climate Report (UN, 2023), more than 75% of all greenhouse gas emissions are due to fossil fuels making them a dominant contributor to climate change. Carbon dioxide and methane are the most common greenhouse gases produced widely while burning fossil fuels (US EPA, 2015). These emissions are dominant in generating power and transport operations. Only 29% of the world's total electricity production is fueled by renewable energy sources (*IEA, 2021*) which create very little to no greenhouse gases or other air pollutants compared to fossil fuels generated electricity (*UNECE*, 2021). Moreover, vehicles, ships, and aircraft are also powered by fossil fuels. Due to the internal combustion and diesel engines used in vehicles, which burn petroleum and diesel as fuels, most of the carbon

dioxide emissions are generated from transportation (Elgowainy et al., 2018). In addition, statistics suggest that over the next few years, energy use for transportation will rise significantly (*IEA, 2022*) to about 9300 GWh by 2030 (Yuan et al., 2021).

Several businesses and governmental organizations are pledging to net zero emissions in the next decades. The expected rise in the transportation market (*Precedence Research, 2023*) raises the question of how to develop transportation sustainably while meeting the needs of society and the environment. Electrification of transportation is a crucial for reaching carbon neutrality in the transportation sector (H. Li et al., 2023). Electric Vehicles (EVs) can help in resolving environmental as well as energy-related problems. The automotive industry has already acknowledged EVs as the future of the automobile industry (*McKinsey*, 2021). The transformation of the conventional ICEVs to EVs is accelerating. In order to transit seamlessly initiatives under raw material extraction, vehicle manufacturing, charging infrastructure, political and social acceptance, and education are required to maximize the benefits, and reduce the cost, social and technological barriers (Boulanger et al., 2011).

EVs can offset a huge environmental impact during the operation phase if clean energy is used for charging the vehicle. However, the production of an EV is quite environmentally intensive compared to the production of a conventional vehicle (Ellingsen et al., 2016). Since the production of batteries for EVs emits more carbon dioxide than Internal Combustion Engine Vehicles (ICEVs) (Y. Li et al., 2019). According to Sen et al., (2019), more than half of the total material carbon footprint and 65% of the direct impacts within in geographical limits of the United States are due to battery manufacturing. Since the battery is an integral part of EVs, to ensure that the production of EVs is sustainable, it is necessary to review battery production processes and analyze the potential to reduce its carbon footprint for sustainability. Out of all available battery technologies, lithium-ion batteries are preferred by EV makers due to their high energy density, prolonged operational lifespan, elevated working voltage, minimal self-discharge, and negligible memory effect (Lai et al., 2021; Lai, Huang, et al., 2022). However, with the increased use of lithium ion batteries, the concerns remain for safe operation, as overcharged cell raises the risk of explosion, undercharged cells reduces the useful life of the battery (Hannan et al., 2017), and reduction of negative environmental impacts in their production processes (Hannan et al., 2018).

Production of a Lithium-Ion Battery (LIB) for an EV requires significant amount of materials like aluminum, copper, cobalt, and lithium (Porzio & Scown, 2021). Olivetti et al., (2017) circumspect that the rapid adoption of EVs could exceed the demand for battery-grade materials like cobalt and lithium soon. Furthermore, they observed that the extraction ratio for cobalt, lithium, and graphite reserves has increased which may lead to finite resource depletion if continued to be extracted at similar rates. Moreover, the processing and refining of these materials are typically more energy intensive. Additionally, 60% of the total cost of battery production is due to material and the cathode contributes to 35% of the total material cost (X. Zeng et al., 2019).

Therefore, the high demand for EVs calls for exploring and researching recycling technologies of battery-active materials. Li-ion batteries must be recycled to maintain economic and environmental sustainability. This must be done to manage end-of-life-cycle trash creation due to its widespread use and ongoing growth in production. A closed-loop recycling system should ideally enable the use of recycled materials in the manufacture of new batteries (Georgi-Maschler et al., 2012). There is a need for recycling strategies to recover valuable contents and close the loop back to the battery production system. Recycling can also be a crucial long-term secondary material supply source in the coming decades (Olivetti et al., 2017) as it reduces the need for new virgin materials, so does the impact of mining and resource extraction for the valuable and scarce battery-grade elements (Nordelöf et al.,

2014). However, recycling operations are also associated with impacts of energy and additional resources to retrieve essential material that may outweigh the benefits (Oliveira et al., 2015).

Hence it is essential to understand the environmental performance of various battery recycling technologies in practice and under study. And the anticipated environmental impact generated due to the implementation of these technologies while producing cells for future use.

1.2 State-of-the-Art

1.2.1 Environmental Impact of Lithium-ion Batteries Production

A number of studies are available for investigating the environmental impact of the production of lithium-ion batteries. Ellingsen et al., (2014); Hao et al., (2017); Kim et al., (2016); Lai et al., (2022); Majeau-Bettez et al., (2011) and Notter et al., (2010) investigated greenhouse gas emissions from the production of lithium-ion batteries. Ellingsen et al., (2014) sought that a significant contribution of the total impact is due to energy use in the production process and material and energy use in the preparation of positive electrode (cathode) paste and negative current collector (usually made of copper). Moreover, the study concluded that cleaner and green energy sources for the production of battery cells will potentially reduce the overall production impact. Hao et al., (2017) compared the carbon footprint of the production of three types of lithium-ion cells chemistries- LFP (Lithium Iron Phosphate), NMC (Nickel Manganese Cobalt), and LMO (Lithium Nickel Manganese Oxide)- in China. The study found that the highest production impact of LFP batteries which can increase the manufacturing impact of an EV by 30%. Also, the impact due to the Chinese electricity mix is observed to be three times higher compared to the USA due to relatively cleaner energy. Kim et al., (2016) found 45% of the total impact in battery manufacturing is due to cell production, and 40% of the cell mass is comprised of electrodes and current collectors. While Lai et al., (2022) compared 4 different NMC cell chemistries with NCA (Lithium Nickel Cobalt Aluminum oxides) and LFP and found cathode have more than 30% of the impact contribution in almost all environmental impact categories and about 90% in Acidification Potential (AP) and Fine Particulate Matter Formation (FPMF). Majeau-Bettez et al., (2011) found with the help of Structural Path Analysis (SPA) that metallurgical activities associated with the extraction of nickel, for producing positive electrodes and metals for current collectors, contribute to more than 80% of Metal Depletion, Terrestrial Acidification and Particulate Matter Formation impacts and more than 70% of the toxicity impacts. The study by Notter et al., (2010) also represented the major contributors to environmental impact are metals and energy use in the development of anode, cathode, and battery pack. These studies pointed out that the major contribution of the critical battery components i.e. electrodes and specifically the positive electrode that is used in cathode manufacturing.

However, there are also certain innovations in improving cell chemistry while reducing the impact of the extraction and processing of battery-grade materials. Replacing cobalt with nickel and manganese can potentially reduce the cost and environmental impact of cell production (Scrosati, 2000). Electricity consumption and blasting operation during mining and extraction of cobalt emits radioactive emissions and harmful particles which are diffused by air all over the environment (Farjana et al., 2019). Despite environmental concerns cobalt mining is also associated with political instabilities due to large reserves presence in high-risk areas of the Democratic Republic of the Congo in Africa, threatening future investments for battery industries and relevant other applications (Kapusta, 2006) and social injustice due unsafe mining practices and child labor (*OEKO, 2011*). Therefore, battery chemistries are now evolving to include low-cost and more environmentally friendly material such as lithium manganese oxides (Robertson et al., 2001).

1.2.2   Cost of Lithium-ion Batteries Production

The relatively high cost of manufacturing lithium-ion batteries compared to lead acid and NiMH batteries is a major obstacle to market penetration (Diouf & Pode, 2015). Manufacturing cost includes both the process and material cost. Coating, drying, formation, and aging make up about 48% of the total cell manufacturing cost while, drying and solvent recovery contribute 47% and the dry room contributes 29% of the total cell manufacturing energy due to high moisture levels of added solvent (Liu et al., 2021) making the process both economically and environmentally unsustainable. Major cell materials including cathode, anode, electrolyte, separator, and cell container, make up about 75% of the total cell cost, and 50% of this material cost is due to cathode (Gaines & Cuenca, 2000).

Certain innovations to reduce the coating and drying energy requirement have been done such as using aqueous binder in coating instead of organic ones (Jansen et al., 2016) and increasing solid content and slurry viscosity with the help of high-pressure extrusion (Haarmann et al., 2020) can enhance the drying efficiency. Also, an approach developed by Maxwell Technologies Inc. to manufacture dry-coated electrodes by removing the use of any solvents thus eliminating the requirement for drying. Furthermore splitting the drying process into three stages can also shorten the drying time (Jaiser et al., 2017). Drying technologies like fiber laser can reduce energy consumption by 50% (Liu et al., 2021). These innovations saved energy costs and hence the carbon footprint. Moreover, to improve the energy performance of the dry room, Huttner et al., (2020) observed short-term (20 min) low-temperature (20°C) argon drying has better electrochemistry performance than conventional vacuum drying. In order to accelerate the formation process to less than half compared to traditional one, Lee et al., (2004) proposed optimizing cut-off voltage, removing the intercalation phase and reducing formation time in lithium carbon cells.

According to Gaines & Cuenca, (2000) using nickel instead of cobalt can increase the energy storage of cell and ultimately reduces the number of cells required in battery packs. Another way to decrease the cost mentioned in the study to replace cobalt is with manganese which is even more economical than a nickel but has less energy storage capacity. Broussely et al., (1997) observed that both nickel and manganese compounds with lithium can be achieved with the same cost by doubling the quantity for manganese-based compounds. Another study by Wood et al., (2015) finds that improving processes using a more economical aqueous binder and solvents, optimizing parameters in electrode manufacturing processes, using thicker electrodes (Patry et al., 2015) and reducing current collector mass can achieve more than 90% cost reduction in electrode processing as represented in Figure 1, where reduced cost is realized by aqueous binders, doubling electrode coats, and shortening wetting and formation times (Wood et al., 2015). Furthermore, studies are available like Berckmans et al., (2017), which predict a considerable decline in the future cost of batteries considering the potential expected changes in the chemical composition of a battery and the evolution of the market with the start of mass production.
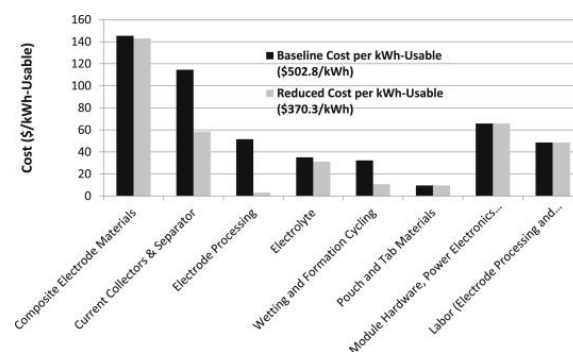

Figure 1- Baseline vs Reduced Pack Costs

1.2.3   Problem Statement

The widespread use of lithium-ion batteries in portable electronics and upcoming use in electric vehicles has the potential to create a massive number of used batteries, which may end up being discarded. Despite being valuable, less than 5% of the lithium ion batteries produced are currently recycled due to various factors (CAS Science Team, 2022). This impending situation has prompted researchers to seek out sustainable and cost-effective solutions to deal with the large stockpile of lithium-ion batteries that will accumulate in the future. Also, excessive demand stimulates high production volumes and eventually high waste or discarded cells during production rejects. While there are safety risks and environmental concerns associated with handling and transporting lithium-ion batteries, the motivation for establishing proper methods of battery recycling goes beyond just mitigating the potential harm of chemicals. It is driven by the goal of conserving resources and reducing the reliance on virgin materials in battery manufacturing.

Extensive research and studies have been conducted to understand the evolution and maturity of lithium-ion battery technology. However, despite these efforts, there are still gaps and limitations that need to be addressed, especially considering the widespread diffusion and dominant role this technology is expected to play in the near future. A number of the available literature is lab scale analysis whereas there is limited information about industrial scale studies which makes analysis uncertain and addressing problems difficult (Keppeler et al., 2021). Confidentiality and transparency of actual life cycle inventory data by manufacturers is also a gap in present research work increasing uncertainty and limiting the quality of results (Pellow et al., 2020). High dependence on secondary data from research labs and limited access to primary data from industrial sectors also causes variation in the energy requirements of recycling processes. (Pellow et al., 2020). Most of the research and studies are focused on isolated production and recycling assessments for lithium-ion batteries. There are several studies integrating impact of production and credit for recycling processes (Dunn et al., 2016; Gaines et al., 2011; Mohr et al., 2020; Raugei & Winfield, 2019; Lai, Chen, et al., 2022; Feng et al., 2022) and even fewer where production with recycled materials is discussed (S. Jiang et al., 2022; Q. Chen et al., 2022; Ciez & Whitacre, 2019; Du et al., 2022; Yoo et al., 2023).

S. Jiang et al., (2022) compared hydrometallurgical and direct recycling of Li-ion batteries in China through a process-based LCA. Direct recycling was found to be advantageous due to its lower energy and chemical requirements. In addition, the study suggests that material recovery and carbon-free energy are integral to realizing the recycling benefits. Q. Chen et al., (2022) also found direct and hydrometallurgy as carbon-efficient recycling methods reducing emissions by 51.8% compared to raw materials. The study emphasized the significance of recycling in the short term and green transition in the long term for sustainable EV production. Ciez & Whitacre, (2019) used life cycle analysis and cost models to compare the three recycling technologies- direct, hydrometallurgy, and pyrometallurgy. The study also indicated better environmental and economic performance of direct recycling. Moreover, policies are found to be crucial to incentivize battery collection and recycling. Du et al., (2022) compared the production of virgin and recycled NMC materials and highlighted the major impacts contribution of the leaching and extraction process. The results showed 74% less energy consumption and up to 57% less environmental impact of recycling compared to virgin materials, therefore, emphasizing the significance of high recycling rates and cleaner technologies for battery and cell production industries. Yoo et al., (2023) explored the environmental performance of battery metal recycling (BMR) technologies namely pyrometallurgical and hydrometallurgical recycling to recover lithium and found a significant reduction in impact compared to the virgin resources.

The recovery rate from Quality Control (QC) rejected cell is higher than the post-consumer scrap collected after the end of life for recycling purposes (Marshall et al., 2020) which makes QC rejects a

strong candidate for recovery of valuable materials from the cell. Around 40% of the cost per kWh of a cell is due to material, 50% of this material cost comprises cell container and terminals, and 20% of cathode precursors (Ciez & Whitacre, 2017). However, there is more potential to reduce the cost of cathode precursor material than cell container and terminal as this hardware material are being in mass cell production for decades and research and optimization techniques have already been applied for cost reduction in these constituents. Active cathode material, as per the EverBatt model, is composed of about 38.8 wt.% for NMC111 total cell material, and battery production setups are interested in recovering these materials for reducing production cost and the associated environmental impacts from mining and extraction of virgin material. Dunn et al., (2016) illustrate the price comparison of cathode materials compared to other components of the cell and found that the recovery of cathode materials could also provide a significant economic advantage. Furthermore, most of the experts interviewed by Few et al., (2018) acknowledged the importance of research regarding the environmental consequences of producing and disposing of LiBs.

The environmental impact of the production of a cell from recycled material is only analyzed by S. Jiang et al., (2022) and Q. Chen et al., (2022). Hence, this study will be an increment toward the already established models and research. The results will be compared with previous established studies and their similarities and differences will be investigated with quantitative and qualitative analysis.

1.2.3.1   *Aim*

The aim of this study is to calculate the overall environmental impact of recovering the cathode active materials from the postproduction scrap cells by combining production and various recycling procedures under various reuse rate scenarios.

1.2.3.2   *Assumptions*

This study examines a hypothetical future scenario in which the recycling facility is integrated into the production plants themselves. In this envisioned setup, the scrap cells generated after end-of-life testing or final QC check are directly transported to the recycling facility. Notably, the study does not take into account any potential impacts arising from logistics and material handling systems involved in the transfer of the scrap cells from the production plants to the recycling facility. Instead, it focuses on an ideal circular economy strategy, aiming to effectively close the loop of battery materials.

1.2.3.3   *Research questions*

With respect to the goals discussed above, this study will be focusing on the following research questions:

1. How to combine and model the production and recycling process of lithium-ion batteries?
2. What will be the combined environmental impact from all recycling models under varying scenarios of recycled and virgin material use?
3. What will be the impact and how sensitive it is to change the geographical input of electricity, heat, and transport supply inputs?

1.2.4   Scope of Study

Due to unavailability of Life Cycle Inventory (LCI) data for in-production scrap from individual production processes, only the scrap generated during the final quality control check, i.e., the last step before final packaging, is considered. The cell is completely assembled at this stage and contains all the necessary materials. Therefore, the environmental impact of these three types of recycling technologies- Direct, Hydrometallurgy, and Pyrometallurgy are calculated and discussed.

This study focuses on the recycling of scrap NMC111 cells from production quality control rejects. In NMC111, nickel, manganese, and cobalt compounds are present in a molar ratio of 1:1:1 or 33.33%

each. These cells were selected as they are one of the most prevalent types of lithium-ion cells used in the EV market, making them a prime target for recycling in the transportation sector. Additionally, NMC111 batteries are the primary focus of most LCA studies related to LIB recycling (Abdelbaky et al., 2021). Furthermore, NMC batteries contain a higher amount of rare and valuable materials than other LIBs on the market, making them more attractive targets for recycling.

Moreover, the impacts from the combined production and recycling model are calculated individually using the energy and transport supply of Norway, Germany, and China. Changing energy and transport inputs to different geographical locations can be used to analyze the sensitivity of the system toward these factors.

The study is based on LCIs of production data points defined by Jinasena et al., (2021), Orangi & Strømman, (2022), and Ellingsen et al., (2014) and the recycling model defined by Dorri, (2022) for hydrometallurgy and pyrometallurgy and direct recycling by Jiang et al., (2022) as represented in Figure 2. These datasets are selected as these are taken from the latest publications with LCI for NMC111 cells designed for similar functional units.

Jinasena et al., (2021) provide a detailed breakdown of electrical and heat energy for individual processes for 1Wh of cell produced. This data is scaled for 224Wh/kg cell energy density manufactured in a theoretical output of 6GWh production capacity. Orangi & Strømman, (2022) has also utilized data and insights from Jinasena et al., (2021) and developed a process-based cost model to investigate the production cost of 10 different battery cell chemistries including NMC111 cell. However, the article discusses the model for prismatic cells, it has been adjusted and proportioned for an NMC111 pouch cell mass (0.93kg/cell) and dimensions ($15.7cm \times 1.05cm \times 25.4cm$). The adjusted model calculated that the cathode and anode components make up 0.507 kg per cell, where both the cathode and anode have a mass fraction of 91:4:5 for active material, carbon, and binder. The required electrolyte volume per cell is 0.051 kg, and it is assumed that 48% of the mass of the cathode and anode solvent is dried and evaporated. Production LCI of 1 battery vehicle pack by Ellingsen et al., (2014) has also been adjusted as per the production model for 1kg NMC111 cell and data for the additional components like a separator, electrolyte, cell container, transport, and infrastructure is taken from this source where necessary.

Dorri, (2022) compiled a comprehensive LCI consisting of materials and energy used in the two recycling models- pyrometallurgical and hydrometallurgical recycling. The first recycling model for lithium-ion batteries comprises a pyrometallurgical process and a supplementary hydrometallurgical process to extract the intended materials. The pyrometallurgical process is based on the Umicore process patented by Cheret & Santen, (2007), while the supplementary hydrometallurgical process is based on the process suggested by Dunn et al., (2012). The efficiency of the pyrometallurgical part was assumed to be 94% and 99% for cobalt and nickel, respectively, while the hydrometallurgical part was assumed to be 100%. The final products of LIBs' recycling processes are not pure elements and wastewater, and CO_2 are other outputs of the system. No pre-treatment was assumed for the process, and both processes are assumed to be happening at the same location.

The second recycling model, for hydrometallurgy, is based on a study conducted by Du et al., (2022) on a leading lithium-ion battery recycling plant in Jiangxi province, China. The study provided reliable data from a real industrial case in China, which is a pioneer country in utilizing hydrometallurgical processes for recycling lithium-ion batteries. The presented LCI in this study is found to be more comprehensive than other similar studies, reducing uncertainties in the LCA. Further Dorri, (2022) also compiled a process flow diagram, identifying the relevant chemicals used and stressors emitted. The study assumed that the main products of the hydrometallurgical recycling process for spent NMC111

cells were cobalt, nickel, lithium, and manganese, with a 100% recovery rate, based on a literature review by Larouche et al., (2020). Stressors and wastewater are also outputs of the system, with stressors determined based on Du et al., (2022) and all other materials assumed to end up in wastewater. Further, the electricity requirement was calculated based on the electricity consumption model by Rinne et al., (2021).
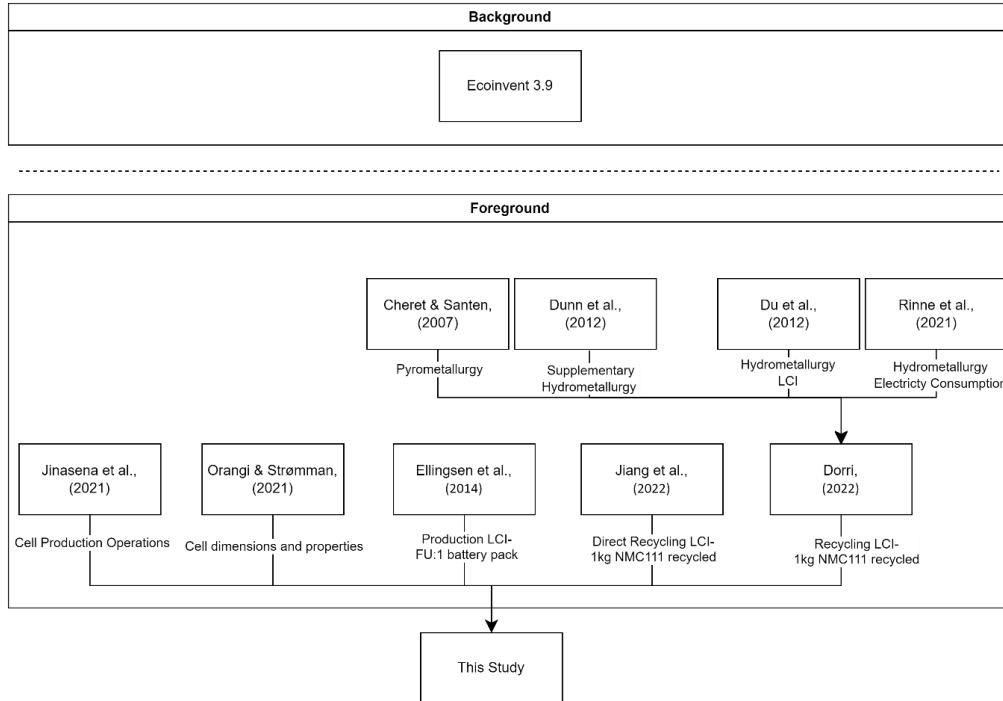
Figure 2- General flow representing the LCI data points taken from past studies.

2   Overview of Production and Recycling

2.1   Production Process of Lithium-ion Batteries

To identify the potential for improvement in future battery production, it is essential to understand and analyze the current infrastructure of production, material and energy requirements, and yield efficiency from the production of lithium-ion cells. A cell is a part of the module, and the module is a part of a pack that is integrated into an electric vehicle as shown in Figure 3. According to the Journal of Industrial Ecology by Ellingsen et al., (2014), a battery pack consists of 12 modules having 30 cells each. A full battery pack in a vehicle traction battery consists of four major components: Cell, Battery Management System (BMS), Cooling System, and Packaging (Ellingsen et al., 2014). BMS consists of Battery Modules Boards (BMB) to monitor voltage and temperature limits and Integrated Battery Interface System (IBIS) to control battery charge and discharge cycles. The cooling system consists of a radiator for equilibrating the temperature of the battery pack and the packaging consists of a battery retention system and a tray to hold modules in position before sealing the battery pack. A battery cell makes up about 60% of its total weight (Ellingsen et al., 2014). Not only does the cell have mass dominance but also economic and impact dominance over the rest of the battery components.
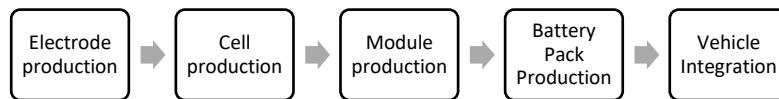
Figure 3- Production Chain of Battery Pack

The production technology and process requirements vary based on the chemistry of the cell and the structural format of the cell. There are two major classifications of cell geometries namely prismatic and cylindrical cells where pouch cells are basically soft-case prismatic cells (Popp et al., 2020). A simplified diagram of these cell geometries is presented in Figure 4 by Zwicker et al., (2020). An overview of the different aspects of these three types of cells is given by Zwicker et al., (2020), Schröder et al., (2017), and *SmartBatt*, (2014) summarized in Table 1. The production and packaging of the cells are done according to the type of cell and application requirements.

Table 1- Comparison of Three Types of Cells

Property	Pouch	Prismatic	Cylindrical
Energy Density	High	Medium	Low
Construction	Light	Heavy	Light
Thermal Stability	Medium	Highest	High
Impact Rigidity	Medium	High	Medium
Size	Large	Large	Small
Casing type	Soft	Hard	Hard
Cell Capacity	Medium	High	Low
Manufacturing Cost	Average	High	Low
Space efficiency	Low	Medium	High
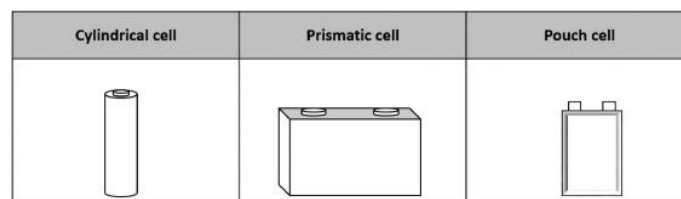
Figure 4- Different types of cells used in automobile batteries

Several studies are available on the description of traditional production processes and resources used to manufacture a lithium-ion cell (Duffner et al., 2021; Jinasena et al., 2021; Liu et al., 2021; Pettinger et al., 2018; Schnell et al., 2019; Tagawa & Brodd, 2009; Väyrynen & Salminen, 2012). Figure 5 shows a general overview of the production processes for a lithium-ion cell.
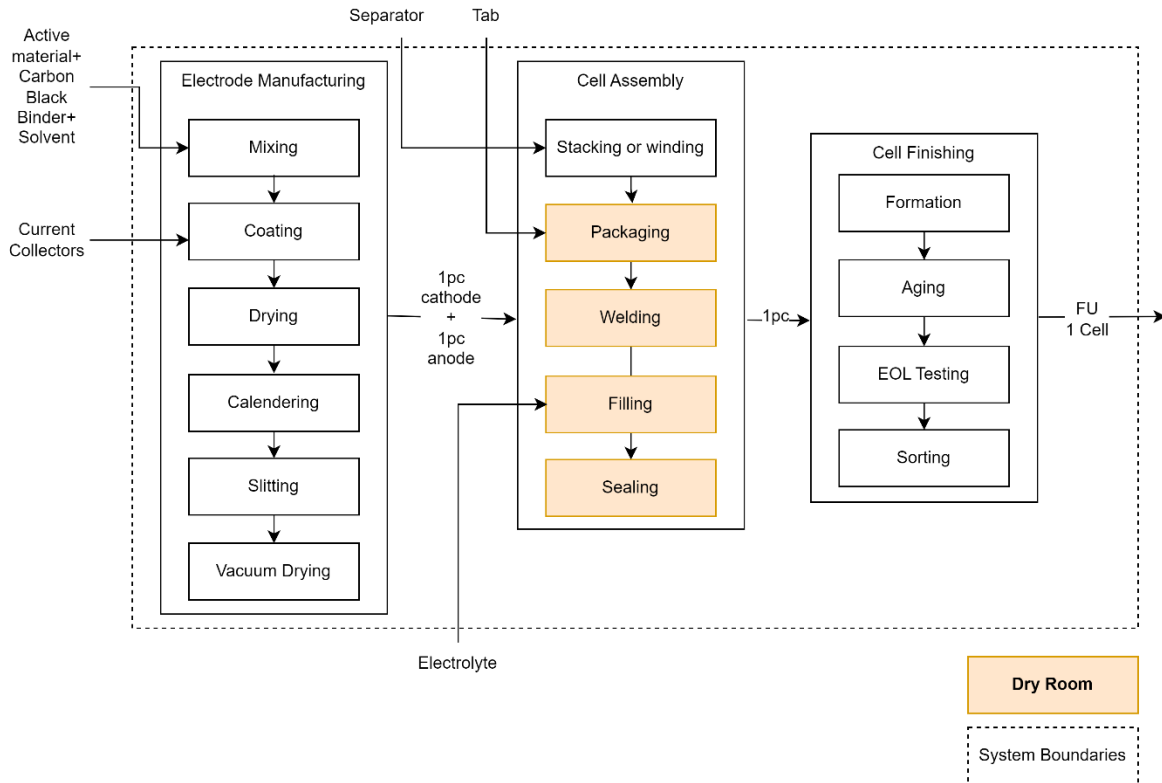
Figure 5- General Production Flow of NMC111 cell

The electrode production starts with mixing active materials, carbon black, and binder. This process is performed in separate mixers for anode and cathode slurry. The contents are first mixed dry and then relevant solvents are added i.e., N-Methyl-2-Pyrrolidone (NMP) for the cathode and deionized water for the anode in the dispersing step. Certain technologies can use to perform the mixing process as mentioned in Table 2. The homogenized slurry of active material is coated with current collectors made of aluminum and copper foils for the cathode and anode respectively. Depending on the structural format of the cell the slurry is coated intermittently or continuously on the aluminum or copper foil via several technologies listed in Table 2. The coating is sequentially done on the top and bottom sides of the foil separately. The coated electrodes are then continuously fed to the drying chamber till the solvent applied in the active material is evaporated and the binder is homogenized with the active material. Drying temperature and speed are integral parameters to achieve suitable adhesion properties in the electrode. The dried electrodes are now statically discharged, cleaned, and pressed through rollers to compress the film and then clean again to remove any traces of external impurities from the electrodes. The calendared rolls are cut into small electrode coils called daughter rolls using rolling knives or laser cutters. The daughter cells are stored in either continuous or infrared dryers for approx. 12 to 30 hours after slitting till the remaining solvents are removed from the coils. The separation process is specific for pouch cells. In this process, the dried coils are unwound and cut down into sheets with the help of laser cutters. Anode and cathode sheets are collected separately to be stacked in the subsequent step. The separated sheets of anode and cathode are stacked alternatively over each other with a separator sheet in between to make a cell stack for pouch cells. In the case of

prismatic and cylindrical cells, electrode rolls are rolled together with a separator roll in between a mandrel and center pin respectively. The current collectors are welded with cell tabs. The cell casing is usually drawn as per the required size of the cell. The stack and rolls are then placed inside the casing and sealed on one side while the other side is kept open for electrolyte filling in the next step. Under vacuum, with a high-precision dosing needle, the electrolyte is filled in a cell case and then the case is vacuum sealed. Roll pressing is an optional process for pouch cells to achieve optimum distribution of electrolytes and reduce rejection rate. The prepared cells are charged and discharged with precisely defined parameters for current and voltage with the help of contact pins in the formation step. During this process, a Solid Electrolyte Interface (SEI) is formed creating a protective layer between graphite particles of anode and the electrolyte. Positive lithium ions are transported and negative electrons are blocked with SEI to ensure electrochemical reactions prevent electrolyte decomposition (Wang et al., 2018). In pouch cells usually after the first charge-discharge cycle, gas is released and captured inside due to the sealed case. This additional degassing step allows to remove the trapped gas and then the cell is sealed again. Aging is basically the storage and monitoring of cell performance parameters under high and low temperatures. The aged cells are discharged back to the shipping state to charge and tested for certain tests and sorted into grades according to their performance. The next step after cell production is module and pack production (Figure 3). A collection of cells is called a module and a collection of modules makes a battery pack, electrically connected, and controlled with the help of a Battery Management System (BMS). A typical battery pack consists of 12 battery modules, each carrying 30 cells (Ellingsen et al., 2014). The cells are first packed in either series or parallel into modules and then modules along with the cooling system and packaging accessories are packed into a battery pack.

Table 2- Production steps of lithium-ion cell; where p~ pouch, pr~ prismatic, c~ cylindrical cells.

Sno	Process	Sub process	Input	Technology
1	Mixing	Dry mixing	active material, binder, additives	Batch mixers
				Continuous mixers
		Dispersing	Solvent	same as mixing
2	Coating	Topcoat Bottom coat	homogenized slurry, current collector	Continuous (pr,c)
				Intermittent (p)
				Both modes
3	Drying		collector coated with electrode paste	convective air heating
				infrared heating
				laser heating
4	Calendaring	statically discharged	dried and adhesed electrode	discharging roller
		cleaning		brushes or air flow
		compacting		compressing roller
		cleaning		brushes or air flow
5	Slitting	cutting	Calendared rolls	rolling knives or laser cutters
		cleaning		suction or brushes
		rewounding		
6	Vacuum Drying		daughter rolls	continuous dryers
				infrared dryers
7	Separation (p)	unwounding	coiled roll	
		cutting	straight roll	laser ablation
				laser cutting
				shear cutting
8	Stacking (p) or Winding (pr,c)		electrode sheets, separator	Laminating heat press pocket pressing
9	Packing & Sealing	welding tabs and current collectors	casing, stacks, and rolls	ultrasonic welding
				laser welding
		Sealing		impulse sealing
				contact sealing
10	Electrolyte filling & Final Sealing	vacuum filling	cell with casing, electrolyte	
		wetting		Roll Pressing (p)
				Vibrating table (pr,c)
				Vacuum sealing (p)
		sealing		Crimping (pr,c)
				Beading (pr,c)
				Welding (pr,c)
11	Roll pressing (p)		cell filled with electrolyte	
12	Formation			
13	Degassing (p)	Degassing	pouch cell	
		Resealing		
14	Ageing	High Temperature	complete assembled cell	Ageing Shelves
		Low Temperature		Ageing Tower
15	End of Line Testing	pulse tests, open circuit voltage tests, leakage tests, internal resistance measurements, optical inspections.	Charged cell	EOL test rig
16	Sorting		Discharged cell	

2.2 Recycling Processes for Lithium-ion Batteries

Recycling lithium-ion batteries is quite challenging and even dangerous compared to lead acid batteries (Gaines, 2014). Still, there are certain recycling methodologies in research and practice to recover valuable contents from scrap and spent cells securely.

Sakti et al., (2015) assume a 95% yield rate in cell stacking step which means that 5% of the input is lost as scrap after cell stacking. The study also proposes about 7.8% scrap from electrode materials, 9.8% from current collectors, and 2% from separators. The Process Based Cost Model (PBCM) implemented in the study suggested that about 200 – 300 MWh of battery production capacity may require to get economies of scale. Considering the 200 MWh capacity of the cell production plant means for a 224 Wh/kg energy density cell, 0.89 million kg cell mass could be produced, of which about 57% mass is of electrode material combined (Daniel, 2008). 7.8% scrap from electrode materials means that out of every 0.51 million kg input electrode material, 0.04 million kg of the electrode materials is lost. The lost material not only contributes to direct resource loss but also the cost incurred due to production and processing and relevant overheads. Hanisch et al., (2015) also compiled in production scrap rates from various studies and declared about 86.7% yield rate for electrode material whereas high scrap rates are observed during the final steps of end-of-life testing and sorting of cells. Marshall et al., (2020) mentioned that about 80% of the material can be recovered by rejected cells during the formal quality control check procedures which are 3% more than the spent cells collected for post-consumer recycling. Hence there is marked potential to recover valuable contents of electrode paste from the rejected cells to be reused again and achieve a circular economy.

Several recycling techniques are in practice and research, depending on the extent to which the contents of the cells need to be recovered namely pyrometallurgical, hydrometallurgical, direct and in-house production scrap recycling. Zhou et al., (2020) summarized the comparisons in the first three recycling processes.

2.2.1 In-Production Scrap Recycling

Figure 6 provides a general overview of in-house production scrap recycling. The principles of circular economy aim to remove the generation of waste at its immediate steps therefore it is important to know the benefits and challenges associated with the recycling opportunities while the batteries are being produced. Hanisch et al., (2015) explained to deal with the scrap generated after the coating process. With the help of certain separation techniques, the electrode paste can be recovered before the addition of electrolytes and utilization of energy-intensive dry room operations. The scrap output consists of electrode paste coated with current collectors which after separation can be reused again for dispersing/mixing followed by coating and the preceding processes for cell manufacturing. Recycling of in-house production scrap electrodes is technically, economically, and environmentally feasible. The cells can easily follow reverse engineering as newly manufactured electrodes have low adhesion due to the not yet developed Solid Electrolyte Interface (SEI) because the cells have not yet undergone formation and charging step downstream. In-Production scrap recycling does not require a high amount of energy as required for the thermal and chemical decomposition of cells in the case of hydrometallurgy and pyrometallurgy nor relatively higher infrastructure cost of direct recycling. Furthermore, it has significantly low risks due to the elimination of disassembling needs to further recycle the important contents of the cells. However, according to Hanisch et al., (2015), there are certain limitations for the in-production recycling of electrode scraps due to dependence on N-Methyl-2-pyrrolidone (NMP) for wet separation, while in the case of dry mechanical process, fine impurities from aluminum foil may affect the resynthesis of cathode active materials.
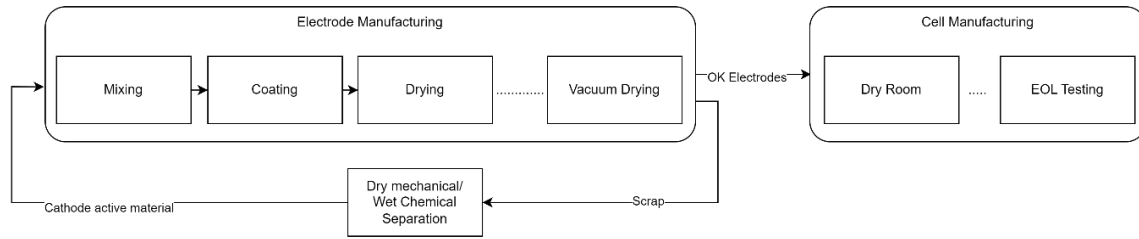
Figure 6- An Overview of In-House Scrap Recycling

2.2.2 Direct Recycling

Figure 7 gives an overview of a general direct recycling process. Direct recycling is the process of recovering materials while retaining the cathode structure, thus saving energy and cost by removing the need for material dissolution and purifying it again (Montoya et al., 2022). Several approaches to direct recycling are cathode-to-cathode, electrochemical, mechanical, and cathode-healing™ technologies (Sloop et al., 2020). Inhouse production scrap will be the dominant source for recycling (*Circular Energy Storage*, 2020) quantity of which varies from 5 to 30 % as per the learning curve and level of expertise of battery producers (Gaines et al., 2021). Direct recycling generally comprises a mechanical and chemical process. The mechanical process involves disassembling the battery, crushing the cell, and separating the degraded active powder.

Further, by using diethyl carbonate on the degraded active powder, it is possible to extract the electrolyte salt and then reformulate it to obtain electrolytes with improved cycling performance (Gaines et al., 2021). Several mechanical and physiochemical separation methods are available like magnetic separation, separation with dissolution in water, and surface chemistry-based separation by froth floatation. Separating cathode active material from collector foil requires excessive use of organic solvents usually NMP (Sloop et al., 2020). The recovered cathode active material is then upgraded by relithiation of lithium ions and/ or upcycled in the subsequent step by changing the chemical composition of the cathode powder thus making it usable for other cell chemistries (Gaines et al., 2021).

Direct recycling proves to be an efficient and low-carbon recycling technology compared to hydrometallurgy and pyrometallurgy (Gaines et al., 2021), however, there are several challenges to implementing this technology similar to the wet separation discussed in in-production scrap recycling. Excessive use of NMP solvent is not cost-effective, and the recycled material can have impurities making it unsuitable for direct use as active material in new electrode production and generating excessive liquid waste. An alternative approach to separate and remove binder from electrode bits is via thermal pyrolysis which is also under conflict due to the production of Hydrogen Floride which besides being toxic to humans also corrosive for cathode recovery (Sloop et al., 2020). The direct recycling model used in this study is adapted from S. Jiang et al., (2022).
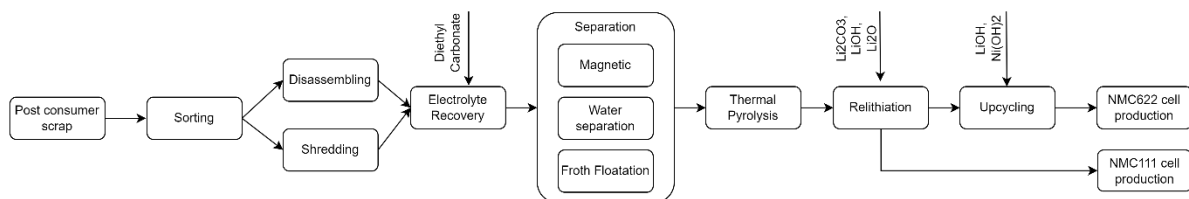
Figure 7- An Overview of Direct Recycling

2.2.3 Hydrometallurgy

Figure 8 provides an overview of the hydrometallurgical recycling model analyzed in this study. Hydrometallurgy is a relatively mature, and reliable method for recycling lithium-ion batteries. The material is recycled following the steps; pre-treatment to remove impurities, leaching to dissolve valuable materials in specific leaching agents, and extraction using certain organic solvents (*Dorri et al, 2022*). Pre-treatment includes sorting and disassembly according to the chemistry of spent cells to decide the type of leaching agent to be used (Mantuano et al., 2006). H. S. Kim & Shin, (2013) has divided the primary physical process into mechanical operations, heat treatment, mechanochemical reactions, and dissolution phenomena. After pre-treatment leaching is done to separate impurities using sulfuric acid (H_2SO_4) with hydrogen peroxide (H_2O_2), and sodium sulfide as a reducing agent (Du et al., 2022). Following leaching, the organic contaminants are segregated, and leachate is directed to the initial solvent extraction unit by modifying the alkalinity of the solution using chemicals like kerosene and sodium hydroxide (NaOH) and extractants (Ma et al., 2022). In this study, Di(2-ethylhexyl) phosphoric acid (P204) and 2-Ethylhexyl 2-Ethylhexyl Phosphate (P507) are used as extractants (Rao et al., 2020). During this stage, kerosene and P204 extractants eliminate metallic impurities, such as iron and manganese (Ekberg & Petranikova, 2015). The remaining materials are then forwarded for the subsequent extraction process aimed at separating other targeted substances. The extraction of cobalt, nickel, and lithium occurs in the subsequent solvent extraction unit and stripping phase (Neumann et al., 2022), which have been merged into a single sector in the model to facilitate the analysis of environmental impacts. Additionally, P507 is another extractant that is utilized to isolate cobalt from the solution, which is subsequently extracted in a stripping process (Neumann et al., 2022). Notably, the input for the hydrometallurgical process typically originates from a recycling plant that employs pyrometallurgical methods.

Although hydrometallurgy has better environmental and health performance relative to pyrometallurgy due to high purity active materials recovery, energy efficiency, less wastewater, and no air emissions generation (L. Li et al., 2015). However, traces of cobalt, nickel, and manganese in the leachate may complexify the lithium metal recovery (Fahimi et al., 2022). Also, high consumption of acid and reducing agents may make the process risky, costly, and unsustainable.
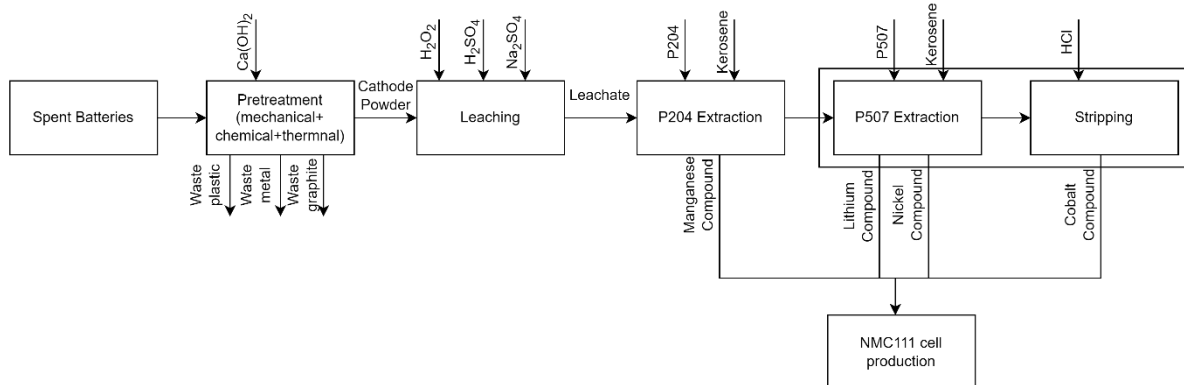
Figure 8- An Overview of Hydrometallurgy

2.2.4 Pyrometallurgy

Figure 9 provides an overview of the pyrometallurgical recycling model analyzed in this study. Pyrometallurgy is another type of recycling process for lithium-ion batteries in two steps, roasting or smelting followed by hydrometallurgical separation (M. Chen et al., 2019). Under pyrometallurgy, end of life batteries may also be pre-treated first by several thermal pre-treatment methods like

incineration, pyrolysis, and calcination to recover cathode active materials (Makuza et al., 2021). Under the pyrolysis zone, plastic parts are burned. This combustion process is typically incomplete, which can result in the production of harmful chemicals like dioxin, furan, and toxic halogens from burning electrolytes and binders. In order to avoid these hazardous gas emissions into the environment, a gas treatment system must be installed in the system. Furthermore, under metal smelting and reduction, slag formers such as CaO and SiO_2, along with coke reductant, play a crucial role in the segregation of materials into alloy and slag during the process (Brückner et al., 2020; Cheret & Santen, 2007; Dai et al., 2019). A formal pyrometallurgical recycling process includes the burning of organic matter under high temperatures, reduction of metal oxides to recover metals, and leaching for refining cobalt and other cathode-active materials (Zheng et al., 2017). It is also called the combination of pyro and hydrometallurgical recycling (Georgi-Maschler et al., 2012). Hence after reduction, hydrometallurgical recycling process steps like leaching and extraction are followed as explained in the previous section. However, oxidation in the presence of H_2O_2 and sintering or firing is done to recover cobalt in the form of lithium cobalt oxide $LiCoO_2$ (Dunn et al., 2012).

Pyrometallurgy besides simplicity and maturity has higher throughput and raw material applicability (Jie et al., 2020). Also, it allows mixed input of end-of-life batteries and delivers an output that can be used in developing various battery chemistries due to the recovery of fundamental "building blocks" (M. Chen et al., 2019). However, high energy consumption, inefficient recovery of materials of interest like lithium, and high emissions due to the burning of organic matter are major drawbacks of this recycling technology (Yadav et al., 2020). Furthermore, high capital and operational cost are also involved with pyrometallurgy (Cheret & Santen, 2007).
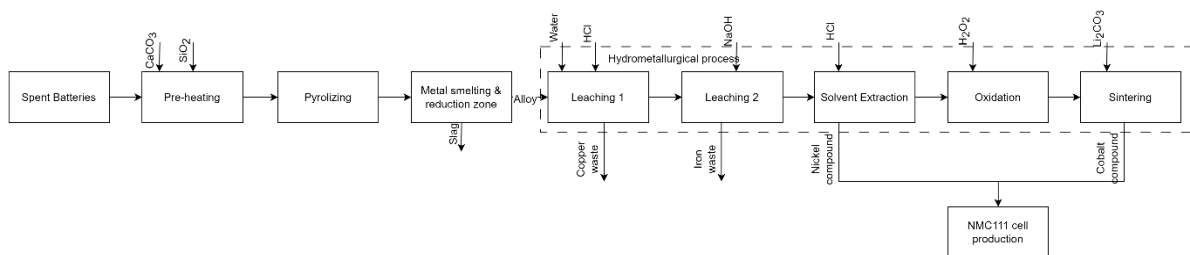
Figure 9- An Overview of pyrometallurgy

3   Case and Methodology

3.1   LCA

The life of a product starts with its design and ends with end-of-life activities like recycling and waste disposal. All the activities involved in a product's life cycle have environmental impacts like resource consumption, emissions of substances, and radiation. Life cycle assessment (LCA) is a framework used to evaluate the environmental impacts of a product throughout its life cycle, including climate change, ozone depletion, pollution, depletion of resources, and their impact on human health and ecosystems (Rebitzer et al., 2004). In an LCA study, the specific product being analyzed is called the foreground, while the background represents the industrial economy as a whole, drawn from reference databases. The foreground is designed by the modeler, while the background is selected and adapted from existing sources or available databases (Kuczenski et al., 2018).

3.1.1   Goal & Scope

3.1.1.1   *Aims of the study*
The aim of this study is to conduct an LCA to evaluate the environmental impacts of combined production and recycling systems for an NMC111 lithium-ion cell.

3.1.1.2   *Functional Unit*
The function of the system under study is to produce an NMC111 cell. Hence the functional unit in this study is **1 kilogram (kg) of NMC111 cell produced** from both virgin and recycled inputs.

3.1.1.3   *Scope of LCA*
As the constituents of positive electrode paste used in the cathode production have both mass and economic dominance in a cell, the recycling inputs of only these constituents are taken into account to produce new cells.

3.1.1.4   *System boundaries*
System boundaries include foreground processes of cell manufacturing and recycling while upstream raw material extraction, energy and transport are modeled outside the system boundaries in the background. In this study cradle-to-gate LCA is performed. Cradle-to-gate LCA includes manufacturing process while not including use, end-of-life, and disposal phases (M. Li & Subramaniam, 2017). Traditional cradle-to-gate LCA is modified here using scrap cells and recycling processes as an input to cell production process. Here in spite of end-of-life cells, recycling is performed for postproduction scrap.

The LCA calculations were accomplished by connecting the foreground system consisting of combined production and recycling models, to a background system, which is the *Ecoinvent 3.9* database consisting of technosphere and biosphere material and emissions flows.

3.1.1.5   *Life Cycle Impact Assessment (LCIA) Method*
This study has selected the attributional LCA methodology as it is the most used, feasible, robust, and the least complex LCA methodology that can describe environmentally relevant flows associated with the life cycle of a product or service system (Ekvall, 2019).

In the impact assessment method, the selection of impact categories, category indicators, classification, and characterization is made. Out of the various impact assessment methods available, the ReCiPe Hierarchist (H) method was chosen for this assessment. The reason for selecting this method is its ability to differentiate between midpoint and endpoint-level indicators. Moreover, it is one of the most relevant and up-to-date impact assessment methods with regional validity for the

European region (*TOSCA Sustainability Framework, 2022*), and it includes most of the relevant impact categories that need to be analyzed (*OpenLCA, 2015*). The Hierarchist (H) perspective is based on scientific consensus and common policy principles, which group assumptions and calculate the plausibility of impact mechanisms over balanced time frames (*LCIA: The ReCiPe Model | RIVM*, 2018).

3.1.1.6   Impact Categories

ReCiPe Midpoint (H) Hierarchist methodology has 18 midpoint indicators pathways to 3 endpoint indicators as shown in Figure 10 by Roux & Gérand, (2014). Out of these 18, Climate Change, Particulate Matter Formation, Fossil Fuels, and Minerals Consumption are selected as they are quite relevant to the objectives of the study for discussing LCIA results.

The impact of emissions on climate change can be compared by using metrics like radiative forcing or temperature response. Each step in the cause-effect chain requires a modeling framework, but simpler indicators can also be used to quantify contributions to climate change. Indicators can be given in absolute or relative terms, with CO_2 often used as a reference gas. The choice of an indicator depends on the climate policy goals, and the Global Warming Potential (GWP) is a commonly used emission indicator for transferring emissions of different gases to a common scale. The GWP was adopted by the IPCC for a time horizon of 100 years to implement the multi-gas approach in the UNFCCC and the Kyoto Protocol (*IPCC, 2014*).

Particulate Matter (PM) is a mixture of solid particles and liquid droplets found in the air, which can range from visible dust to tiny particles only detectable by an electron microscope. The two most common types of PM are PM_{10} and $PM_{2.5}$, with diameters of generally 10 micrometers or smaller and 2.5 micrometers or smaller, respectively. PM can come from various sources, including emissions from power plants, industries, automobiles, and fires. Fine particles ($PM_{2.5}$) are the most harmful and can cause serious health problems when inhaled, including reduced visibility and haze in national parks and wilderness areas. Based on Bell et al., (2007) research, the selection of the seven chemical constituents of $PM_{2.5}$, which include sulfate, nitrate, ammonium, elemental carbon, organic carbon, silicon, and sodium ion, was determined by their contribution of greater than 1% of $PM_{2.5}$ mass for either seasonal or annual averages, and their combined total constitutes at least 79-85% of $PM_{2.5}$ mass. $PM_{2.5}$ also contains heavy metals like As, Cd, Co, Cr, and Ni which can cause significant harm to health when they enter the human body (Zhao et al., 2021). The EPA regulates inhalable particles and has implemented national and regional rules to reduce PM emissions to meet air quality standards (US EPA, 2016). Giovanis, (2015) found a significantly negative relation between recycling solid municipal waste and $PM_{2.5}$ emissions as recycling reduces waste in landfills and pollution in the environment. Recycling helps to conserve natural resources and reduce the amount of pollution released into the air. The manufacturing of products from recycled materials generates less air pollution than creating products from mined materials. Recycling also reduces the incineration process, which produces air pollutants. Recycling waste materials conserves energy and helps to reduce the acquisition of virgin materials from the natural environment (Morris, 1996).

Besides Human Health and Ecosystem Quality, Natural Resources is also an Area of Protection (AoP) encompassed in LCA. As battery production besides energy requires materials extracted from the lithosphere. Therefore, two indicators Surplus Ore Potential (SOP) and Fossil Fuel Potential (FFP), relevant for scarcity are selected for analyzing the impact of these resources deeply in lithium cell production.

Lithium batteries require an extensive amount of minerals comprising abundant elements like silicon, aluminum, iron, etc., and elements prone to the risk of future supply like lithium and cobalt in their production. Therefore, an indicator selected for explaining impact results is mineral resource

consumption. Mineral resource groups consist of chemical elements, minerals, and aggregates, which exist naturally or as part of a human-made supply and can have value for human use in technology. The focus on protecting mineral resources lies in their potential value for human use in technology and any harm to this potential, caused by human activity, is measured as a reduction or loss of value (Berger et al., 2020). ReCiPe uses an indicator called Surplus Ore Potential (SOP) to measure this loss by calculating the additional amount of ore that can be mined per unit of resource extracted in the future (Vieira et al., 2017).

To obtain the fossil resource scarcity indicator in the ReCiPe package, the higher heating value of extracted fossil resources is divided by the higher heating value of crude oil (Huijbregts et al., 2017). The indicator for fossil fuel depletion can be considered a measure of the scarcity of fossil energy as extracting more fossil energy in the future is likely to increase the energy demand required for extraction (Arvidsson et al., 2021). Similar to extraction, processing, transportation, and relevant upstream processes in production, while downstream processes such as disposal and recycling may also be powered by fossil resources. That makes Fossil Fuel Depletion Potential (FDP) or Fossil Fuel Potential (FFP) an interesting indicator for this study.
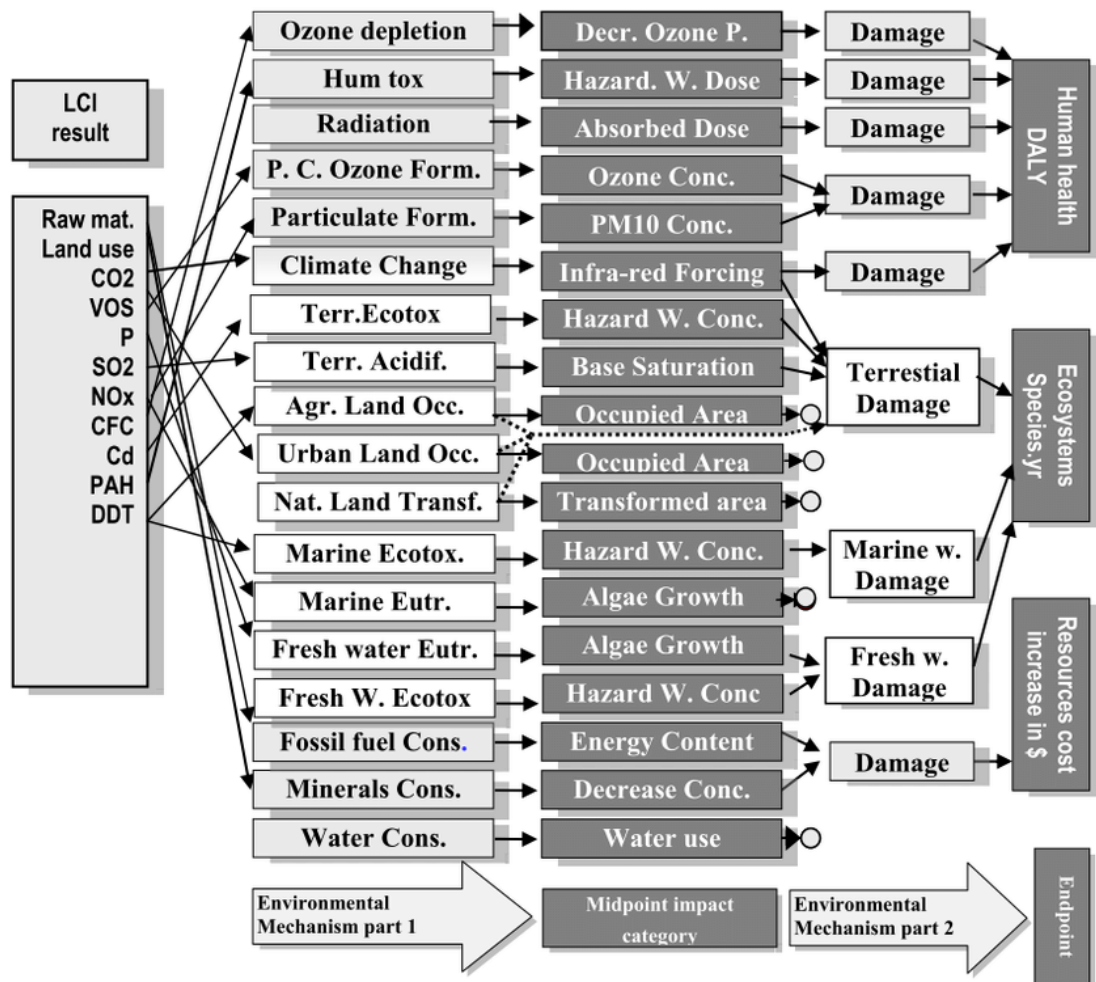

Figure 10- LCI to midpoint and endpoint LCIA mechanism for ReCiPe methodology

3.2 Modelling the Combined Production and Recycling System

3.2.1 Model in ARDA

The calculations were carried out using ARDA, an LCA software tool based on MATLAB interface created by researchers at the Industrial Ecology Program at NTNU (*Majeau-Bettez, G. and A. H. Strømman, 2016*). Arda utilizes an input-output analysis approach to perform mathematical calculations for LCIA. The LCIA calculation process through Arda involves the assessment of environmental impacts based on the unit processes, materials, and energy required to achieve the functional unit. To connect all the materials and energy consumed in unit processes to the functional unit, specific vectors and matrices are formed.

The key matrices and vectors are given in Table 3 and the generalized representation of A matrix is demonstrated in Table 4. A and L are square matrices where rows (*i*) are equal to columns (*j*), A_{ij} (*i=j*), contains components that correspond to the energy and materials needed for unit processes to produce a single unit of output for existing foreground processes. Each component is represented in the format of a_{ij}, indicating the output flow required from process *i* to process *j* per unit production in process *j*. The matrix also displays connections between foreground (*f*) and background (*b*) processes through the *ff, fb, bf,* and *bb* indices.

Table 3- Matrices and Vectors used in LCIA contribution analysis.

Name	Description	Dimension	Nature
A	Process Input Requirements Matrix	*pro X pro*	Matrix
Y	External Demand Vector	*pro X 1*	Vector
X	Output for a given External Demand Vector	*pro X 1*	Vector
L	Output per unit External Demand Matrix	*pro X pro*	Matrix
S	Stressors intensities per unit Output Matrix	*str X pro*	Matrix
e	Total Stressors from all Processes for a given External Demand Vector	*str X 1*	Vector
C	Characterization Matrix	*imp X str*	Matrix
d	Total Impacts from all Processes for a given External Demand Vector	*imp X 1*	Vector
E	Stressor from each Process for a given External Demand Matrix	*str X pro*	Matrix
D_{pro}	Total Impacts from each Process for a given External Demand Matrix	*imp X pro*	Matrix
D_{str}	Impacts from each Stressors for a given External Demand Matrix	*imp X pro*	Matrix
D_{pro, f}	Total Impacts due to Foreground Process	*imp X pro*	Matrix
D_{pro, ff}	Impacts generated in Foreground due to Foreground Processes	*imp X pro*	Matrix
D_{pro, bf}	Impacts generated in Background due to Foreground Processes	*imp X pro*	Matrix

Table 4- General representation of A matrix

Foreground to Foreground: This section includes input from foreground processes per unit output to foreground process. A_{ff}	**Foreground to Background:** This section includes input from foreground processes per unit output to a background process. This is usually zero unless the foreground system is so huge that it has an impact on the background economy. $A_{fb}=0$
Background to Foreground: This section includes input from background processes in Ecoinvent Database per unit output to foreground process. A_{bf}	**Background to Background:** This section includes input from background processes per unit output to a background process. A_{bb}~ Ecoinvent 3.9

The general formula of all matrices and vectors given in Table 3 is derived from the production balance equation given in Equation 1:

$$X = AX + Y$$

Equation 1

$$X - AX = Y$$

$$X(I - A) = Y$$

$$X = (I - A)^{-1}Y$$

Where $(I - A)^{-1} = L$

$$X = LY$$

Equation 2

$$e = SLY = SX$$

Equation 3

$$d = CSLY = CSX = Ce$$

Equation 4

$$E = S\hat{X} = S\widehat{LY}$$

Equation 5

$$D_{pro} = CS\hat{X} = CS\widehat{LY}$$

Equation 6

$$D_{str} = C\hat{e} = C\widehat{SX} = C\widehat{SLY}$$

Equation 7

Furthermore, to calculate the total impacts due to foreground processes, the equation for $D_{pro, f}$ is derived as:

$$D_{pro,f} = D_{pro,ff} + D_{pro,bf}$$

Equation 8

Arda requires, in a specific template, data of foreground matrix (A_{ff}), background to foreground matrix (A_{bf}), demand vector (Y), and foreground stressors matrix (S_f also called F_f) while characterization matrix (C), background matrix (A_{bb}) and background stressors matrix (S_f) are built in Ecoinvent database in terms of Technosphere and Biosphere matrices. With this information, Arda calculates, with the help of the MATLAB program, Output Vector (X), Total impact Vector (d), total stressors vector (e), and Total Impact due to foreground processes ($D_{pro, f}$).

Arda template and Ecoinvent 3.9 database is then uploaded to the MATLAB through a Graphical User Interface (arda_gui or lca_gui) and impact calculation is done with the help of MATLAB program. Structural Path Analysis (SPA) to find the most intensive emissions pathways and Taylor Series to analyze the most intensive tiers can also be calculated using this program.

3.2.2   Issues with Close Loop Modelling

In order to understand the major issues with calculating the impact in closed-loop recycling models, an example is discussed from LCA literature by NTNU (*Life Cycle Analysis - TEP4223 - NTNU*, 2022).

A circular model presented in Figure 11 runs in infinite loops. Here '*y*' represents the amount of recycled content that is fed back to the production system and combined with the virgin material '*1-y*' to deliver a functional unit '*1*' for use. Setting up an *A* matrix based on the model in Figure 11 where the Production Process is represented as P, Use as U, Recycling as R, and Virgin Mining as V.
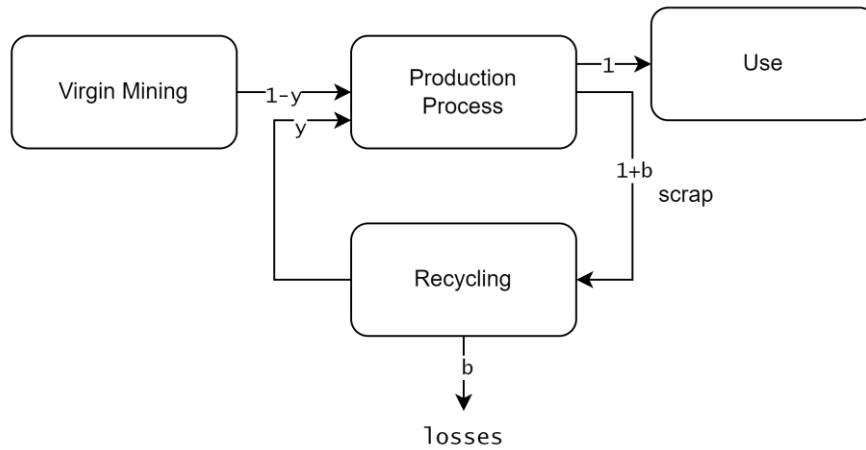
Figure 11- An Infinite Loop Circular Model for Production Scrap Recycling

			P	U	R	V
		P	0	1	0	0
A	=	U	0	0	1+b	0
		R	y	0	0	0
		V	1-y	0	0	0

Leontief inverse L can be expanded using geometric series as:

$$\sum_{i=0}^{\infty} A^t = I + A + A^2 + \ldots A^n = L = (I - A)^{-1} \qquad\qquad if\, \rho(A) < 1$$

<div align="right">*Equation 9*</div>

Where *t* represents the number of tiers of production starting from the demand or functional unit in foreground and stretching back along the value chain in background system, and *ρ(A)* is the spectral radius equal to the maximum eigen value of matrix *A*. Multiplying demand vector *y* of both sides of Equation 9 gives,

$$\sum_{i=0}^{\infty} A^t y = y + Ay + A^2 y + \ldots A^n y = Ly = (I - A)^{-1} y = X_t \quad if\, \rho(A) < 1$$

Where,

$$X_0 = A^0 y = y$$

As zeroth tier output is the functional unit or demand placed in the system. The output of the first tier is given by:

$$X_1 = A^1 y = Ay = AX_0$$

Hence, generally,

$$X_t = A^t y = AX_{t-1}$$

Using Equation 10, the outputs in tiers for the circular model under study are calculated as follows:

Tier	0	1	2	3	3_{acc}
P	1	0	0	y(1+b)	1+y(1+b)
U	0	0	y(1+b)	0	y(1+b)
R	0	y	0	0	y
V	0	1-y	0	0	1-y

(with $X_t =$ to the left of the table)

In tier 3, the production process places a demand on the recycling process with *y(1+b)* which is not the actual demand from the recycling process as the actual demand from the recycling process was *y*. This is the recycled material of the previous round, and it can be interpreted as that the material required for production before it is recycled from production scrap.

Typically, the concern is not an infinite number of recycling cycles, but rather only the impact of a single cycle. To analyze this impact, a non-circular system is modeled with a beginning and an end, rather than a loop. This approach disregards previous and subsequent cycles. The tricky part is determining where to break the cycle.

A linear single cycle model is proposed for the same system in Figure 12 where a dummy process is created named scrap cells to break the closed loop of producing a functional unit.
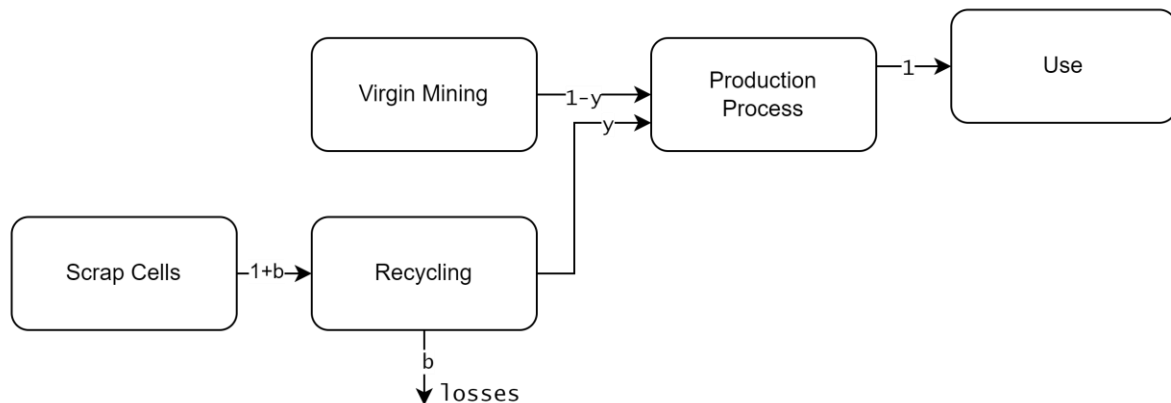
Figure 12- Single Cycle Linear Model for Production Scrap Recycling

Setting up an *A* matrix again based on the linear model presented in Figure 12 where the Production Process is represented as P, Scrap cells as S, Use as U, Recycling as R, and Virgin Mining as V.

	P	S	U	R	V
P	0	0	1	0	0
S	0	0	0	1+b	0
U	0	0	0	0	0
R	y	0	0	0	0
V	1-y	0	0	0	0

(with $A =$ to the left of the table)

Using Equation 10, the outputs in tiers for the linear model under study are calculated as follows:

Tier	0	1	2	3	3_{acc}
P	1	0	0	0	1
S	0	0	y(1+b)	0	y(1+b)
U	0	0	0	0	0
R	0	y	0	0	y
V	0	1-y	0	0	1-y

$$X_t =$$

The accumulated output in this single cycle linear model is *y(1+b)* demand placed on scrap cells by the production process where scrap cells belong to the scrap generated by production in the previous production round.

3.2.2.1 *Issues with Combining Models of Different Functional Units.*

The study is combining the two LCA models each having LCI based on different functional units. The production model has a functional unit of 1 kg of cell produced while the recycling model has a functional unit of 1 kg of cell recycled. The output of the recycled model cannot be directly supplied as an input in the production model as the recovered content is less in each type of recycling methodology than the amount required to produce 1 kg NMC111 cell.

One approach is to scale the output of the recycling linearly up to the input required to produce 1kg of the cell. However, this approach was not proven to be effective as a dramatic rise in recycling impact is observed as shown in Figure S 1 in Supporting Information. Pizzol et al., (2021) discuss the limitations of the linear framework of LCA to calculate impacts for scalable and emerging technologies. It explains that while the potential environmental impacts associated with demanding a product increase linearly with demand, due to economies of scale, industrial synergies, efficiency gains, and system design, activities at different scales will perform differently in terms of life cycle impact in a non-linear way. Thus, LCA is linear in terms of functional unit dependence but the coefficients that define each activity can be based on the technologies as defined by the practitioner, making it possible to model systems with non-linear properties.

Hence, parametrized LCA could be more useful while modeling such systems. Where parameterization is another approach to displaying LCA information in the form of raw data and equations, as opposed to the calculated values in unit process datasets contained in databases (Cooper et al., 2012) as these databases are composed of highly aggregated data that may lead to low-resolution inventories compromising data precision and hence accuracy in results (Manjong et al., 2021). However, when it comes to performing parametric LCA for lithium-ion cells, the lack of sufficient and up-to-date data on their production and recycling is a great challenge as mentioned earlier due to the unavailability and transparency of data from industrial-scale studies (Keppeler et al., 2021).

Therefore specifically for modeling the recycling model, the allocation approach is selected according to the guidelines of *ISO-14044, (2006*) i.e. first try dividing the unit process into sub-processes or expanding the product system to include co-products to avoid allocation whenever possible. However, if the division of processes based on multiple products is not possible, the inputs and outputs of the system should be partitioned among its different products in a way that reflects the underlying physical relationships between them. The guidelines further elaborate that reuse and recycling processes may allocate inputs and outputs associated with unit processes among multiple product systems. However, due to limited knowledge about the recovery quality of the recycled material, it is assumed that the inherent properties of materials are not altered after recycling as extensive analysis of the chemical properties falls beyond the scope of the study and academic limitations of the thesis writer.

3.2.3 Mass Allocation-Based Combined Model

To equalize the functional unit as 1kg of NMC111 produced, the study proposed a mass allocation-based input from the recycling model. With this approach, the recycling model is made independent of the quantity of cell input, by allocating the input requirements of every recycling process step with respect to the amount of recovered cathode active materials.

Figure 13 represents the generalized representation of production and recycling models of 1kg cell produced and recycled respectively. According to Figure 13, in the production model, X and Y materials are required in x_1 and y_1 quantities to produce positive electrode paste. While in the recycling model, only x_1' and y_1' quantities of X and Y materials are recovered by recycling 1kg of spent or scrap cells, which is quite less than the input requirement of positive electrode paste. Hence positive electrode paste cannot be produced by x_1' and y_1' quantities of X and Y materials, therefore more than 1kg of cells needs to be recycled to fulfill the production requirement. The requirements for producing 1kg of cell and output from recycling 1kg cell are different as they belong to two different functional units of production and recycling models, that is, 1kg cell produced and 1kg of cell recycled respectively. The challenge is transforming the recycling model in a way that it can be connected with the production model, and therefore, can be used to calculate the total impact of producing 1kg of cell with virgin and recycled constituents of positive electrode paste.
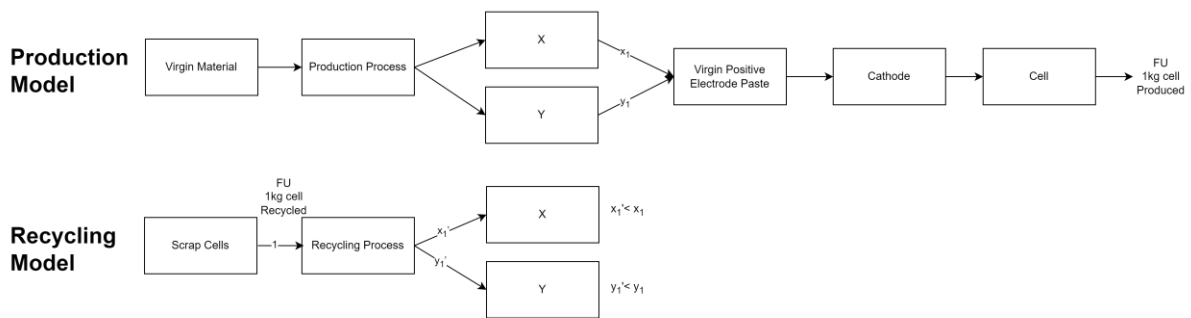
Figure 13- Generalized representation of Production and Recycling Models

Figure 14 represents a possible way of transforming the initial recycling model R having a functional unit of 1kg of cell recycled and producing x_1' and y_1' quantities of X and Y as recycled products. The recycling process requirements are attributed to recycled products X and Y with the help of mass-based allocation. The recycling process (R) is split between the products X and Y in such a way that the total impact of the initial recycling process (R) will be the same as the total impact of transformed recycling processes (R1+R2). This is done to transform the functional unit of the recycling model from per kg of cell input to per kg of output recovered from the process. As the initial recycling model is developed for 1kg of NMC cell recycled, the input is kept with the same consistency in the transformed model. As the interest is to connect the outputs or products recovered from the cell to the production model, the products obtained from the recycling processes are scaled as per the requirement of the production of positive electrode paste irrespective of the input of 1kg of the cell. The more the material requirement of producing positive electrode paste from the recycling process, the more the impact is due to mass allocation. In this way, the model basically burdens the materials recovered or products obtained from the recycling processes individually.
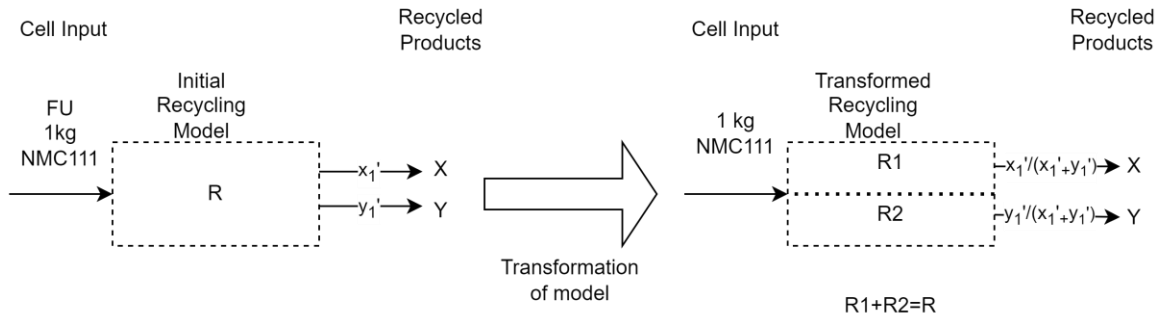
Figure 14- Transformation of Recycling Model

Figure 15 shows the combined model of cell production with 90% virgin and 10% recycled positive electrode paste. Where $x_1'/(x_1' + y_1')$ and $y_1'/(x_1' + y_1')$ are the allocated outputs from the recycling process for X and Y respectively. The recycling model is now connected with the production model. The combined model developed using the allocation methodology discussed above is applied individually with cases of cell production with virgin and recycled through direct, hydrometallurgy, and pyrometallurgy. Impact from each of these cases is tested by changing proportion of recycled and virgin content in cell production.
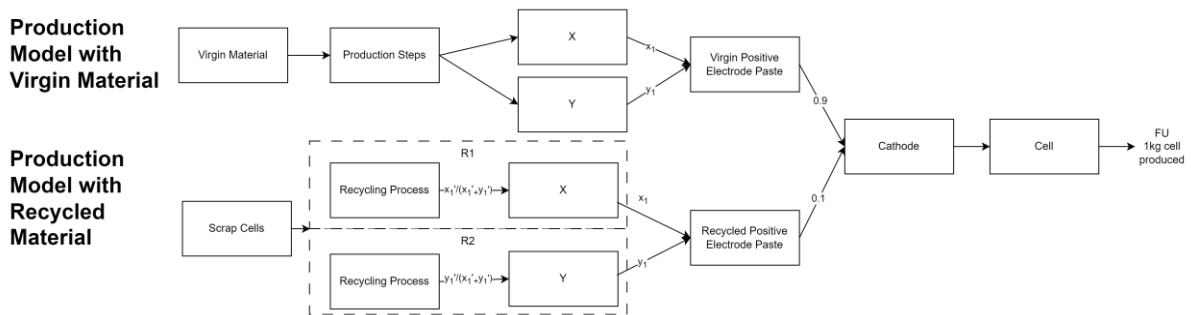
Figure 15- Combined Production Model

3.2.3.1   Virgin Production Model and LCI

Materials and energy inputs are required to produce a cell. As discussed earlier the foreground system consists of the specific product system under analysis. The components and material of interest are selected in the foreground system as shown in Figure 16. Figure 16 combines the production and recycling model of producing positive electrode paste. The major components of the cell are the cathode, anode, container, electrolyte, and separator. Whereas production operations include electricity and heat energy requirements for mixing, coating, drying calendaring, slitting, stacking, filling, formation, floor heating, dry room, and miscellaneous cell production operations as discussed in Production Process of Lithium-ion Batteries section earlier.
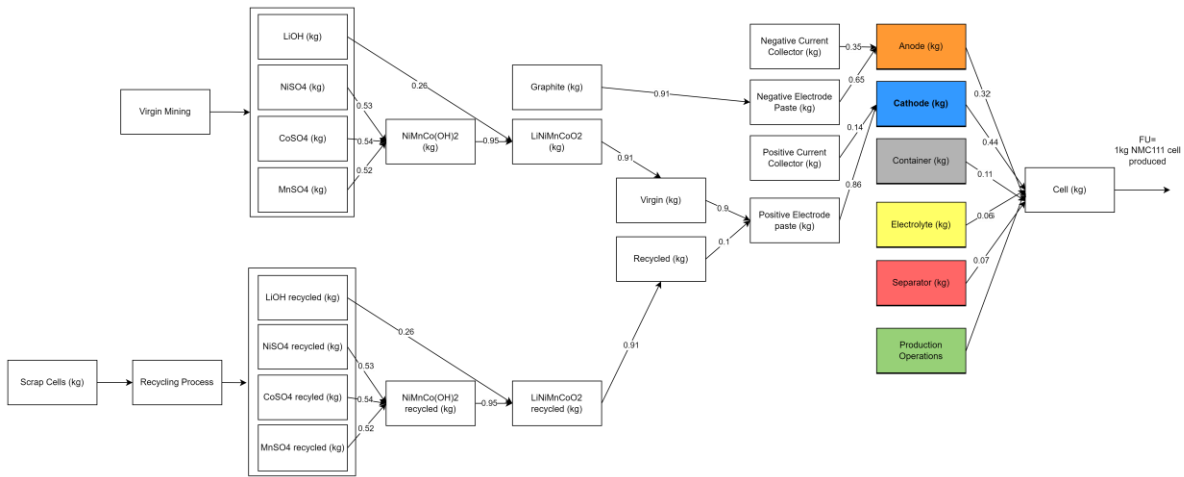
Figure 16- Materials of Interest

The cathode is made of positive electrode paste and the positive current collector made of aluminum foil while the anode is made of negative electrode paste, mainly consisting of graphite, and the negative current collector made of copper foil. The development process of positive active material for positive electrode paste is explained by Majeau-Bettez et al., (2011) as shown in Figure 17. Positive electrode paste is prepared as a binder, additives, solvent, and active material are mixed in the mixing step. Precursor Nickel-Manganese-Cobalt-Hydroxide ($Ni_{0.33}Mn_{0.33}Co_{0.33}(OH)_2$) is created with precipitation of Nickel Sulfate ($NiSO_4$), Cobalt Sulfate ($CoSO_4$) and Magnesium Sulfate ($MnSO_4$). Calcination of $Ni_{0.33}Mn_{0.33}Co_{0.33}(OH)_2$ with Lithium Hydroxide (LiOH) produces active material Lithium-Nickel-Manganese-Cobalt-Oxide ($Li_{0.33}Ni_{0.33}Mn_{0.33}Co_{0.33}O_2$).
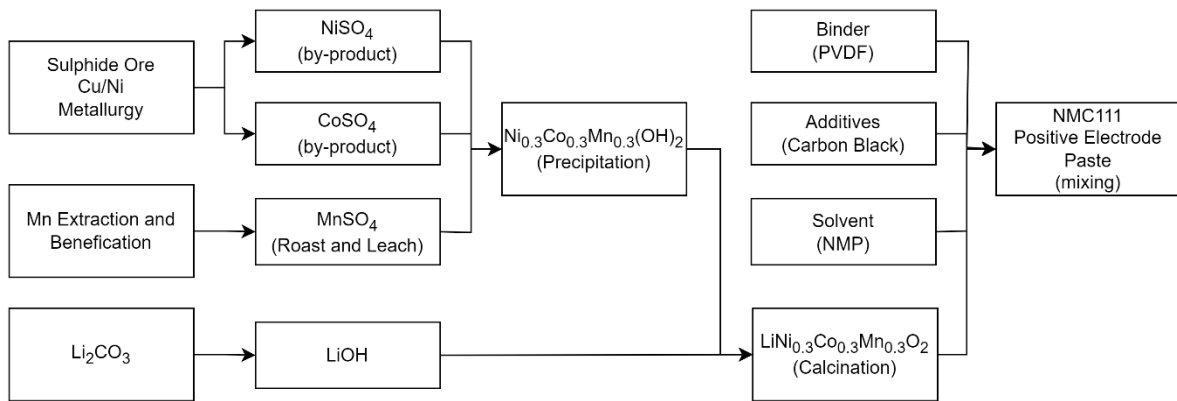
Figure 17- Production steps of NMC111 Electrode Paste

Using the virgin route of production model developed in Figure 16, the complete Life Cycle Inventory of producing 1kg NMC111 cell is given in Table S 1 in Supporting Information.

3.2.3.2 Production with Direct Recycling and LCI
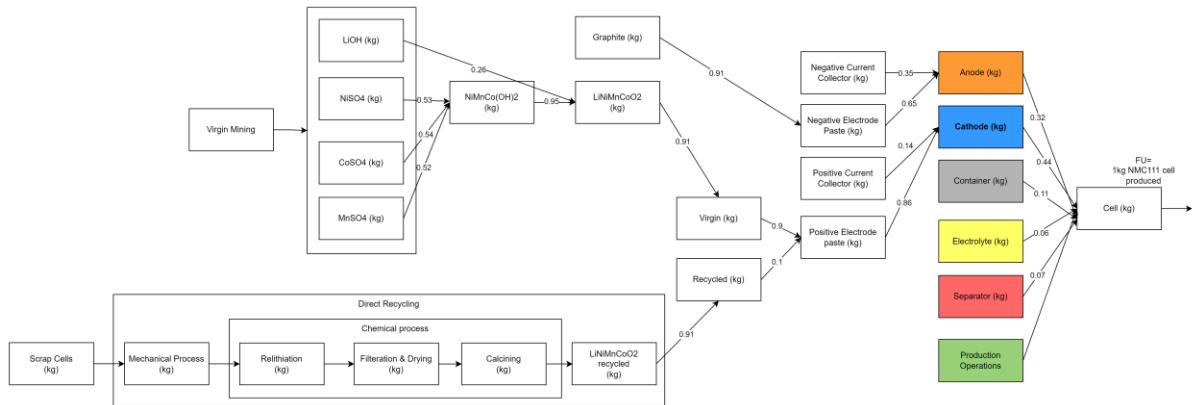
Figure 18- Production Model with Direct Recycling

The direct recycling model is presented in Figure 18, based on Jiang et al., (2022) while the material and energy losses in direct recycling processes are adjusted using waterjet-based direct recycling analysis by Kurz et al., (2021). A generic scenario is presented with 90% virgin and 10% recycled positive electrode paste. Direct recycling, as discussed in the literature, consists of mechanical shredding and disassembly of components to follow the chemical separation process. After mechanical separation, the cell is dismantled, cell container, copper, and aluminum collectors are discarded as scrap while the degraded active material contains positive and negative electrode paste. The output from mechanical shredding and separation enters electrochemical relithiation where lithium hydroxide (LiOH) is added to refill the loss of lithium ions in the degraded active paste. For NMC, the relithiated powder undergoes washing with water to remove residual LiOH after reaction with LiOH for relithiation. The powder is then dried and calcined with lithium carbonate (LiCO$_3$) to create recycled LiNiMnCoO$_2$.

After the generation of recycled cathode active material, the recycling model is combined with the production model, and recycled LiNiMnCoO$_2$ is used to produce positive electrode paste used in the cathode and eventually the cell. Following the allocation approach described in the Mass Allocation-Based Combined Model the recycling processes are attributed to the burden of producing recycled LiNiMnCoO$_2$. The complete LCI of producing LiNiMnCoO$_2$ by 1kg of NMC111 cell through direct recycling is given in Table S 2 in Supporting Information.

3.2.3.3 Production with Hydrometallurgical Recycling and LCI
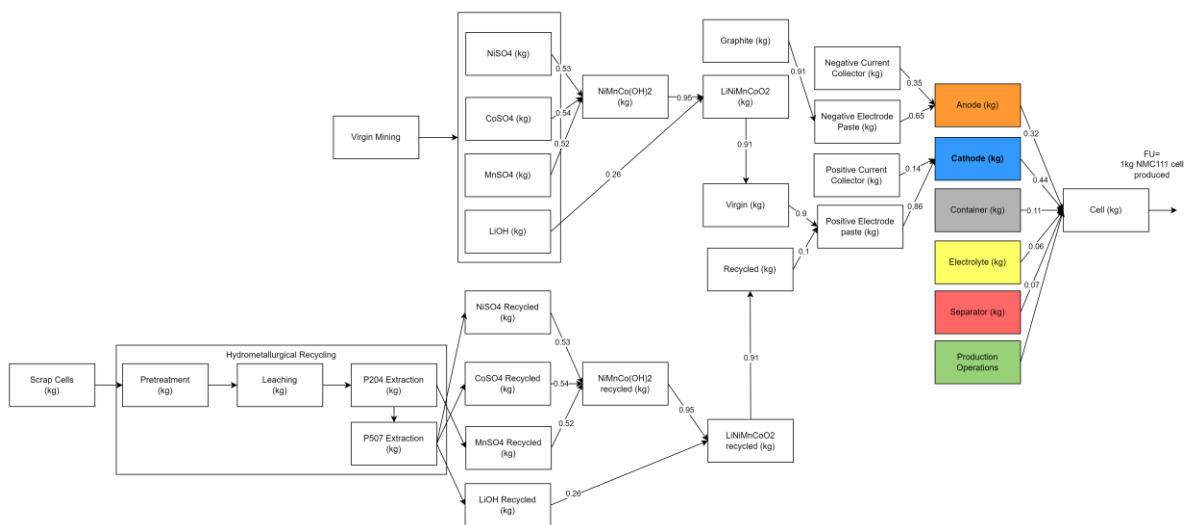
Figure 19- Production model with Hydrometallurgical Recycling

Figure 19 depicts the hydrometallurgical recycling process combined with the cell production model. The hydrometallurgical process commences with pre-treatment, where active cathode materials are separated into powder form. This powder mainly contains nickel, cobalt, lithium, and manganese from positive electrode paste along with copper, aluminum, iron, and graphite impurities from positive current collector, negative current collector, cell container, and negative electrode paste respectively. Scrap metal and plastics are discarded after pre-treatment while the remaining powder is then transferred to the leaching step which is the most chemical-intensive process in hydrometallurgy recycling. During leaching, leaching agents' sulfuric acid, water & hydrogen peroxide and reducing agents, sodium sulfate, and sodium hydroxide are added to dissolve all inputs into a leaching solution and to eliminate impurities of particles from current collector and graphite from anode paste. After leaching, organic impurities are separated, and residual leachate is sent to the first solvent extraction process where P204 extractant and kerosene are added to remove metallic impurities like iron and liberating manganese compound to be reused for resynthesis of cathode precursor. The remaining output from P204 extraction undergoes P507 extraction to separate cobalt and stripping to extract nickel and lithium compounds. Due to the high use of chemicals and water during leaching and extraction, hydrometallurgy releases severely contaminated wastewater after treating cells. Complete LCI of producing $NiSO_4$, $CoSO_4$, $MnSO_4$, and LiOH by 1kg of NMC111 cell through hydrometallurgical recycling is given in Table S 3 in Supporting Information.

3.2.3.4   *Production with Pyrometallurgical Recycling and LCI*
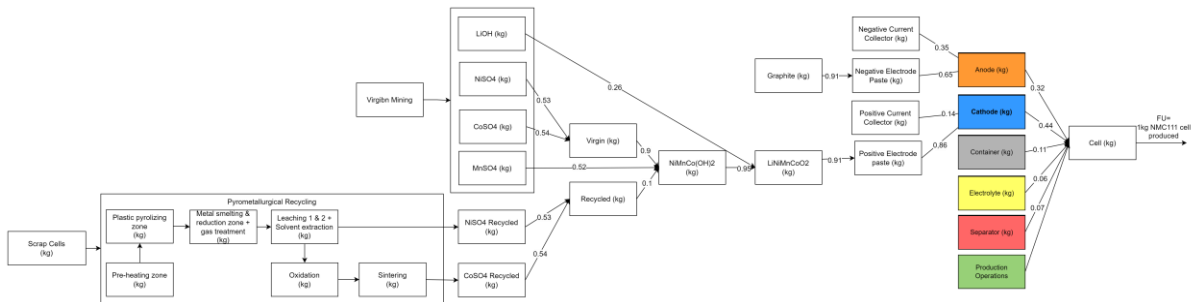
Figure 20- Production model with Pyrometallurgical Recycling

The production model with pyrometallurgical recycling is displayed in Figure 20. The process starts with preheating scrap cells in a blast furnace along with coke and slag forming agents, sand, limestone, and metal slag from steel manufacturing. The slag formed during the preheating process is fed into the metal smelting and reduction zone + gas treatment where slag and metal alloy are separated. Slag is disposed of in landfills while the alloy is treated further with a two-stage leaching process followed by solvent extraction to extract nickel as nickel hydroxide (Ni $(OH)_2$). After extracting nickel, the output from leaching is oxidized using hydrogen peroxide (H_2O_2) followed by sintering in the presence of lithium carbonate (Li_2CO_3) to extract cobalt in the form of Lithium cobalt oxide ($LiCoO_2$). This method is also referred to as pyro-hydrometallurgy where preheating, pyrolysis and smelting is a part of pyrometallurgy while leaching, solvent extraction, oxidation, and sintering are part of the hydrometallurgy. As the study is connecting recycling with the production model, here the model assumes that output nickel and cobalt compound has the same functionality as required to produce a precursor for positive active material production. The complete LCI of producing $NiSO_4$ and $CoSO_4$ by 1kg of NMC111 cell through pyrometallurgical recycling is given in Table S 4 in Supporting Information.

The LCIs of the associated virgin and recycling methodologies discussed above are incorporated in terms of foreground matrix A_{ff}, background to foreground matrix A_{bf}, stressors matrix F_f with the demand vector y in the specified Arda template available to calculate the impacts through the MATLAB program.

4 Results and Analysis

This section presents the life cycle impact assessment results for producing 1kg NMC111 cell based on the ReCiPe midpoint H impact assessment method. Overall, the results represent that cells produced with direct recycling will be the most effective under all impact categories and virgin cathode is the highest contributor in most of the impact categories (Figure 26). Direct and hydrometallurgical recycling has comparable impacts. Only cathode contribution in the overall impact is observed to vary depending on whether it is produced using materials mined traditionally or recycled from scarp cells rejected during quality control procedures. While the cathode in the recycling process- direct, hydrometallurgy, and pyrometallurgy- also contain the respective recycling impact as only $LiMnNiCoO_2$ as an active material in positive electrode paste is being recycled while recycling of other elements of positive electrode pastes and positive current collectors is not considered.

The results along with the contribution analysis of the four impact categories selected in the Goal & Scope section are presented below:

4.1 Contribution Analysis of Production with Virgin and Recycled Cathode

According to Ekvall et al., (2020), LCA is primarily utilized to provide insight into the ecological consequences of different activities and services by revealing their negative effects. Furthermore, it identifies the primary drivers of various environmental impacts, which enables us to mitigate or eradicate their adverse consequences. Additionally, it is also crucial to comprehend the procedures or supply chains that produce these harmful factors. This aspect examines the principal contributors and their associated processes.

4.1.1 Global Warming Potential:

Figure 21 presents the climate change impact breakdown of the cell production using virgin raw materials vs recycled materials for cathode production, where GWP-100 is Global Warming Potential measured over 100 years. The results show that the cathode including positive electrode paste and current collectors contributes about 65% of the total cell's GWP impact made of virgin cathode active materials while recycled cells with direct recycling technology will reduce the impact of cathode production by 90%. It is observed that 64% of the contribution of impact is due to cobalt as a virgin material and with recycled, cobalt its impact reduces to even less than 6% as shown in Figure 22. Furthermore, as the recycling technologies only recycled positive electrode paste and its constituents, it is observed that the cell container that is typically made of aluminum dominates in GWP in recycled cell impact for direct and hydrometallurgical recycling followed by electrode and cell manufacturing operations. However, in pyrometallurgical recycling, the cathode is still observed to be the highest impacting component followed by the cell container.

According to the results, the cathode is a dominant contributor to GWP, SOP, and FFP impacts under virgin and pyrometallurgical recycled cell production. When the contribution of impact is analyzed deeply, it is observed that among active materials, cobalt is the main driver of GWP as shown in Figure 22. According to Farjana et al., (2019), cobalt production requires extensive use of electricity and blasting causing significant environmental impact and emissions, with global warming being the largest impact category. Fossil fuel-based carbon dioxide is the biggest contributor to global warming in cobalt mining. Hence either reducing the cobalt content in NMC cells or reusing the recycled cobalt from the scrap cells is an essential aspect to reduce the climate impact of cell production.
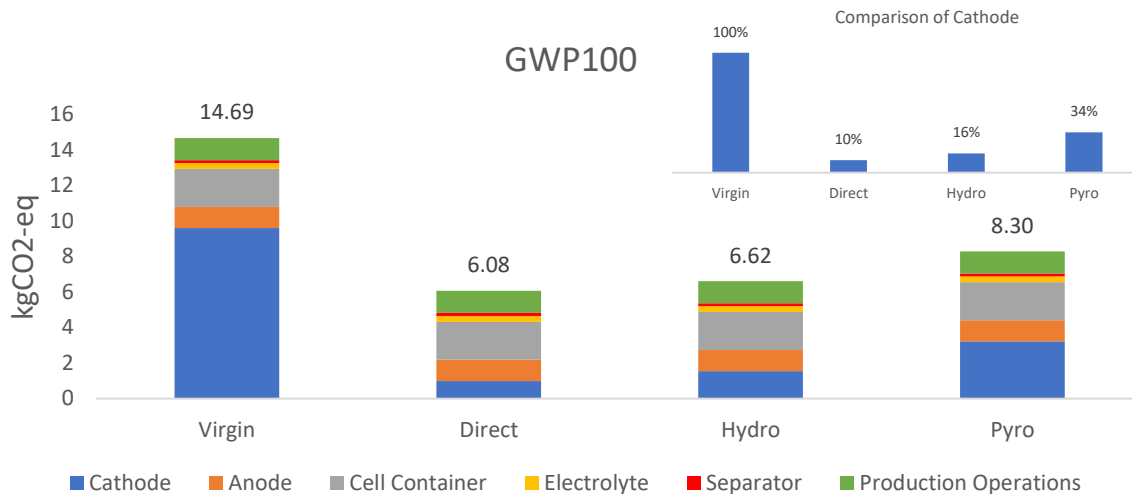
Figure 21- Climate Change Impact Contribution

Furthermore, cell containers are significantly from aluminum due to their lightweight and robust properties, enabling them to endure high temperatures and corrosion. Its excellent conductivity allows for effective heat dissipation, a critical factor in averting battery overheating, which could pose a safety risk or lead to damage. Additionally, aluminum's recyclability and minimal ecological footprint make it an eco-friendly option for battery manufacturing. However, according to the *World Economic Forum*, (2022), the production of aluminum necessitates energy-intensive methods for extracting alumina from bauxite and converting it into aluminum, with over 70% of the energy derived from fossil fuels, for powering smelters (for primary aluminum) and electric induction furnaces (for recycled aluminum). 58.9% of the total world's primary aluminum output is generated in China (International Aluminium Institute, 2023). The aluminum production industry contributes to about 2% of all man-made emissions, making it one of the most emission-intensive industrial materials, with seven times the emissions of steel (*World Economic Forum*, 2022). Therefore high GWP under the cell container is due to the direct CO_2 emissions, accounting for more than 90% of the total, caused by alumina refining and aluminum smelting in the production of aluminum (*Aluminum Tracking Report*, 2022). There exist various low-emission alternatives to aluminum for cell containers in batteries. For instance, lightweight and eco-friendly plastic materials like polycarbonate or polypropylene are practical options (SAE, 2023). Alternatives material for cell containers, like carbon fiber-reinforced plastic (CFRP) and glass fiber-reinforced plastic (GFRP) components offer excellent mechanical properties, such as high stiffness and strength, coupled with low weight (Altin Karataş & Gökkaya, 2018). An instance of this is the battery case manufactured from CFRP, which has the potential to reduce weight by up to 40% compared to aluminum or steel (*Battery Cases for Electric Vehicles*, 2019). Other potential substitutes include materials such as titanium, known for its high strength and low density *(Britannica*, 2023), or stainless steel, which is corrosion-resistant and durable. Although several alternatives are available, the selection is dependent on certain factors like cost, performance, market availability, and recyclability of materials.
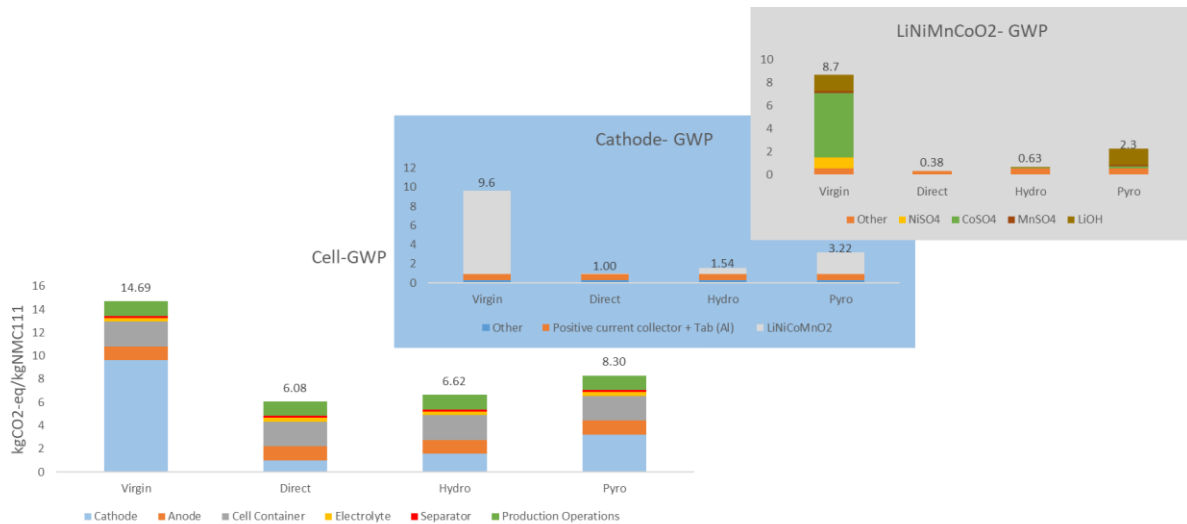
Figure 22- GWP-100 Impact Contribution of Active Materials

4.1.2   Particulate Matter Formation Potential:

In particulate matter formation, 54% of the impact is due to the virgin cathode followed by the anode while in the case of recycled cells, the anode is the most significant contributor, contributing even more than half of the total impact as shown in Figure 23. Cell operations and electrolytes have negligible impact. This implies that the active materials used in both cathode and anode manufacturing are the main contributors to particulate matter formation.
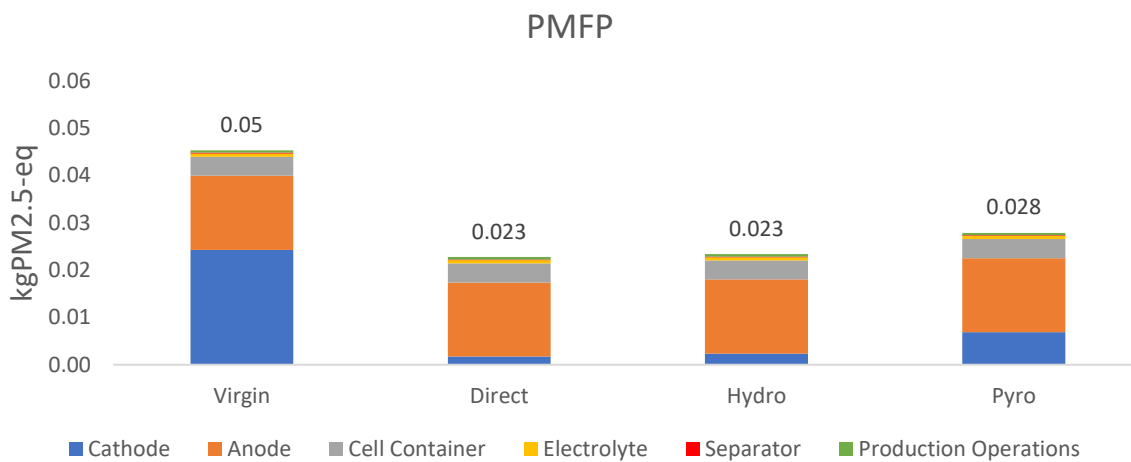
Figure 23- Particulate Matter Formation Potential (PMFP) Contribution

Particulate matter formation dominates in both cathode and anode manufacturing. Whereas the high impact of PMFP in the virgin cathode is justified by the high mass of the cathode relative to the anode. In cathode, PMFP dominates due to cobalt and nickel mining, while in the anode due to copper current collector and tab. Cobalt has relatively high $PM_{2.5}$ emissions compared to nickel as explained by Dai et al., (2019), high PM emissions from $CoSO_4$ production can be attributed to specific cobalt ore mining practices in Congo. Low-grade ores that are excavated are usually kept stockpiled until all the high-grade ores in the deposit are extracted and processed, which can take up to 20-30 years. Due to wind erosion over such a lengthy period, the stockpile produces significant amounts of PM emissions. In addition, the transportation of diesel and reagents required for mining and processing copper-cobalt

ore in Congo involves importing them and hauling them by trucks over long distances, primarily on dirt roads, which also leads to significant PM emissions (*Argonne GREET® Model*, 2018).

While for a current collector, copper extraction involves mining, crushing, milling, ore concentration, smelting, refining, and waste management, which generate by-products contaminated with metals and metalloids, including toxic ones such as As, Cd, Hg, and Pb (Izydorczyk et al., 2021). Smelting releases large volumes of SO_2 and solid particles containing high amounts of heavy metals such as As, Cd, and Pb (Serbula et al., 2013). Dust with small particle sizes can remain in the air indefinitely, and fine fractions of metallurgical dust containing toxic metals (As, Pb) can cause human health and environmental problems (Izydorczyk et al., 2021). NMC111 cells commonly employ copper foil and tab as the negative current collector. Nevertheless, researchers are currently investigating alternative current collector materials such as carbonaceous materials, nickel, titanium, and conductive polymers to enhance the performance and safety of these cells. Zhu et al., (2020) compared several materials commonly used as current collectors in lithium-ion batteries, based on electrochemical stability, electrical conductivity, mechanical property, density, and sustainability. The use of carbonaceous materials as current collectors in lithium-ion batteries is promising but problematic due to low weldability (Pan et al., 2022). Polymers have the potential as current collectors due to their lower density and better corrosion resistance, but their low electrical conductivity and thermal stability need improvement (Augustyn et al., 2021). Furthermore, recycling or reusing conventional current collectors can reduce the PMFP impact and provide a secondary source of valuable materials, but their recyclability and reusability need to be investigated further.

4.1.3   Surplus Ore Potential:

Cathode being highly rich in scarce materials contributes to about 84% of the surplus ore potential as presented in Figure 24. However, unlike other impact categories, if the cathode materials are recovered from recycling, the electrolyte becomes the second highest contributor except for pyrometallurgical recycling which does not recover lithium and magnesium from the scrap cell. Also analyzing the factor contributing most to electrolyte, it is observed that rare earth element mine operation and beneficiation from bastnaesite and monazite ore contributes about 70% of the impact in direct and hydrometallurgically recycled cathode and spodumene production contributes about 70% of the impact in pyrometallurgically recycled cathode. Spodumene is an important ore of lithium and a source of ceramic materials.
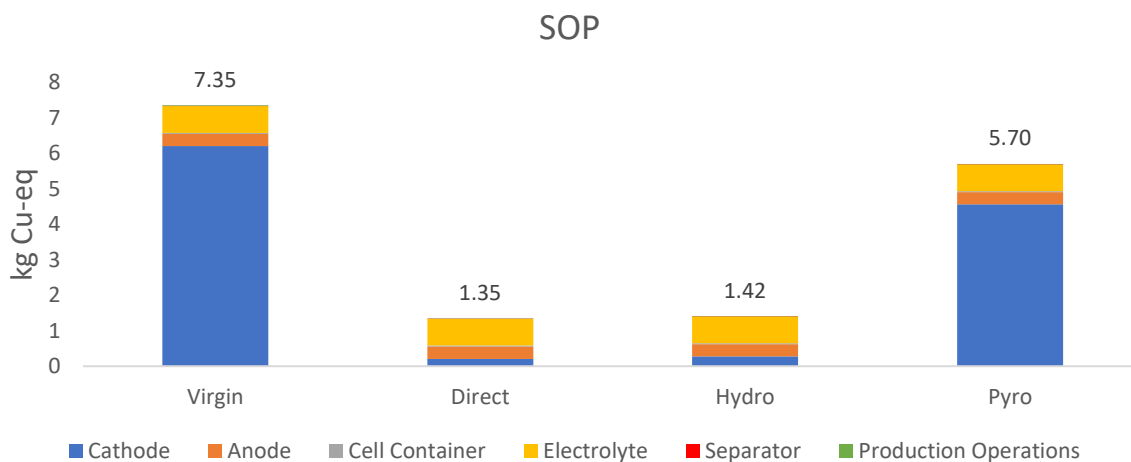
Figure 24- Surplus Ore Potential (SOP) Contribution

As the cathode and its components constitute about 44% of the total cell mass (Table S 1), the SOP for the cathode is also found to be higher in both virgin and pyrometallurgical recycled cathodes. Furthermore, the contents of positive electrode paste specifically $CoSO_4$ and $LiOH$ have a high contribution to SOP due to the continued extraction of these materials for meeting the demands of batteries (Searcey et al., 2021; Greim et al., 2020). As explained in Impact Categories, SOP measures the loss of resource by calculating the additional amount of ore needs to be mined per unit of resource extracted in the future. Similarly higher SOP impact was observed in pyrometallurgical recycled cathode due to the inability of pyrometallurgy recycling technology to replace lithium compound. Moreover, pyrometallurgy required additional lithium carbonate Li_2CO_3 to extract cobalt in the form of Lithium cobalt oxide $LiCoO_2$.

However, if the cathode is produced with directly recycled or hydrometallurgically recycled materials, the SOP impact is found to be increased under the electrolyte component rather than the anode. While investigating the composition of the electrolyte used in this study, electrolyte Lithium Hexafluorophosphate ($LiPF_6$) used with ethyl carbonate (EC) and dimethyl carbonate (DMC), is commonly utilized due to its thermal stability in solvents (J. Jiang & Dahn, 2003; Z. Zeng et al., 2014; F. Zhou et al., 2009). $LiPF_6$ is mainly synthesized through the reaction between hydrogen fluoride (HF) and phosphorus trichloride (PCl_3) (Boll et al., 2015; Susarla & Ahmed, 2019). HF is primarily manufactured by the reaction between fluorspar (CaF_2) and sulfuric acid (H_2SO_4) (Dahlke et al., 2016) whereas CaF_2 is produced by the reaction of calcium with rare earth fluorides (*Britannica*, 2023). Mining operations and beneficiation of rare earth elements from bastnaesite and monazite ore (Behrsing et al., 2014) required in the production of electrolyte $LiPF_6$ is, therefore, a strong candidate for SOP. While researchers have been trying to develop new electrolytes for lithium-ion batteries, however, none of the new salts have offered significant advantages over the industry standard $LiPF_6$ salt, which continues to be the predominant salt used for most commercial Li-ion batteries (Henderson, 2014). Furthermore, the suitability of any electrolyte depends on the trade-offs between different properties and the specific application requirements.

4.1.4 Fossil Fuel Potential:

The virgin cathode contributes to about 67% of fossil fuel potential (Figure 25) as the constituents of positive electrode paste are produced in various parts of the world using a high share of fossil energy in their total energy mix. Similarly, cell container 80% of which is composed of aluminum dominates the fossil fuel potential in direct and hydrometallurgical recycling where cathode components are recycled adequately.

Under fossil fuel potential (FFP), virgin cathode is the highest contributor followed by cell container and anode. Extraction of hard coal from mines to energize the systems extracting battery active materials and aluminum for a cell container contributes to about 20% of the FFP. Also, Yudhistira et al., (2022) compared the different chemistries of lithium-ion and lead-acid batteries for grid energy storage using Product Environmental Footprint Categorical Rules (PEFCR) and observed in the case of battery cells, the most significant fossil resource use impact is attributed to the manufacturing of electrodes, which applies to both cathodes and anodes in the case of lithium-ion batteries. Whereas high fossil fuel potential under a cell container is justified due to the indispensable usage of fossil fuels in both alumina refining and recycled aluminum production (*Aluminum Tracking Report*, 2022). As FFP and GWP are strongly correlated, the impact contribution is also found similar in both indicators. Likewise, reducing virgin cobalt extraction and aluminum in cell containers can be a potential aspect of FFP reduction.

FFP
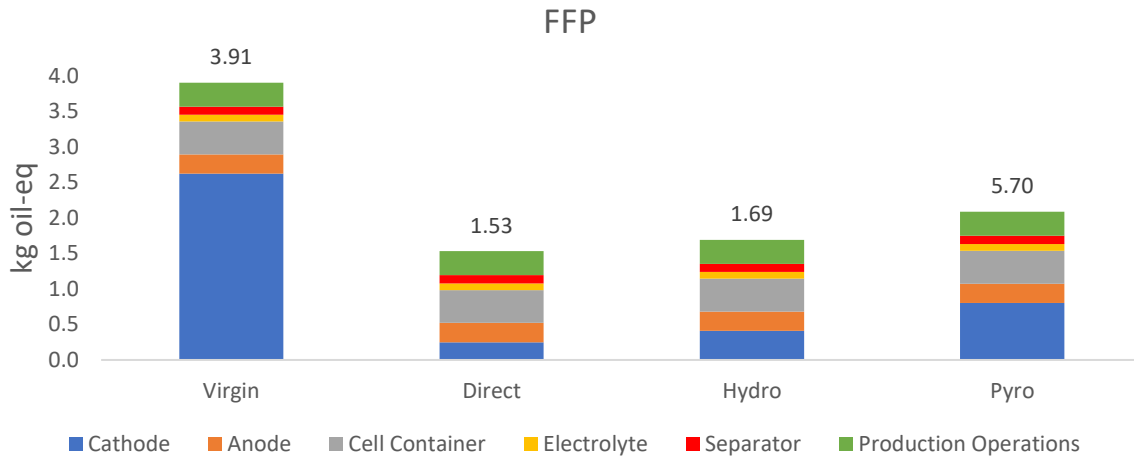
Figure 25- Fossil Fuel Potential (FFP) Contribution

Figure 26 represents the impact contribution map of all 18 ReCiPe midpoint indicators- Impact contribution of all 18 ReCiPe midpoint indicators- freshwater ecotoxicity potential (FETP), marine ecotoxicity potential (METP), terrestrial ecotoxicity potential (TETP), human toxicity potential (HTPnc), photochemical oxidant formation potential: humans (HOFP), photochemical oxidant formation potential: ecosystems (EOFP), human toxicity potential (HTPc), ionising radiation potential (IRP), terrestrial acidification potential (TAP), particulate matter formation potential (PMFP), global warming potential (GWP100), ozone depletion potential (ODPinfinite), water consumption potential (WCP), surplus ore potential (SOP), fossil fuel potential (FFP), agricultural land occupation (LOP), marine eutrophication potential (MEP), freshwater eutrophication potential (FEP). Besides the cathode, the anode is observed to be another potential contributor in these impact categories. Under all impact categories, the magnitude of impact follows the condition $d_{direct} < d_{hydro} < d_{pyro} < d_{virgin}$ where d refers to the total impact under all impact categories.

Impact Contribution of all 18 ReCiPe Indicators
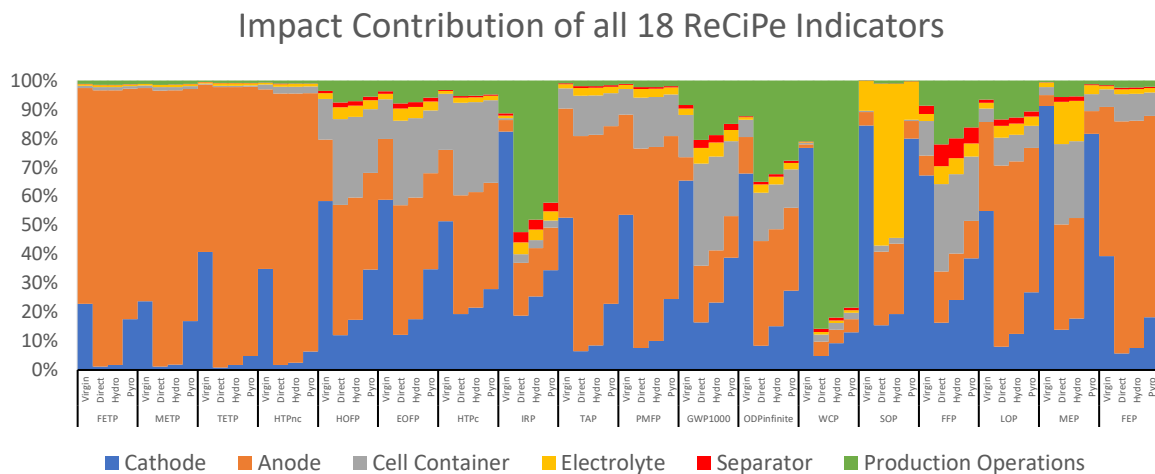
Figure 26- Impact contribution of all 18 ReCiPe midpoint indicators

4.2   Sensitivity Analysis

Sensitivity Analysis (SA) involves examining the correlation and impact of the inputs on the outputs of a system and addresses how changes in the inputs affect the overall outcome of a system (Razavi et

al., 2021). The results are further analyzed on two conditions that may significantly affect the impact results. Only climate change impacts are selected for this analysis as described below:

4.2.1 Changing the Recycled Content:

Given the current statistics by CAS Science Team, (2022), less than 5% of the spent batteries are being recycled, therefore, it is not easy to acquire 100% of the recycled contents in cell production. Therefore, it is necessary to identify the potential impacts of the combined virgin and recycled cathode under varying percentages as shown in Figure 27. It is notable that the impact due to cells produced with even 100% of pyrometallurgical recycled cathode material is greater than the cell produced with cathode recovered with 90% direct or hydrometallurgical recycling that makes direct and hydrometallurgical recycling preferable to pyrometallurgy recycling technology. Figure 28 represents the relative GWP impact while changing the contribution of input from virgin and recycled routes as modeled in the Case and Methodology. Impact reduces as the recycled content is increased, however, using 10% recycled cathode reduces the impact ranging between 4-6%, and 100% recycled cathode reduces the impact ranging between 53-59% depending on the recycling technology used. This indicates that even producing cells with 100% recycled contents of positive electrode paste reduces nearly half the impact of cell production.
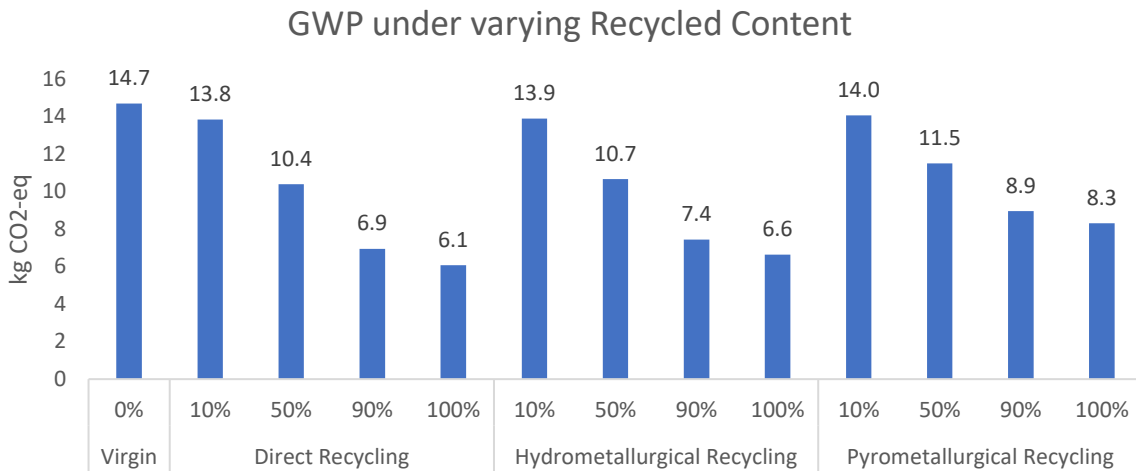
Figure 27- GWP under varying recycled content
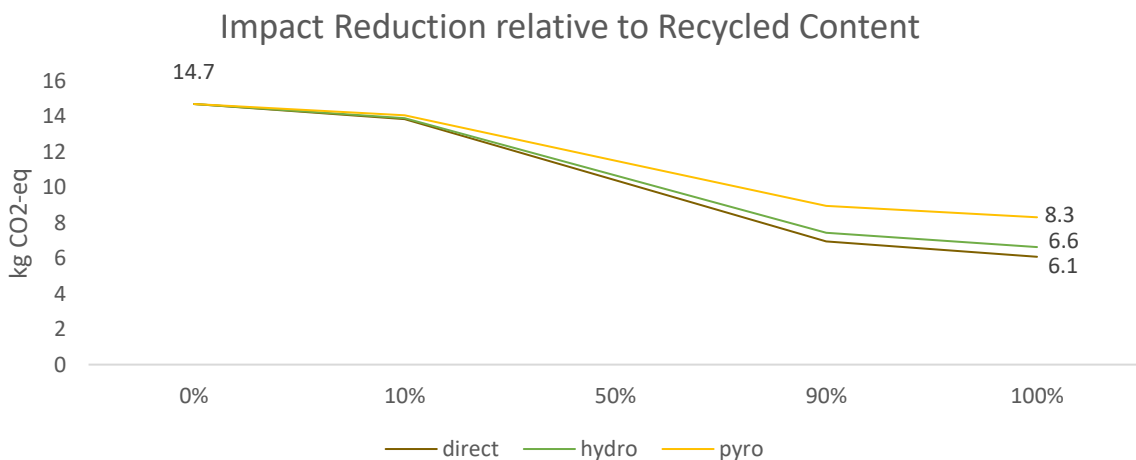
Figure 28- Change in Impact relative to the Recycled Content

The impact of producing 1kg NMC111 cell by the combining of production and recycling model vary depending on the recycling technology. The general condition of the impact $d_{direct} < d_{hydro} < d_{pyro} < d_{virgin}$ still seems to be valid. The impact of pyrometallurgy recycling is relatively higher than other recycling methodologies as it demands extensive heat energy, for example, in sintering for the recovery of cobalt from metal alloy.

Despite the fact that pyrometallurgy has drawbacks such as high energy consumption, harmful emissions, and the inability to extract lithium, it is the most commonly used technique to recover metals at an industrial scale as it is more versatile and simpler (Latini et al., 2022; Mossali et al., 2020). From an environmental and resource perspective, numerous studies have suggested that hydrometallurgy is a more appropriate recycling technique than pyrometallurgy (Ganesh et al., 2021; Vieceli et al., 2021) due to its ability to recover a greater proportion of elements while consuming less energy, generating significantly lower greenhouse gas emissions, producing minimal air emissions, and enabling purification. However, it is a complex process that is heavily influenced by cathode chemistry, making it difficult to implement on an industrial scale. Furthermore, direct recycling of lithium-ion batteries besides being ecofriendly, can also be affected by various factors, including battery sorting, extensive pre-treatment, the accumulation of defects and impurities in the active materials, variations in cell composition, and battery state-of-health (Larouche et al., 2020). Additionally, since it was developed for specific types of batteries, direct recycling may be more susceptible to market fluctuations and the emergence of new battery chemistries.

The GWP impact from either direct and hydrometallurgy are quite comparable which means that implementing either of the technologies will be almost equally beneficial and influence in a similar way in terms of climate change.

4.2.2 Changing Geographical Location:

Another sensitive factor to the results calculated is the geographical location. Regionalized LCIA and relevant LCIs give close-to-reality results (Frischknecht et al., 2019). High spatial resolution minimizes uncertainties and differentiates when accessing the impacts of emissions on various locations (Manneh et al., 2010). Regionalization can be applied at both LCI and the LCIA levels. Due to the unavailability of specific regionalized LCIs, the variation in results is analyzed by changing the input sources of energy and transport geographically as electricity mix and transportation services vary significantly from country to country. The spatially differentiated impact of producing the same cell with 100% recycled cathode is presented in Figure 29. Here the impact is following the general condition $d_{Norway} < d_{Germany} < d_{China}$, that is, if the same cell is produced using Chinese electricity, heat, and transport, the impact is higher than otherwise if it is produced in Germany and Norway. It is worth noticing that production with virgin cathode material in Norway is better than if the cell is produced with 100% recycled cathode in China and comparable to the pyrometallurgical recycled cathode in Germany.

The results of sensitivity analysis indicate that certain assumptions can significantly impact the outcomes, such as the assumed energy mix and geographical region. Producing the same NMC111 cell in Europe would have likely resulted in a lower CO_2 impact, because Europe has a less carbon-intensive average energy mix, mainly due to a greater proportion of hydro and nuclear power (Melin, 2019). Conversely, manufacturing the battery in China would have increased its CO_2 impact due to a higher proportion of coal in the energy mix (*Global Electricity Review 2023*, 2023; Ritchie et al., 2022). Thus, the electricity mix is a crucial factor in determining the environmental impact of NMC111 cell production with either virgin or recycled material. The carbon footprint of lithium-ion batteries during their entire life cycle stages is relatively small in comparison to the production stage, with the

electricity mix at the production site being the main factor affecting the overall environmental impact (Costa et al., 2021) and varies significantly among countries.
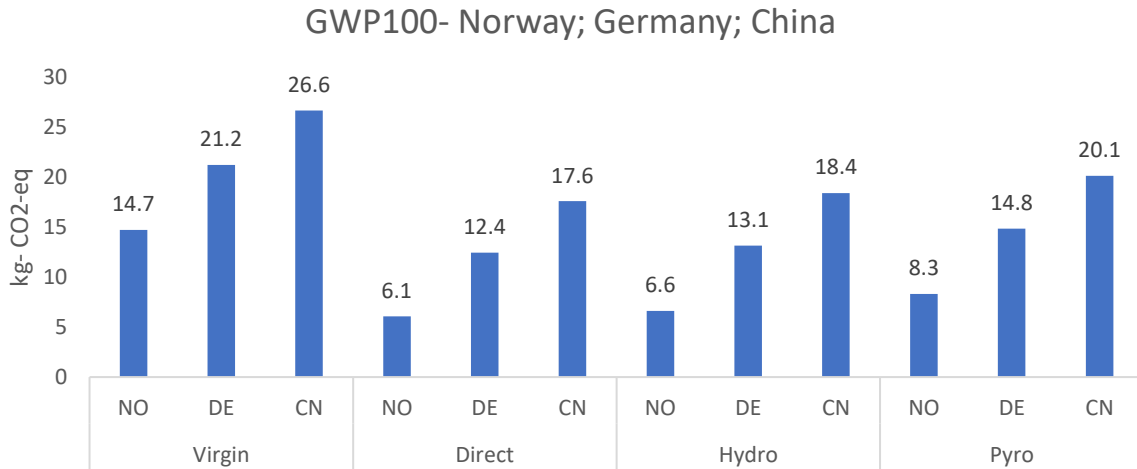
Figure 29- GWP of 1kg recycled cathode produced in Norway (NO), Germany (DE) and China (CN)

The benefits of recycling lithium-ion cells can be maximized using renewable energy sources during the recycling process. Therefore, the combination of recycling and decarbonization is essential for a sustainable circular economy to mitigate climate change. Recycling reduces the environmental impacts of new cell production by reutilizing cell scrap, but it must be powered by renewable energy to minimize greenhouse gas emissions. Decarbonization eliminates greenhouse gas emissions from human activities to mitigate climate change's effects. Therefore, recycling strategies discussed will be more beneficial if implemented in line with the decarbonization of energy sources to reduce greenhouse gas emissions, conserve natural resources, and reduce environmental impacts associated with waste and resource use.

4.3   Taylor Series Analysis
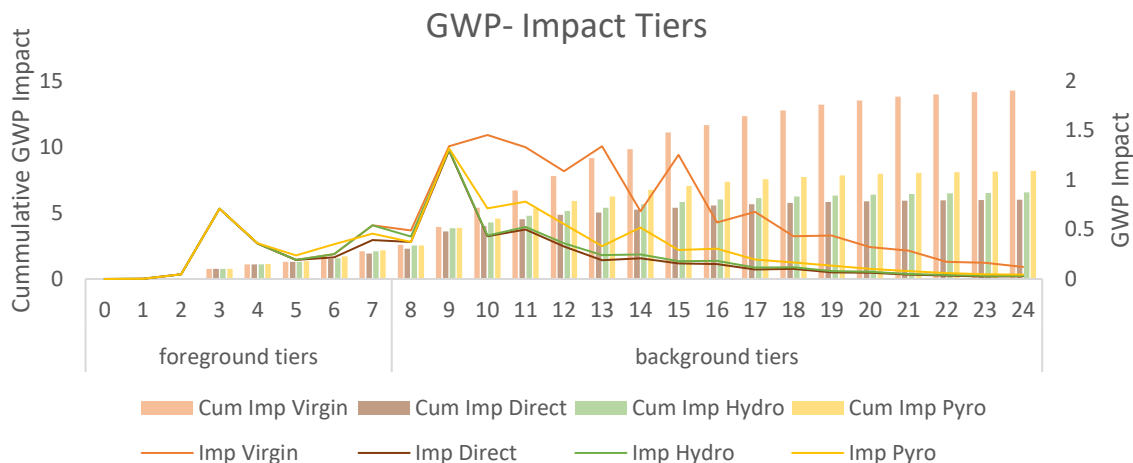
Figure 30- GWP Impact along the Production tiers

Figure 30 represents the impact in individual tier of cell production with line graph represented by *'imp'* and cumulative impact with bar graph represented by *'cum'*. The cumulative impact represents how impact adds up from foreground to background tiers for producing 1kg NMC111 cell. It is evident

that the impact regardless of the production is nearly similar until the 9th tier where the 10th tier represents the first marked difference among the production of the cell using vs recycled cathode. This difference in impact is due to the energy inputs required in the background to produce lithium and manganese in cells produced with pyrometallurgical recycled cathode, and further cobalt and nickel in a cell produced with virgin cathode. Thus, cell production impact is dominated by the background energy requirements for extracting active materials while in the case of recycled active materials, the impact can be reduced significantly (about 70% in the case of hydrometallurgy and direct recycling and about 51% in the case of pyrometallurgical recycling) eliminating the need of excess energy required for mining and refining. As this study only considered the recycling of cathode active elements, the impact can be further reduced if all the components of the cell can be recycled thus eliminating the need for raw mining. The results also indicate even if recycling is an important aspect of reducing GWP impact; it alone is not the complete solution to climate change. The recycling process itself can have environmental consequences, and it is not enough to meet the increasing demand for battery systems (kbaker, 2022; *Fraunhofer ISI*, 2023; Reid, 2022). Additionally, the quality of the recycled material to be reused in the production of new NMC111 cells is another factor that needs further investigation. Therefore, to address the underlying causes of climate change holistically, recycling strategies discussed in this study should also be accompanied by other environmental initiatives in practice.

5   Discussion

In this section, the implications of the results, limitations of this study and future perspectives are discussed. The results are then compared with the previous literature available. The objective of the study was to combine the traditional production model with several recycling methodologies and calculate the impact under varying recycling content and specific geographical parameters like electricity, heat, and transport.

The results indicate direct recycling is the most preferred recycling method in terms of environmental impact. However, this technology is just been implemented on a lab scale (Bai, Muralidharan, Li, et al., 2020; Bai, Muralidharan, Sun, et al., 2020; Gaines & Wang, 2021; J. Li et al., 2020; Yu, Li, et al., 2022) therefore, its industrial scale implications are not totally clarified yet. Whereas hydrometallurgy and pyrometallurgy are already commercialized and mature, however, their ecological values are not that ideal due to chemically intensive wastewater in hydrometallurgy and smelting emissions in pyrometallurgy (M. Chen et al., 2019). There is continued research demonstrating the positive potential of the direct recycling process on a commercial scale (Gupta et al., 2023; Sloop et al., 2020; Yu, Yu, et al., 2022) that could shortly bring this technology into practice safely by recycling investors.

Upstream material availability and procurement issues are additional factors motivating the recycling industry. In addition to environmental, artisanal cobalt mining is associated with high resource criticality, geopolitical risks, and social injustice (Campbell, 2020). However there is no geological scarcity of lithium reserves (Speirs et al., 2014), its continued extraction will be costly in future (Kesler et al., 2012) and achieving required quality for battery grade materials will be a constraint for continued growth of EV market (Narins, 2017). Additionally, the use of fossil resources for these metallurgical processes not only depletes fossil resources, but also generates extensive GHG and PM emissions.

The allocation-based model developed in this study has been able to produce convincing results. It is observed that generally high impact is associated with virgin cathode components due to their high mass and valuable contents. Further, as the recycling content is increased the impact is shifted from the cathode to other components depending on the impact category under analysis which assures that even if the cathode is recycled there are other components of the cell that will be dominating in total production impact. Also, as the recycling content is increased the impact is decreased making it obvious that recycling energy and resource utilization is less than the utilization for extracting virgin resources in the case of cell production. Also, the results vary depending on the recycling technology being used, which justifies the distinction in resource utilization of each technology evaluated in this study. Furthermore, the sensitivity analysis justified the dependence of GWP impact on the intensity of clean energy use.

The expected rise in the demand for lithium-ion batteries will accelerate production, and so does the amount of waste or rejected scrap cells after QC checks during manufacturing. Recycling these rejected batteries can provide a sustainable solution to waste management, especially as the number of giga-cell production factories commence operations. The various recycling strategies discussed in this study can recover valuable materials such as lithium, cobalt, and nickel, reducing the need for new mining activities and contributing to the circular economy. Besides conserving these resources, recycling also reduces the amount of waste generated during production that, otherwise, ends up in landfills, which is critical for achieving sustainability. The tangible benefits from recycling as indicated by the results are the direct reduction of greenhouse gas emissions, whereas indirectly recycling facilities will also generate jobs and revenue.

However, the results obtained from this study support recycling from an environmental perspective, implementing recycling practices are not easy as it will require a huge supply of scrap cells. Scrap cells recycling analyzed in this study constitutes a smaller proportion of the expected total secondary cell supply. EV Stock Channel, (2020) reported scrap generation to be about 5 to 10% depending on the scale of cell production facility. Therefore, the scrap rate from production is quite dynamic and difficult to measure. However, a lot more of the secondary cell supply is expected when the mass produced cells dispatching today will be returning back at the end of life (Gaines et al., 2021). With this study, using scrap cells, an idea is captured that the investment in recycling research and facility will be worthwhile in coming years.

In order to realize the benefits of recycling, industrial and political actors have a crucial role. From an industrial side, cells that are designed considering the ease of disassembly and eliminating irreversible disintegration, maintain the component integrity for reuse and recycling efficiently (L. Thompson et al., 2020). On the other hand, government policies, and regulation has played a vital role in recycling lead-acid batteries achieving about 90% of the recycling rate globally (*Report Linker, 2021*). Enforcing optimal policies can result in multiple benefits to industries, government, society, and the environment. First, standards can be established, and targets may be set for the use of recycled materials in new cell production. Thus, creating a demand for recycled material which incentivize industries to invest in recycling facilities. Second, the government can also support normalizing these facilities by funding their research and construction. This helps in overcoming technical challenges and increasing the recycling process's efficiency. Thirdly, the government can promote awareness of the benefits of battery recycling among the public and encourage participation in recycling programs. Finally, product-related instruments like environmentally related product taxes, and tax differentiation according to OECD (2015) play a significant role in facilitating the recycling of lithium-ion batteries.

Implementing environmental product taxes on the sale of lithium-ion batteries can act as a deterrent, discouraging the production and consumption of batteries that harm the environment. These taxes are designed to consider the environmental impact of the batteries, creating financial disincentives for their use and encouraging the adoption of more eco-friendly alternatives. Additionally, they can generate revenue to support environmental clean-up initiatives and fund the development of advanced recycling technologies for lithium-ion batteries. Tax differentiation involves modifying existing tax structures to align with environmental objectives. In the context of lithium-ion batteries, this entails adjusting tax rates based on their environmental characteristics. In this way, batteries with high recyclability and minimal hazardous components could be subject to lower tax rates and incentivize the production and consumption of environmentally friendly batteries while discouraging the use of less sustainable options. These instruments incentivize the recycling of production scrap itself. Additionally, post-consumer scrap recycling can also be incentivized by Deposit-Refund Systems (DRS) and Extended Producer Responsibility (EPR) as described by (OECD, 2015). DRS encourages consumers to return the batteries they buy by taking a small deposit which will be refunded only if the owner returns the product back. Thus, motivating consumers to properly manage and dispose of batteries. While EPR makes sure that battery manufacturers have to collect and recycle batteries and holds manufacturers accountable for safely disposing and recycling their products. By adopting these product-related instruments, policymakers can establish economic incentives and regulatory frameworks that promote the efficient and sustainable recycling of lithium-ion batteries. Such measures facilitate the transition to a circular economy, mitigate environmental impacts, and conserve valuable resources for future use. Hence, government policies are crucial for creating a sustainable and environmentally friendly energy storage system.

Although the combined virgin and recycled model presented in this study suggests recycling can be an effective solution for reducing greenhouse gas emissions and preserving natural resources, it is not the only solution to climate change. As also observed in sensitivity analysis where production of lithium cells from recycled material in China is found to be even more GWP intensive than the production of cells from virgin raw material in Norway. Several factors contribute to the higher emission intensity of recycling in China compared to the virgin production of lithium-ion cells in Norway. The extraction of valuable metals from recycled cells requires significant energy input, often via high-temperature furnaces. However, in China, about 62% of the electricity is derived from coal-fired power plants (*Global Electricity Review, 2023*; Ritchie et al., 2022), which emit about 1kg/kWh of greenhouse gases over their lifetime that can offset the environmental benefits of recycling. Moreover, recycling facilities in China often lack the technology to effectively capture and treat hazardous waste generated during the recycling process (Wei & Liu, 2012). This can result in toxic substances escaping into the environment and contributing to pollution. Another factor is the high demand for manual labor (South China Morning Post, 2023) due to low labor costs in China (US Bureau of Labor Statistics, 2017), which can result in poor working conditions, as well as increased environmental and health risks from exposure to hazardous waste. In contrast, Norway's production of lithium-ion cells is relatively low-emission, with renewable hydroelectricity powering the energy-intensive production process (*Business Norway, 2023*). In conclusion, while recycling can offer environmental benefits, it is crucial to consider country-specific circumstances and methods. China's reliance on coal-fired power plants, lack of hazardous waste treatment technology, and poor working conditions contribute to higher emission intensity compared to Norway's lithium-ion cell production. The use of net zero emission technologies like Carbon Capture and Storage (CCS) in Chinese existing electricity infrastructure (Q. Chen et al., 2022) in line with the transition to renewable energy can be useful for decarbonization. The Chinese battery industry is expected to reach approximately 1.5 TWh by 2030 (Yuan et al., 2021), dominating the world energy supply. Therefore, the adoption of sustainable and environmentally friendly practices is essential for effective recycling operations.

In addition to the recycling of lithium-ion batteries, various strategies can contribute significantly to addressing climate change like reducing overall energy consumption through enhanced energy efficiency measures or production process innovations such as *24M^{TM} Technologies* (MIT, 2022), simplifying the cell production process with its SemiSolid cell design reducing production cost up to 40%, and development of hybrid cell with graphite electrodes (*Power Japan Plus*, 2014) with a longer functional lifetime. Advancements in manufacturing process and chemical composition of the cells is anticipated by academic and industrial experts depending on factors of interest like cost, energy density and (Few et al., 2018). Cobalt free alternatives are expected where energy density is not a high priority that has a potential for reducing the environmental impacts from cobalt mining.

In addition to renewable energy sources, utilization of energy-efficient technologies to power battery production and recycling processes, also plays a pivotal role in achieving energy conservation objectives. When considering the recycling of lithium-ion batteries, it is imperative to prioritize sustainable and environmentally conscious practices. This necessitates the development and implementation of advanced recycling technologies capable of minimizing the utilization of hazardous chemicals and reducing waste generation. Adhering to environmentally responsible recycling processes is essential to safeguard ecosystems and mitigate further environmental degradation.

It is also crucial to recognize that there are inherent limitations associated with the recycling of lithium-ion batteries. Specifically, challenges arise from recycling inefficiencies related to certain constituents such as graphite (Rey et al., 2021) and electrolytes (Zhang et al., 2022). These limitations underscore the necessity for continuous scientific research and innovation in the field of battery recycling to

identify superior approaches, overcome existing hurdles, and enhance the overall sustainability of the recycling process. The growing demand for lithium-ion batteries in the automotive and energy storage industries (*IEA, 2021*) also poses challenges to recycling efforts as it generates more waste that requires significant resources and infrastructure to manage. Sustainable and environmentally friendly recycling methods must be implemented in line with measures such as reducing energy consumption and transitioning to renewable energy sources to effectively combat climate change.

5.1   Assumptions and Limitations

However, the model proves beneficial in combining the virgin and recycled production chains, there are also some assumptions implied to derive these results that may affect the quality of the results obtained. Also, the LCI datapoint gathered in this study are taken from the previous publications. Therefore, the study is purely based on secondary data due to inaccessibility of primary resources. Hence, there is some uncertainty associated with the results due to reliance on secondary sources.

The allocation-based combined model used in this study may have some limitations as using mass-based allocation assume a similar impact for all products based on mass recovered from the recycling process, regardless of the fact if the product recovered is cobalt or nickel for instance.

Furthermore, in this study, only the constituents of cathode paste are recycled and reused in manufacturing new cells. However, the recycling technologies are designed for recovering as much as possible materials and constituents of the cells including aluminum and copper in current collectors, graphite and carbon black in anode paste, aluminum in a cell container, and electrolyte specifically in Direct Recycling etc. Therefore, the impact may be overestimated as the study does not credit the impact on these constituents from recycling.

Moreover, the model used in this study is also proven feasible for fewer recovered materials as allocation of recycling is being done individually for the recovered masses. Using each and every possible recovered material can be problematic and complex to model.

The recycling model LCIs are designed for spent cells after post-consumer use. However, this study assumes the scrap cells are rejected after post-production during QC checks. As mentioned earlier in the literature review, QC rejects can recover 80% of the cell contents while End of Life (EOL) cells can recover 77% as changes are observed in the composition, morphology, and alterations in the characteristics of these two types of cell categories (Marshall et al., 2020). The study has used a cathode comprised of 75% Lithium Manganese Oxide (LMO) spinel and 25% Lithium Nickel Cobalt Aluminum Oxide (NCA) unlike Nickel Manganese Cobalt (NMC) used in this thesis. It was observed that transition metals Mn: Co: Ni ratios were 74:3:23 and 73:6:22 for QC reject and EOL cells respectively, indicating a higher percentage of cobalt and low dissolution of manganese in the EOL cells. The study aimed to recover most of the valuable materials from the cells, therefore, the recovery rate cannot be compared directly with the recovery of only the positive active material constituents as 80% of the recovered cell contents from scrap cell includes aluminum, copper, polymers from the separator films, and the black mass from the electrodes. Therefore, it remains ambiguous whether QC scrap cells or EOL spent cells are strong candidates for positive electrode recovery, however, the recovery also depends on several factors e.g., the ease of disassembly, reverse engineering, and infrastructure availability.

The assumption that the recycled material can be fed directly into the production system is another factor that requires further investigation. The recovered material from the different types of recycling strategies discussed in the study may require further mechanical, chemical, or physiochemical treatment depending on the condition of the scrap cell. To feed the recycled material into the

production system, it needs to be ensured that the recovered material meets the standards completely so that the quality of the new cell produced with the recycled material is not compromised. This could be a critical aspect as if the cell is not produced with the desired quality, the scrap rate will be increased, and resources utilized in production will be wasted. Therefore, additional quality checks should be provided before the recycled product enters as an input to the production chain. The inclusion of these additional treatment processes and their respective impacts can further improve the quality of the results obtained.

Although there are certain limitations, the model is still useful for demonstrating how combining virgin and production chains in varying proportions may impact the overall GHG emissions associated with Li-ion cell production. Therefore, the model offers a systematic and methodological framework for conducting a more thorough process based LCA analysis of cell recycling. Enhancing the model reliability through parameterization as mentioned in the literature review and collaboration with production and recycling sectors, and the LCA community can further minimize the uncertainty in the results obtained. Furthermore, the sensitivity analysis is performed using an average electricity mix of countries. However, China has a huge geographical area with varying electricity mixes among various regions causing different carbon emissions (Q. Chen et al., 2022). Therefore, the use of regionalized LCI and LCIA characterization factors may also maximize the resolution of the calculated results.

5.2 Comparison with Other Studies

The outcomes of our model display the diversity of each recycling methodology, while also generating results that fall within the acceptable ranges reported in various LCAs of battery production as given in Table 5.

Table 5- Comparison with other studies

	Ref	Cell	FU	Method	Specific Energy kWh/kg	Rec Comp	Region	Virgin Prod	Direct	Hydro	Pyro
										kg CO_2-eq	
1	This Study	NMC111	1kg cell prod	ReCiPe midpoint (H) V1.03	0.224	Cathode	Norway	14.7	6.1	6.6	8.3
							Germany	21.19	12.42	13.13	14.82
							China	26.61	17.56	18.39	20.08
2	(S. Jiang et al., 2022)	NMC111	1kg cell prod	IPCC 2014	0.150	Cathode Al foil Cu foil	China	19.4	14	16.7	
3	(Q. Chen et al., 2022)	NMC811	1kWh battery	ILCD, 2022	~0.226	All	China	91.21 ~22.2	43.92 ~9.8	60.77 ~13.6	86.86 ~19.45
4	(Ciez & Whitacre, 2019)	NMC622	1kg battery cell prod	GREET 2016	0.27	All except materials not recovered through recycling	US Average (GWP20)	9.8	8	9	12
5	(Du et al., 2022)	NMC	1 kg NiCoMn ternary cathode materials	ReCiPe Midpoint		Ternary cathode materials	China	53.5		20.8	
6	(Yoo et al., 2023)	NMC8111	1kg CAM prod	GREET, 2021		Cathode Active Materials	Elec: South Korea 2017 Gas: US	24.9-28.6		15	

GWP is chosen as the basis for comparing the results of this study with those of other studies. This table presents a summary of a comparison between the results of this research and some previous studies, all of which focused on the production and recycling of NMC paste, cells or batteries. As is obvious from the table the results differ among studies. However, there are various reasons for the differences in the results of different studies. For example, different production and recycling processes may use different methods, technologies, and chemicals to manufacture and recycle LIBs, resulting in

varying chemical usage and energy consumption. Additionally, the background data differ depending on the geographical location of these studies. For instance, the environmental impacts of chemicals produced in a production and hydrometallurgy and direct recycling plant in China may differ from those produced in a similar LIB recycling plant in Europe. Furthermore, the results may also be influenced by the LCIA method used, including the characterization factors that may change over time. As shown in Table 5, different studies used different LCIA methods, such as ReCiPe, GREET, and IPCC, and were conducted in different years. Also, there is a marked difference in the cell type, energy density, and components recovered from recycling.

S. Jiang et al., (2022) conducted a process-based life cycle assessment to quantify the environmental impacts of hydrometallurgical recycling of two common lithium-ion traction batteries and reusing materials in their manufacturing in China. The study besides recycling the cathode, also recycles positive and negative current collectors based on Al and Cu foils respectively which justify less impact compared to the one obtained in this study.

Q. Chen et al., (2022) uses cradle to cradle LCA to investigate the carbon footprint and reduction potential of lithium-ion batteries in China. In order to compare the results, the impact due to functional unit of 1kWh are converted to 1kg as used in this study, according to the battery mass and capacity defined by Q. Chen et al., (2022). It is observed that the adjusted impact results are approximately close to what this study has calculated for pyrometallurgy. However, marked variation is observed under other categories. On the other hand, the results are found to be lying between this study and S. Jiang et al., (2022). Q. Chen et al., (2022) analyzed NMC811 where high nickel content demands higher energy for extraction and processing of nickel compared to cobalt and manganese, whereas S. Jiang et al., (2022) analyzed NMC111 where every element is under same ratio.

Ciez & Whitacre, (2019) utilizes attributional life-cycle analysis and process-based cost models to evaluate the greenhouse gas emissions, energy inputs, and costs associated with producing and recycling lithium-ion cells with three common cathode chemistries. It is notable that the GWP emissions in pyrometallurgy are even greater than the virgin production. This discrepancy is because the study has calculated the combined impact of manufacturing and recycling 1kg of battery and subtracted the emission offsets for NMC battery outputs to give recycling credit to production. Therefore, the 12 kg CO_2-eq does not represent the production of NMC batteries with pyrometallurgical recycled material but the combined impact of NMC battery production and recycling through pyrometallurgical treatment based on the US average electricity grid. Another highlighting factor is that the virgin cell production impact in Ciez & Whitacre, (2019) is lower compared to this study due to different cell chemistry being used. According to Emilsson & Dahllöf, (2019), NMC622 has 11% less GWP impact than NMC111 due to a lower fraction of cobalt. Another factor could be due to a change in the time horizon for GWP, this study has calculated the climate impact for 100 years while Emilsson & Dahllöf, (2019) calculated it for 20 years using GWP20.

Du et al., (2022) estimated 20.8 kg-CO_2-eq for producing 1kg of recycled NiCoMn ternary cathode materials whereas this study has calculated 18.39 kg-CO_2-eq impact for producing 1kg NMC cell. As it is obvious from the impact contribution that ternary cathode materials are more emission-intensive than other components of the cell, the higher impact estimated by Du et al., (2022) is justified as 1kg of NMC has only about 34% (i.e. 0.34kg) of the ternary cathode materials. Similar to Du et al., (2022), Yoo et al., (2023) also calculated the impact for Cathode Active Materials (CAM). Although impact calculated by Du et al., (2022) is higher which could be due to regional differentiation.

Numerous other factors, including the energy density of a cell, also influence the GWP of cell production. Typically, increasing the energy density of a cell can lead to a decrease in its GWP. Mohr

et al., (2020) observe the lowest production impacts and, therefore, the lowest net GWP, which can be attributed to their higher energy densities because less cell mass is needed to provide the same capacity, and less production energy is required to manufacture them. However, it is important to note that the relationship between energy density and GWP is not always straightforward, and other factors like the manufacturing process and raw material sources may also impact the GWP of cell production. Therefore, a comprehensive LCA is crucial to accurately compare the GWP of cells with different energy densities.

Despite the existence of some variations, the outcomes of this study generally align with the range of findings from previous research. This suggests that the generic models and life cycle inventories (LCIs) used in this study are effective, and therefore, the presented results can be considered valid.

This thesis selected the most interesting and valuable component of a cell, as all components cannot be reused as is. Also, there is a limitation of useful lifetime and contamination issues. Even recovered cathodes require further relithiation, however, the circular economy for battery materials involves reusing components depending on their reclamation quality, repairing, and lastly remanufacturing the remaining ones. The production of a completely recycled cell from recovered components is unrealistic currently, however, this study provides an overview of the anticipated effects if this needs to be done in the near future.

6   Conclusion

The study conducted a life cycle assessment of the production of lithium-ion cells by combining the conventional production process and recycling strategies. Mass allocation is used to partition the recycled product impact and combine it with the production model. The environmental impacts of producing 1kg of a cell are calculated by changing the proportion of virgin and recycled content. And finally, the country-specific factors of electricity mix, heat supply, and transport are tested to analyze the sensitivity of the calculated results.

Four impact categories, Global Warming Potential, Particulate Matter Formation Potential, Surplus Ore Potential, and Fossil Fuel Potential are selected for the impact assessment. Irrespective of the impact category, the results follow this sequence: $d_{direct} < d_{hydro} < d_{pyro} < d_{virgin}$ which aligns with the findings of the previous studies. Furthermore, it is found that increasing the recycling content to produce new cells reduces the impact, however, it depends on the recycling technology used where 100% recycling with pyrometallurgy is found to be high impact than 90% recycling with hydrometallurgy. The results also indicate a strong correlation between cell production and recycling impact with the carbon intensity of country-specific energy mix where virgin cell production in Norway is found to be more beneficial than recycled cell production in China.

However, the result from the study lies well within the range of existing bodies of knowledge, nonetheless, the variations in impact results are observed due to certain factors like cell chemistry, recycled components, impact assessment method, spatial differentiation, etc.   Recycled cell production is found to be environmentally beneficial, but government policies and consumer behavior are vital to realizing these recycling benefits. The study also suggests besides recycling, reducing cobalt content in positive electrode paste, transitioning to renewable energy, and reducing overall energy consumption will be integral steps towards sustainable cell production.

The allocation-based model used in the study also has certain limitations like assuming a similar impact of products based on the mass recovered. The scope of the study was only to cathode paste constituents while recycling processes are designed to recover as much as possible. The assumption that recycled material is fed directly to the production system without additional treatment requires further investigation to maintain the quality and standards of the new cell produced using recovered cell material. Implementing quality checks and treatment processes can enhance the reliability and outcomes of using recycled materials in the production chain.

Despite these limitations, the model remains useful in demonstrating the impact of combining virgin and recycled production chains in Li-ion cell production. The model's reliability and results' resolution can be further improved through parameterization, industrial collaboration, spatial differentiation, and LCA community insights.

In conclusion, the study addressed and identified the impacts expected while pursuing the electrification of transport through cell production and recycling. The findings emphasize the integration of decarbonization of electricity, research in environmentally positive cell production and recycling practices, and the application of relevant government policies are vital for sustainable cell production.

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8 Supporting Information
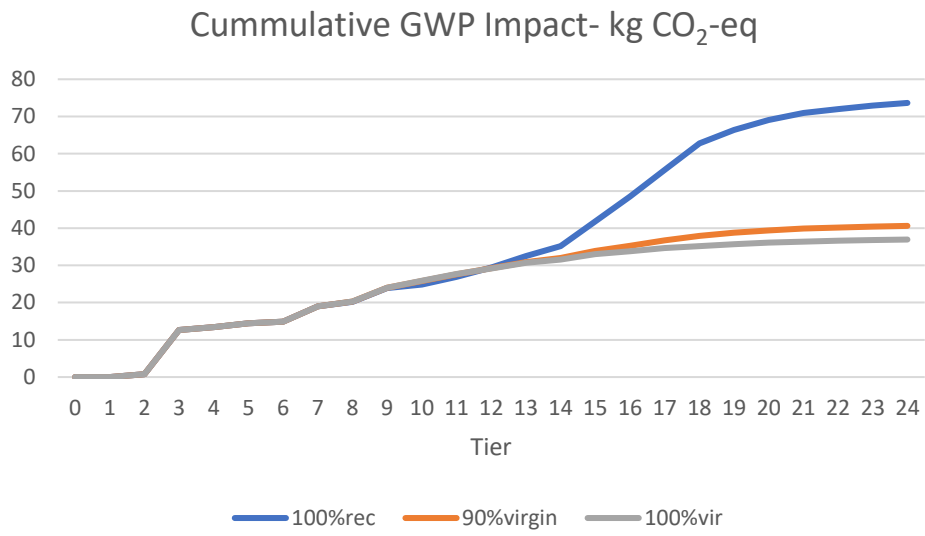
Cummulative GWP Impact- kg CO_2-eq

Figure S 1- Scaling effects in combining virgin and recycling production model.

Table S 1- Production LCI of 1kg NMC111 cell

Description	Input	Output	Unit	Ref	Ecoinvent 3.9 process
Functional unit					
NMC111 cell		1.00	kg	This Study	
Material input					
Cathode	0.44		kg	Ref-1	
Anode	0.32		kg	Ref-2	
Cell Container	0.11		kg	Ref-3	
Electrolyte	0.06		kg	Ref-4	
Separator	0.07		kg	Ref-5	
Energy input					
mixing anode	0.00		kWh		market for electricity, medium voltage / NO / kWh
mixing anode	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
mixing cathode	0.03		kWh		market for electricity, medium voltage / NO / kWh
mixing cathode	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
coating anode	0.00		kWh		market for electricity, medium voltage / NO / kWh
coating anode	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
coating cathode	0.00		kWh		market for electricity, medium voltage / NO / kWh
coating cathode	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
drying anode	1.03		kWh		market for electricity, medium voltage / NO / kWh
drying anode	1.36		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
drying cathode	5.33		kWh		market for electricity, medium voltage / NO / kWh
drying cathode	7.52		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
calendaring anode	0.00		kWh		market for electricity, medium voltage / NO / kWh
calendaring anode	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
calendaring cathode	0.00		kWh		market for electricity, medium voltage / NO / kWh
calendaring cathode	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
slitting anode	0.00		kWh		market for electricity, medium voltage / NO / kWh
slitting anode	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
slitting cathode	0.00		kWh		market for electricity, medium voltage / NO / kWh
slitting cathode	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
stacking	0.01		kWh		market for electricity, medium voltage / NO / kWh
stacking	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
filling	0.00		kWh		market for electricity, medium voltage / NO / kWh
filling	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
formation	3.05		kWh		market for electricity, medium voltage / NO / kWh
formation	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
floor heating	0.00		kWh		market for electricity, medium voltage / NO / kWh
floor heating	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
dry room	2.46		kWh		market for electricity, medium voltage / NO / kWh
dry room	4.96		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
miscellaneous	0.57		kWh		market for electricity, medium voltage / NO / kWh
miscellaneous	0.00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
Infrastructure Input					
Metal Refinery	8.96E-11		unit		precious metal refinery construction / SE / unit
Transport Input					
Transport Freight rail	0.13		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Transport lorry >32	0.02		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km

Description	Input	Output	Unit	Ref	Ecoinvent 3.9 process
Ref-1					
Functional unit					
Cathode		1.00	kg		
Material input					
Positive Electrode Paste	0.86		kg	Ref-6	
Positive current collector + Tab (Al)	0.14		kg		market for aluminum, primary, ingot / IAI Area, EU27 & EFTA / kg
Ref-2					
Functional unit					
Anode		1.00	kg		
Material input					
Negative Electrode Paste	0.65		kg	Ref-7	
Negative current collector +Tab (cu)	0.35		kg		market for copper collector foil, for Li-ion battery / GLO / kg
Ref-3					
Functional unit					
Cell Container		1.00	kg		
Material input					
Cell Container (aluminum)film	0.80		kg		market for aluminum, primary, ingot / RoW / kg
Plastic PET	0.04		kg		polyethylene production, high density, granulate / RER / kg
Nylon 6-6	0.12		kg		market for nylon 6-6 / RER / kg
CPP Proxy	0.04		kg		polypropylene production, granulate / RER / kg
Transport Input					
Transport Freight rail			tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Transport lorry >32			tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km
Ref-4					
Functional unit					
Electrolyte		1.00	kg		
Material input					
LiPF6	0.15		kg		market for lithium hexafluorophosphate / GLO / kg
EC	0.43		kg		market for ethylene carbonate / GLO / kg
DMC	0.43		kg		dimethyl carbonate production / RER / kg
Infrastructure Input					
Chemical factory	5.30E-12		unit		chemical factory construction, organics / RER / unit
Transport Input					
Transport Freight rail	0.60		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Transport lorry >32	0.20		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km
Ref-5					
Functional unit					
Separator		1.00	kg		
Material input					
LDPE	1.00		kg		polyethylene production, linear low density, granulate / RER / kg
Infrastructure Input					
extrusion, plastics	1.00		unit		extrusion, plastic film / RER / kg
plastics processing	6.05E-12		unit		plastic processing factory construction / RER / unit
Transport Input					
Transport Freight rail	0.20		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Transport lorry >32	0.10		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km

Description	Input	Output	Unit	Ref	Ecoinvent 3.9 process
Ref-6					
Functional unit					
Positive Electrode Paste		1.00	kg		
Material input					
LiNiMnCoO2	0.91		kg	Ref-8	
Additive	0.04		kg		market for carbon black / GLO / kg
CMC	0.02		kg		market for carboxymethyl cellulose, powder / GLO / kg
Binder	0.03		kg		market for polyvinylfluoride / GLO / kg
Solvent	0.01		kg		N-methyl-2-pyrrolidone production / RER / kg
Infrastructure Input					
Chemical factory	3.14E-11		unit		chemical factory construction, organics / RER / unit
Transport Input					
Transport Freight rail	0.21		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Transport lorry >32	0.10		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km
Ref-7					
Functional unit					
Negative Electrode Paste		1.00	kg		
Material input					
Graphite	0.91		kg		market for graphite / GLO / kg
Carbon black-anode	0.04		kg		market for carbon black / GLO / kg
Binder SBR/CMC-anode	0.02		kg		market for carboxymethyl cellulose, powder / GLO / kg
Binder PVDF-anode	0.03		kg		market for polyvinylfluoride / GLO / kg
Water	0.09		kg		market for water, decarbonised / CH / kg
Infrastructure Input					
Chemical plant	1.88E-11		unit		market for water, decarbonised / CH / kg
Transport Input					
Transport Freight rail	0.58		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Transport lorry >32	0.10		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km
Ref-8					
Functional unit					
LiNiMnCoO2		1.00	kg		
Material input					
NiCoMn (OH)2	0.95		kg	Ref-9	
LiOH (solid)	0.26		kg		market for lithium hydroxide / GLO / kg
Energy input					
Heating	0.92		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ
Infrastructure Input					
Chemical factory	4.00E-10		unit		chemical factory construction, organics / RER / unit
Transport Input					
Transport Freight rail	0.72		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Transport lorry >32	0.12		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km
Ref-8					
Functional unit					
NiMnCo(OH)2		1.00	kg		
Material input					
NiSO4 (anhydrous)	0.56		kg		market for nickel sulfate / GLO / kg
CoSO4 (anhydrous)	0.56		kg		market for cobalt sulfate / RoW / kg
MnSO4 (anhydrous)	0.55		kg		market for manganese sulfate / GLO / kg
Water	9.11		kg		market for water, decarbonised / CH / kg
NH4OH	0.26		kg		market for ammonia, anhydrous, liquid / RER / kg
NaOH	0.29		kg		market for sodium hydroxide, without water, in 50% solution state / GLO / kg
Energy input					
Electricity	0.96		kWh		market for electricity, medium voltage / NO / kWh
Heating	1.90		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland
Infrastructure Input					
Chemical factory	0.00		unit		chemical factory construction, organics / RER / unit
Transport Input					
Transport Freight rail	0.88		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Transport lorry >32	1.53		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km

Table S 2- LCI for Direct Recycling Model

Description	Input	Output	Unit	Reference	Ecoinvent 3.9 process
Functional unit					
LiNiMnCoO2 recycled		100%		Allocation of output-This Study	
Mechanical Process					
Material Input					
NaCl	7.35E-03		kg		sodium chloride production, powder / RER / kg
Energy Input					
Electricity	2.06E-01		kWh		electricity, high voltage, production mix / NO / kWh
Infrastructure Input					
Mechanical factory	4.60E-10		unit		mechanical treatment facility construction, waste electric and electronic equipment / GLO / unit
Transport Input					
Freight rail	1.05E+00		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Freight lorry	1.05E-01		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km
Product output					
Copper scrap		1.14E-01	kg		market for copper scrap, sorted, pressed / GLO / kg
Aluminum scrap		8.31E-02	kg		market for aluminum scrap, new / RER / kg
Emissions to air					
Dust		7.35E-04	kg		market for filter dust from Al electrolysis / GLO / kg
Volatile organic compounds		7.35E-05	kg		VOC, volatile organic compounds /air /unspecified /kg
Solid Waste					
Residue to landfill		2.94E-02	kg		process-specific burdens, residual material landfill / Europe without Switzerland / kg
Chemical process					
Relithiation					
Material Input					
LiOH	4.40E-02		kg		lithium hydroxide production / GLO / kg
Energy Input					
Electricity	6.47E-02		kWh		electricity, high voltage, production mix / NO / kWh
Infrastructure Input					
Chemical factory	1.53E-10		unit		chemical factory construction, organics / RER / unit
Transport Input					
Freight rail	3.50E-01		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Freight lorry	3.50E-02		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km
Filtration & drying					
Energy Input					
Electricity	6.47E-02		kWh		electricity, high voltage, production mix / NO / kWh
Heating	2.00E+00		MJ		steam production, as energy carrier, in chemical industry / RER / MJ
Infrastructure Input					
Chemical factory	1.53E-10		unit		chemical factory construction, organics / RER / unit
Transport Input					
Freight rail	3.50E-01		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Freight lorry	3.50E-02		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km
Emissions to air					
Dust		1.35E-04	kg		market for filter dust from Al electrolysis / GLO / kg
Volatile organic compounds		6.07E-05	kg		VOC, volatile organic compounds /air /unspecified /kg
Calcining					
Material Input					
LiCO3	3.75E-03		kg		lithium carbonate production, from concentrated brine / GLO / kg
Energy Input					
Electricity	6.47E-02		kWh		electricity, high voltage, production mix / NO / kWh
Infrastructure Input					
Chemical factory	1.53E-10		unit		chemical factory construction, organics / RER / unit
Transport Input					
Freight rail	3.50E-01		tkm		market for transport, freight train / Europe without Switzerland / metric ton*km
Freight lorry	3.50E-02		tkm		transport, freight, lorry >32 metric ton, EURO3 / RER / metric ton*km

Table S 3- LCI for Hydrometallurgical Recycling Model

Description	Input	Output	Unit	Ref	Ecoinvent 3.9 process
Functional Unit					
NiSO4 recycled		37%		Allocation of output- This Study	
CoSO4 recycled		37%			
MnSO4 recycled		13%			
LiOH recycled		13%			
Pretreatment					
Material Input					
water	6.60E-02		kg		market group for tap water / RER / kg
(Ca(OH)2) - slaked lime	1.56E-02		kg		market for lime / RER / kg
Energy Input					
Electricity	2.59E+00		kWh		market for electricity, high voltage / NO / kWh
Product output					
Scrap plastics		2.10E-01	kg		market for waste plastic, consumer electronics, sorted / GLO / kg
scrap metal		1.92E-01	kg		market for metal part of electronics scrap, in copper, anode / GLO / kg
scrap graphite		1.43E-01	kg		market for graphite / GLO / kg
Emissions to air					
Dust		3.60E-05	kg		market for filter dust from Al electrolysis / GLO / kg
CO2		4.23E-03	kg		Carbon dioxide, fossil /air /unspecified /kg
HF		1.16E-05	kg		Hydrogen fluoride /air /unspecified /kg
Emissions to water					
wastewater		1.08E+01	m3		market for wastewater from ground granulated blast furnace slag production / RoW / m3
Solid Waste					
waste		2.70E-02	kg		market for inert waste / Europe without Switzerland / kg
Leaching					
Material Input					
NaOH - Sodium Hydroxide	1.26E+00		kg		sodium hydroxide to generic market for neutralising agent / GLO / kg
Na2SO4	2.51E-04		kg		sodium sulfate production, from natural sources / RER / kg
H2SO4	1.18E+00		kg		market for sulfuric acid / RER / kg
NaClO	2.60E-02		kg		market for sodium hypochlorite, without water, in 15% solution state / RER / kg
water	6.97E+00		kg		market group for tap water / RER / kg
H2O2	6.12E-01		kg		hydrogen peroxide production, product in 50% solution state / RER / kg
Energy Input					
Electricity	1.42E-03		kWh		market for electricity, high voltage / NO / kWh
Emissions to air					
H2SO4 mist		9.80E-07	kg		Sulfuric acid /air /unspecified /kg
NMVOCs		7.14E-05	kg		NMVOC, non-methane volatile organic compounds /air /unspecified /kg
Solid Waste					
waste		1.75E-01	kg		market for inert waste / Europe without Switzerland / kg
P204 extraction					
Material Input					
Iron powder	2.29E-03		kg		market for iron pellet / GLO / kg
O2	7.99E-01		kg		market for oxygen, liquid / RER / kg
P204	7.18E-05		kg		market for organophosphorus-compound, unspecified / GLO / kg
Kerosene	4.28E-04		kg		market for kerosene / Europe without Switzerland / kg
Energy Input					
Electricity	3.46E-03		kWh		market for electricity, high voltage / NO / kWh
Product output					
Combination of Al, Cu, and Fe		5.87E-02	kg		market for scrap aluminum / Europe without Switzerland / kg
Emissions to air					
O2		2.88E-01	kg		Oxygen /air /unspecified /kg
P507 extraction & Stripping					
Material Input					
NaF	9.69E-02		kg		sodium fluoride production / GLO / kg
P507	7.12E-05		kg		market for organophosphorus-compound, unspecified / GLO / kg
Kerosene	4.28E-04		kg		market for kerosene / Europe without Switzerland / kg
HCl	1.01E-01		kg		hydrochloric acid production, from the reaction of hydrogen with chlorine / RER / kg
Energy Input					
Electricity	2.03E-02		kWh		market for electricity, high voltage / NO / kWh
Emissions to air					
HCl mist		3.68E-05	kg		Hydrochloric acid /air /unspecified /kg
Emissions to water					
wastewater		1.08E-02	m3		market for wastewater from ground granulated blast furnace slag production / RoW / m3

Table S 4- LCI for Pyrometallurgical Recycling Model

Description	Input	Output	Unit	Ref	Ecoinvent 3.9 process
Functional Unit					
$NiSO_4$ recycled		80%		Allocation of output-This Study	
$CoSO_4$ recycled		20%			
Pre-heating zone					
Material Input					
Slag from steel industry	1.70E-01		kg		market for blast furnace slag / GLO / kg
Limestone ($CaCO_3$)	3.00E-01		kg		market for limestone, unprocessed / RoW / kg
Sand (SiO_2)	1.50E-01		kg		market for silica sand / GLO / kg
Coke	3.30E-01		kg		petroleum coke production, petroleum refinery operation / Europe without Switzerland / kg
Metal smelting & reduction zone + gas treatment					
Material Input					
O_2	1.86E-01		kg		market for oxygen, liquid / RER / kg
water	1.00E+00		kg		market group for tap water / RER / kg
$Ca(OH)_2$ (Calcium hydroxide)	3.00E-02		kg		market for lime / RER / kg
Energy Input					
Electricity	1.30E+00		kWh		market for electricity, high voltage / NO / kWh
Solid Waste					
Slag		7.90E-01	kg		market for inert waste / Europe without Switzerland / kg
Dust		1.00E-02			market for filter dust from Al electrolysis / GLO / kg
Emissions to water					
wastewater		1.00E-03	m3		market for wastewater from ground granulated blast furnace slag production / RoW / m3
Leaching 1 & 2 + Solvent extraction					
Material Input					
water	9.10E+00		kg		market group for tap water / RER / kg
HCl	2.10E-01		kg		market for hydrochloric acid, without water, in 30% solution state / RER / kg
NaOH	1.87E+00		kg		market for sodium hydroxide, without water, in 50% solution state / GLO / kg
Energy Input					
Electricity	6.00E-02		kWh		market for electricity, high voltage / NO / kWh
Product output					
Copper compound	1.20E-01		kg		market for scrap copper / Europe without Switzerland / kg
Iron compound ($Fe(OH)_2$)	1.01E+01		kg		market for iron scrap, sorted, pressed / RER / kg
Oxidation					
Material Input					
H_2O_2	8.00E-02		kg		market for hydrogen peroxide, without water, in 50% solution state / RER / kg
Emissions to water					
wastewater		-1.01E-02	m3		market for wastewater from ground granulated blast furnace slag production / RoW / m3
Sintering					
Material Input					
Li_2CO_3 (Lithium Carbonate)	5.00E-02		kg		market for lithium carbonate / GLO / kg
Energy Input					
Natural gas	4.76E+00		MJ		market for heat, district or industrial, natural gas / Europe without Switzerland / MJ