

Should high-cobalt EV batteries be repurposed?

Using LCA to assess the impact of technological innovation on the waste hierarchy

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Abstract

Lithium-ion batteries (LIBs) are a key technology in decarbonizing the transportation and electricity sectors, yet the use of critical materials, such as cobalt, nickel, and lithium, lead to environmental and social impacts. Reusing, repurposing, and recycling mitigate battery impacts by extending their lifespan and reducing reliance on virgin materials. Innovation that reduces demand for these problematic materials and increases battery efficiency also reduces impacts. Two examples of this technological innovation include, (1) the development of energy dense cathode chemistry containing less cobalt, a material with high social and environmental impacts; and (2) the use of columnar silicon thin film anode, which results in increased energy density compared to the commonly used graphite anode. This research assesses whether these technological innovations change the currently understood waste hierarchy, which prioritizes reuse or repurposing prior to recycling. This is of interest because retired high-cobalt batteries could supply their constituent materials sooner if recycled immediately and be used in low-cobalt, higher-performing batteries. The assessment considers the life cycle environmental impacts of two end-of-life management routes for a high-cobalt LIB: first, recycling the battery immediately after the first use life to produce a new, and less material intensive battery, and second, repurposing the battery for a stationary storage application followed by recycling. Findings show that battery reuse reduces life cycle environmental impacts relative to immediate recycling. Thus, from an environmental perspective, the waste hierarchy holds, and steps to retain the batteries in their highest value use, such as through repurposing, should still be prioritized.

KEYWORDS

cobalt, industrial ecology, lithium-ion battery, recycling, second life, waste hierarchy

1 | INTRODUCTION

The electrification of transportation, enabled by the lithium-ion battery (LIB), has the potential to drastically decrease carbon emissions (Rogelj et al., 2018). Life cycle assessment (LCA) has been used to analyze the environmental impact of electric vehicles (EVs) to understand whether

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eliminating tailpipe emissions leads to real reductions in emissions on a life cycle basis, and to identify life cycle stages and materials that are hotspots of environmental impact (Bauer et al., 2015; Pero et al., 2018). Previous LCAs have demonstrated benefits are highly dependent on the source of electricity, the cathode chemistry used, and the service lifetime (Archsmith et al., 2015). Second to electricity generation, except in cases of low-carbon electricity, the upstream production of battery materials represents the majority of impacts. Specifically, impacts from the production of materials (Dai et al., 2019).

In addition to environmental impacts from battery production, research has demonstrated the social impacts related to the mining of materials are detrimental to the well-being of miners and the surrounding communities (Sovacool, 2019). Batteries are being designed with less cobalt because of these associated impacts, the susceptibility to geopolitical risk due to the geographical concentration in the Democratic Republic of Congo, and the increasing material prices (Benchmark Mineral Intelligence, 2022; Sovacool, 2019; The White House, 2021).

Impacts from battery manufacturing and concerns over resource constraints will continue to increase with LIB demand. As these LIBs age, a wave of batteries will begin retiring, and governments and industries are attempting to figure out the best use for this retired supply. The waste hierarchy is a guiding principle that has long been used by the European Union and United States as a prioritization framework for handling waste. This hierarchy is now being applied to retired LIBs and is as follows: prevent, reduce, reuse, recycle, recover, and dispose (Environmental Protection Agency, 2021; European Commission, 2020). The order is based on the ability of processes to save the most resources and result in the least environmental impact (van Ewijk & Stegemann, 2016).

Maximizing the lifespan and use of the battery can prevent the demand for new batteries and technological development can reduce the demand for materials through increased energy density, both of which decrease initial demand (Lander et al., 2021; Woody et al., 2020). The benefits of repurposing and recycling LIBs for stationary storage prior to recycling has been validated by Richa et al. (2017a) in an LCA reviewing the waste hierarchy and circularity, as well as in other LCAs more broadly focused on LIB second-use applications (Bobba et al., 2018; Casals et al., 2017; Cusenza et al., 2019; Richa et al., 2015).

Despite these earlier findings, the waste hierarchy for high-cobalt end-of-life (EOL) LIBs has recently been questioned due to the rapidly changing LIB technology (Emma Wiesner & NorthVolt battery, 2020; Harper et al., 2019). Previous studies do not consider that the hierarchy may not apply under changing technology conditions. For example, the use of silicon instead of graphite in the anode is being researched due to the potentially significant material efficiency gains. In addition, batteries manufactured today use up to four times less cobalt and have higher energy density. This displaced cobalt sulfate through recovery has been demonstrated to represent a significant fraction of the environmental gains from recycling (Kallitsis et al., 2022). If less efficient batteries retired today are recycled instead of reused, the conversion of materials to more developed technology is accelerated.

The overall lower material demand per kWh for new batteries could offset future demand and decrease long-term extraction (Dunn et al., 2021). To understand the most EOL route for high-cobalt batteries, an LCA of different EOL pathways is needed. Environmental LCA will not provide a full view of the tradeoffs, considering it does not include social impacts and the economics, although it can provide partial insight into the optimal waste hierarchy.

This research will assess the environmental impact of this technological innovation using LCA to evaluate if recycling should be prioritized for batteries with a high-cobalt content, or if their lifespan should be extended through repurposing. It will also contribute to a greater discussion around the evaluation and potential flexibility of the waste hierarchy.

1.1 | Literature review

1.1.1 | Lithium-ion battery manufacturing life cycle assessments

A number of LCAs have compared impacts of LIB cathode chemistries (Peters et al., 2017) demonstrating chemistries containing cobalt and nickel have a higher percent contribution to battery production GHG emissions (Ambrose & Kendall, 2016). Impacts also vary based on the levels of cobalt and nickel (Bonalumi & Tabrizi, 2022; Crenna et al., 2021; Degen & Schütte, 2022; Jinasena et al., 2021). Winjobi et al. (2022) found that with decreasing levels of cobalt and increasing nickel (NMC111 compared to NMC811) the greenhouse gas (GHG) emissions decreased by 7.5% but sulfur oxide (SO_x) increased by 104% due to emissions from the production of nickel. Kallitsis et al. (2020) also analyzed the impact of varying nickel-manganese-cobalt (NMC) stoichiometries but included the substitution of a graphite anode with a graphite and silicon mix, finding the mixed anode results in a 40%–50% decrease in all ecotoxicity 2008 ReCiPe midpoint categories.

1.1.2 | Lithium-ion battery technological modifications

Technological modifications that increase energy density have the potential to decrease environmental impacts through more efficient use of materials. Examples of this, as previously discussed, are the transition to lower cobalt cathode chemistry and the switch to a silicon anode. Silicon is a

promising replacement for graphite because it has a higher theoretical capacity, but batteries with a silicon anode are still not commercialized and plagued with the issue of anode volume change when cycling, leading to cracking and early failure (Baasner et al., 2020).

There are various types of silicon anodes in development to accommodate the issue of volume expansion, including nanostructured silicon (Ellingsen et al., 2016) and columnar silicon thin film anodes (Piwko et al., 2017). Nanostructured anodes are more porous than the typical graphite in order to allow for fluctuation (Baasner et al., 2020; Zuo et al., 2017), although the added required area leads to the loss of energy density provided by silicon (Piwko et al., 2017). Recent LCAs have found in some cases these anodes increase the environmental impacts due to (1) this lack of density improvement, and (2) the impact of processing silicon into a nanostructured anode (Wang et al., 2019; Wu & Kong, 2018). Research has shown that nanostructured anodes have potential environmental benefits if produced at industrial scale, but at current scale are still lacking (Deng et al., 2019; Li et al., 2014). Columnar silicon thin film anode appears to be a better option than nanostructured due to its high specific capacity and ability to have a higher anode to cathode ratio (Baasner et al., 2020). In this study we model the columnar silicon thin film anode because of the potential life cycle environmental benefits it can provide.

1.1.3 | Lithium-ion battery EOL life cycle assessments

The repurposing and remanufacturing of LIBs have been evaluated by a plethora of LCAs, demonstrating that in most cases it is environmentally advantageous to extend the lifespan (Cusenza et al., 2019; Richa, Babbitt, & Gaustad, 2017). The LCA results vary depending on the reference scenario, second-life use, allocation factor, and other assumptions made in the analysis. Various second-life storage applications have been analyzed, including solar PV system support (Bobba et al., 2018; Kamath, Shukla, et al., 2020), load shifting (Sathre et al., 2015), peak shaving (Berzi et al., 2020; Faria et al., 2014; Kamath, Shukla, et al., 2020; Sathre et al., 2015), and EV fast-charging stations (Kamath, Arsenault, et al., 2020). Sathre et al. (2015) found that when the storage is used for diurnal energy shifting it enables an increase of renewables on the grid. Richa et al. (2015) did a similar analysis and found the load shifting results in a reduced net cumulative energy demand due to the ability to store renewable energy instead of curtailing it (Richa et al., 2015).

In the waste hierarchy, after the battery has been used to its fullest extent through reuse and repurposing, recycling can provide environmental benefits through the recovery of materials (Sun et al., 2020). Research has demonstrated the benefits from recycling differ based on the type of batteries recycled, with benefits highest from batteries containing cobalt and nickel (Kallitsis et al., 2022), and from using high recovery processes such as hydrometallurgical or direct recycling (Ciez & Whitacre, 2019).

Pyrometallurgy is the most energy-intensive recycling technique and recovers the least materials, resulting in a higher net environmental impact than the alternatives of hydrometallurgy and direct cathode recycling (Anwani et al., 2020; Richa, Babbitt, & Gaustad, 2017). Hydrometallurgy is occurring at industry scale and recovers the constituent materials while direct recycling is still at pilot scale and recovers the full cathode active materials (Gaines et al., 2018; Harper et al., 2019).

1.2 | Contribution to literature

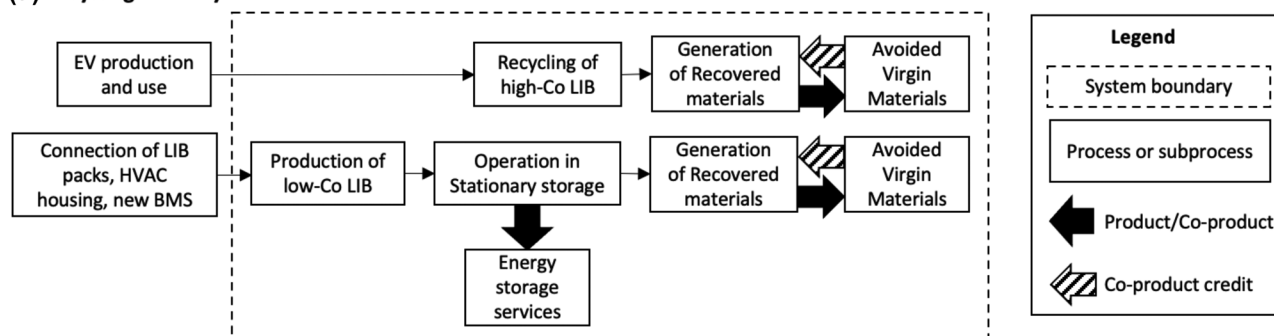
The literature review shows an extensive body of LCA work spanning all life cycle phases of the LIB (production, use in an EV, repurposing, second-life use, and recycling). However, the influence of technological innovation on the tradeoff between repurposing and recycling has not been explored by LCA.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

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

2 | METHODS

LCA evaluates environmental impacts throughout the life cycle of a product and throughout the supply chains that support a product system. The LCA conducted in this research largely conforms to ISO 14040 standards, and the following sections step through the three primary phases of the

(a) Recycling Pathway



(b) Repurposing Pathway

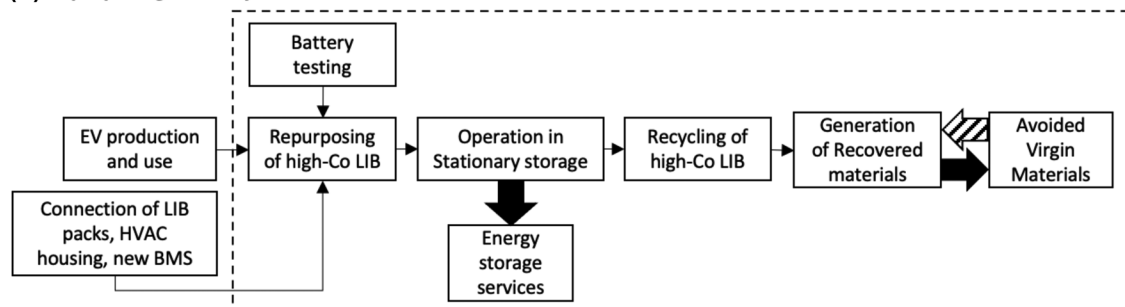


FIGURE 1 The two pathways modeling in this LCA are (a) the Recycling Scenarios and (b) the Repurposing Scenarios.

LCA methodology: goal and scope definition, life cycle inventory (LCI) analysis, life cycle impact assessment (LCIA), and interpretation of the results (International Organization for Standardization (ISO), 2006).

2.1 | Goal and scope definition

This study is a comparative LCA with the goal of identifying whether it is environmentally preferable to extend the life of a high-cobalt battery or recycle it after first use. The intended audience includes policymakers and parties responsible for retired batteries. To make the comparison, both management options need to be modeled through to final disposition of the battery via recycling, but also need to deliver the same service. The system boundary of this analysis starts with a retired EV battery, then two pathways are considered; a direct-to-recycling pathway where no repurposing of the battery is considered, but instead a new battery is manufactured for the stationary storage application, and a Repurposing Pathway where the battery is repurposed in a stationary storage application and then recycled. Because the initial production of the EV battery is identical between the two scenarios it is outside the scope of analysis.

The service provided by these two pathways is the cycling of electricity that is produced by solar photovoltaic panels in the United States. The functional unit represents a reference unit of the service provided by each product system (International Standard Organisation, 2006). Since the batteries analyzed are able to provide a different quantity of energy over their lifespan, the functional unit in this study is normalized to 1 kWh of cycled electricity through an LIB for a stationary energy storage service. This analysis does not consider other grid storage services or the differences in the capability across cell chemistries. A system expansion approach is used to provide the same function for the recycling and Repurposing Pathways. In the case of the Recycling Pathway, where no repurposing occurs, a new battery produced for the same energy storage service is included. The compared pathways are described in greater detail in the following text and in Figure 1. Note that capacity is defined in this work as the nominal capacity of the battery in comparison to the initial rated capacity.

Recycling Pathway: An LIB containing a high-cobalt cathode chemistry is retired from use in an EV when it reaches 80% capacity and sent directly to hydrometallurgical recycling. An LIB containing a different material makeup is manufactured in Europe for stationary storage use in the United States. When the stationary storage LIB reaches 50% capacity, the battery is recycled using hydrometallurgical processing.

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

TABLE 1 The total scenarios used in the comparative LCA.

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

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

In the Recycling Pathway, a new LIB stationary storage system is included within the system boundary to deliver the stationary storage energy service. The recycling is modeled as an open-loop system; recycled materials generated via recycling are assumed to displace virgin materials, but not necessarily for use in batteries.

2.1.1 | Scenario development

The future dominance of different LIB cathode materials, anode materials, and recycled content, is uncertain. In addition, current and previously manufactured EV LIBs have taken on a number of different chemistries, even within the NMC family of LIB chemistries, making the chemistry of a retired EV LIB uncertain as well. To address these uncertainties, scenario analysis is used to examine the effect of potential technology evolution for the new stationary storage LIB manufactured in the Recycling Pathway, as well as the chemistry of the retired EV battery. There are a total of 24 scenarios (Table 1): four scenarios vary the cathode chemistry of the retired EV LIB and the chemistry of the new LIB manufactured; two scenarios vary the anode material of the new LIB manufactured; and three scenarios vary the level of recycled content in the newly manufactured LIB. For the Repurposing Pathway, there are a total of three scenarios which represent different LIB cathode chemistries.

2.2 | Life cycle inventory analysis

2.2.1 | Lithium-ion battery life cycle inventory

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

Altering the cathode LCI

The cathode and anode scenarios in Table 1 were created by altering the $N_xM_yC_z$ ratio and anode material of NMC111-Gr. The six cell cases studied, are as follows: NMC111-Gr, NMC622-Gr, NMC811-Gr, NMC111-Si, NMC622-Si, and NMC811-Si. All dimensions and mass of the cell were kept constant, except for the active electrode material. This simplifying assumption, also used by Kallitsis et al. (2020), allows us to only consider the effects of changing the anode and cathode. In reality, changes to the chemistry will also result in changes to the design and alteration of other materials such as the electrode. Despite the simplification, we believe this analysis is still robust because when cell chemistry is modified by Winjobi et al. (2022), the impacts associated with changes to the other components are shown to be small.

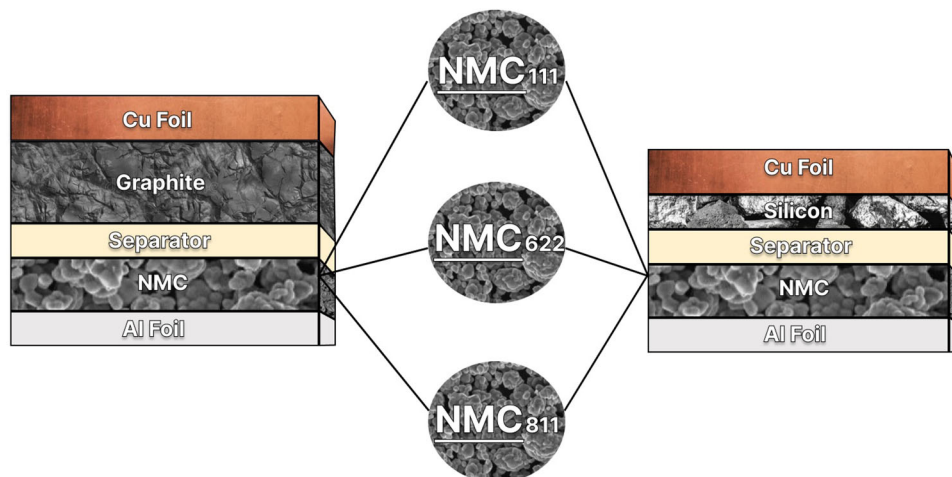


FIGURE 2 Visual depiction of the six cell chemistries: NMC111-Gr, NMC622-Gr, NMC811-Gr, NMC111-Si, NMC622-Si, and NMC811-Si.

To calculate the amount of nickel, manganese, and cobalt for each cathode chemistry scenario, we first calculate the mass of the cathode: NMC111, NMC622, and NMC811. Due to their compositional differences each mass will vary and is calculated using Equation (1):



Using these stoichiometric values of “x,” “y,” and “z” to assess the mass of metal sulfide reactants needed for the NMC-hydroxide precursors, the inventories of the unique NMC chemistries were calculated based on Equation (2):



Altering the anode and relevant LCIs

A major consideration in calculating the anode mass in the different cell chemistries was the theoretical capacity difference between graphite and silicon. Silicon has a theoretical capacity that is an order magnitude higher than graphite (~3579 and 372 mAh g⁻¹, respectively), a major benefit of transitioning from a graphite to columnar silicon thin film anode. This capacity difference translates to less active material needed in the anode to deliver the same energy density (Baasner et al., 2020).

To translate the base case configuration, NMC111-Gr to NMC111-Si, we assume the mass of silicon required to deliver the same theoretical capacity as graphite is lower by an order of magnitude. In addition, the density difference between graphite and silicon is assumed to be negligible (2.27 and 2.33 g cm⁻³, respectively), and thus is not considered. This calculation process uses the theoretical capacity (TC) and mass loading (ML) of graphite (Gr) and silicon (Si), as illustrated in Equations (3) and (4):

$$\text{TC}_{\text{Gr}} (\text{mAh g}^{-1}) \times \text{ML}_{\text{Gr}} (\text{g cm}^{-2}) = \text{TC}_{\text{Si}} (\text{mAh g}^{-1}) \times \text{ML}_{\text{Si}} (\text{g cm}^{-2}) \quad (3)$$

$$\text{ML}_{\text{Si}} (\text{g cm}^{-2}) \times \text{NMC111 Gr area} (\text{cm}^2) = \text{Mass of Si} (\text{g}) \quad (4)$$

This mass of silicon, calculated in Equation (4), is then scaled for pack level analysis and the mass of the cathode material is adjusted to maintain the 1.2 N/P ratio typical of silicon. Finally, both masses were normalized to maintain overall pack level mass.

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

The energy density varies between batteries and the capacity is calculated using equations from a model by Wentker et al. (2019). These calculations include varying cell voltage, practical discharge capacity, crystallographic density, and anode to cathode capacity ratio. Calculations are in Table S1 of Supporting Information S2. The cell manufacturing electricity usage also varies between chemistries and has been updated using data from Jinasena et al. (2021). The LCI of all batteries modeled in this study (NMC111-Gr, NMC622-Gr, NMC811-Gr, NMC111-Si, NMC622-Si, and NMC811-Si) can be found in Table S1-S22, and a visual depiction is shown in Figure 2.

TABLE 2 The lithium-ion battery capacity and total delivered kWh throughout the stationary storage lifespan.

Battery chemistry	Capacity (kWh)	Total delivered kWh from 100% to 50% capacity	Total delivered kWh from 80% to 50% capacity
NMC111-Gr	26.6	19,644	4624
NMC622-Gr	27.9	20,604	4850
NMC811-Gr	30.7	22,672	5337
NMC111-Si	34.3	25,326	5962
NMC622-Si	37.1	27,400	6450
NMC811-Si	42.0	30,985	7294

2.2.2 | Stationary storage use-phase inventory

The use phase of the stationary storage LIB consists of charging and discharging electricity generated by a 530 kWh solar photovoltaic panel in the Western Electricity Coordinating Council grid region of the United States. The delivered electricity is calculated by using the rated capacity of the battery (C_r), capacity at the current cycle y (C_y), 95% depth of discharge (DoD), and efficiency at the current cycle y (E_y). Equation (5) represents the kWh delivered (discharged) and Equation (6) represents the kWh consumed. The two equations differ by the efficiency, which decreases as the battery ages, thus requiring more kWh consumed per kWh discharged.

$$\text{kWh delivered} = \sum_y^c (C_r \times C_y \times \text{DoD} \times E_y) \quad (5)$$

$$\text{kWh consumed} = \sum_y^c (C_r \times C_y \times \text{DoD}) \quad (6)$$

Degradation of the battery over the lifespan is modeled using a capacity and energy efficiency fade. Capacity fade is based on data of an NMC111 battery from Yang et al (2018) and used to calculate C_y . The degradation rate is linear, until it decreases rapidly due to a “knee” at ~70%. The energy efficiency fade is modeled as a linear decline, similar to Richa et al. (2017) and Ahmadi et al. (2014), to calculate E_y used in Equation (5) (Section S3 of the Supporting Information S1). A shortcoming of this model is it does not include degradation or efficiency variance between chemistries, when in reality, higher cobalt chemistries degrade faster due to reduced cycling stability (Jung et al., 2018).

The Repurposing Pathway has a substantially lower number of cycles due to the shorter lifespan capacity and efficiency fade (Yang et al., 2018). The resulting lifespan kWhs delivered, as shown in Table 2, is used to calculate the functional unit. Calculations are in Table S2 of Supporting Information S2.

2.2.3 | Recycled materials inventory and allocation

Prior to recycling, the battery is removed from the EV, manually disassembled to the module level, and mechanically crushed. Hydrometallurgical recycling is then used to recover the constituent materials. The allocation of recovered materials from recycling is done assuming an open-loop process with no market disequilibrium, using the BPX 30-323-0 method (Allacker et al., 2014). In both pathways the recycled material is credited with the amount of material recovered based on the ability to substitute virgin materials. The 50/50 allocation approach is used to split the burdens and credits from recycling between the prior and subsequent product (Allacker et al., 2014). The EOL LCIs can be found in Tables S25–S26 and LCI results calculations can be found in Table S3 of Supporting Information S2.

2.3 | Life cycle impact assessment and interpretation of results

Life cycle impacts are calculated using ReCiPe 2016 midpoint and endpoint characterization factors. The midpoint characterization factors include: metal depletion factor (MDP), freshwater eutrophication (FEP), human toxicity (HTP), global warming potential (GWP), fresh water ecotoxicity (FETP), terrestrial ecotoxicity (TETP), freshwater consumption (WCP), human toxicity non-cancerous (HTPnc), land use (LOP), fossil depletion (FDP), ionizing radiation (IRP), photochemical ozone formation (POFP), terrestrial acidification (TAP), photochemical ozone formation—ecosystem

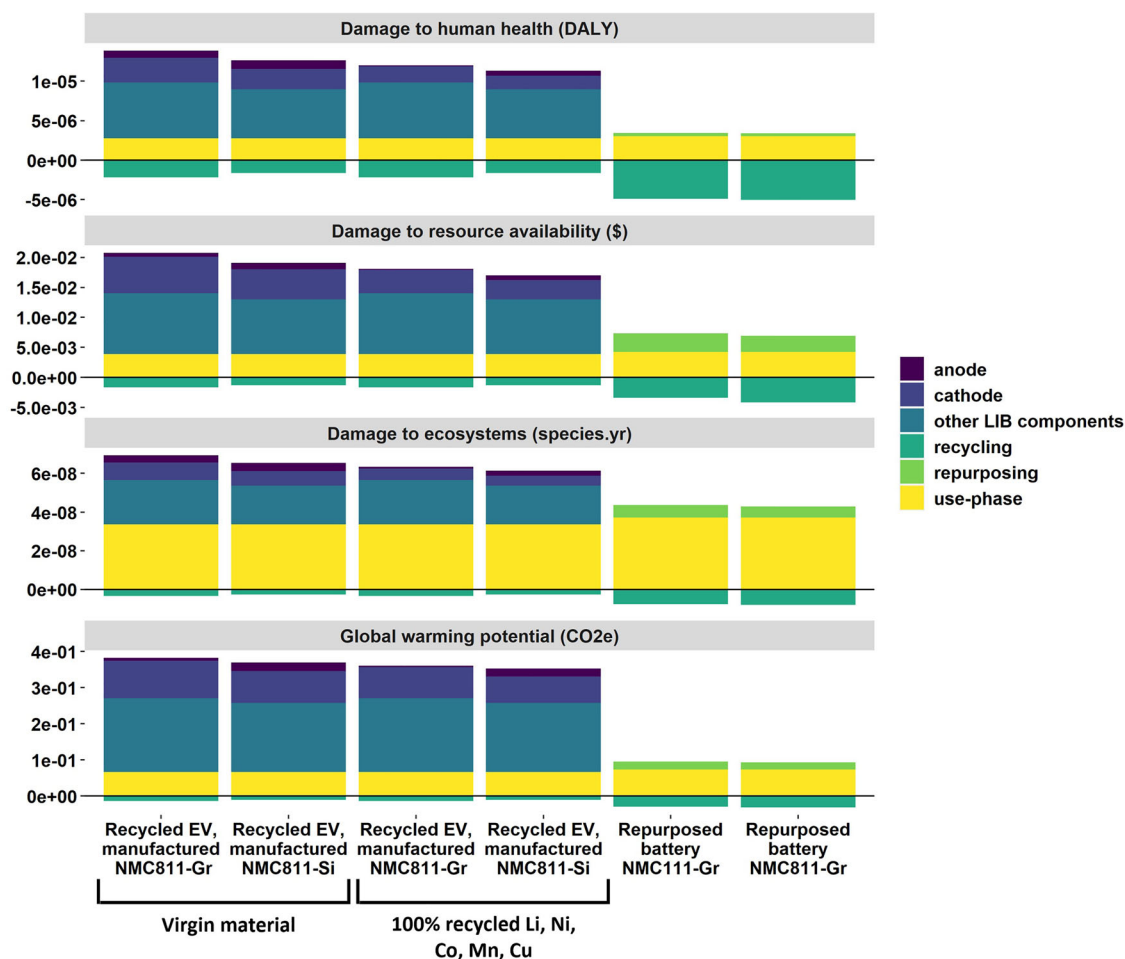


FIGURE 3 The impact per kWh of the cathode in a 253 kg lithium-ion battery is modeled for the Recycling Pathway scenarios (columns 1–4) and the Repurposing Pathway (columns 5 and 6). Within the Recycling Pathway, the impact of using recycled content in manufacturing and the use of a graphite and columnar silicon thin film is reviewed. The underlying data for this figure can be found in Supporting Information S2.

(EOFP), marine eutrophication (MEP), fine particulate matter formation (PMFP), marine ecotoxicity (METP), ozone depletion (ODP). The endpoint characterization factors include: Damage to Resource Availability, Damage to Human Health, and Damage to Ecosystems.

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

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

3 | RESULTS

In all impact categories the Repurposing Pathway results in less environmental impact than the Recycling Pathway (MDP, FEP, HTP, GWP, FETP, TETP, WCP, HTPnc, LOP, FDP, POFP, TEP, EOFP, MEP, PMFP, METP, ODP, Damage to Resource Availability, Damage to Human Health, and Damage to Ecosystems). This is due to the low impact of repurposing and the benefits from recycling; the net benefit from recycling the battery at EOL leads to negative impacts in the Repurposing Pathway. The recycling benefits are doubled in the Recycling Pathway due to two batteries being processed, but this is offset by the impacts of manufacturing, leading to net positive impacts. The benefits from recycling are larger in the Repurposing Pathway because it is cycled less times (Table 2, Figure 3), thus the benefits are divided by a smaller number.

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

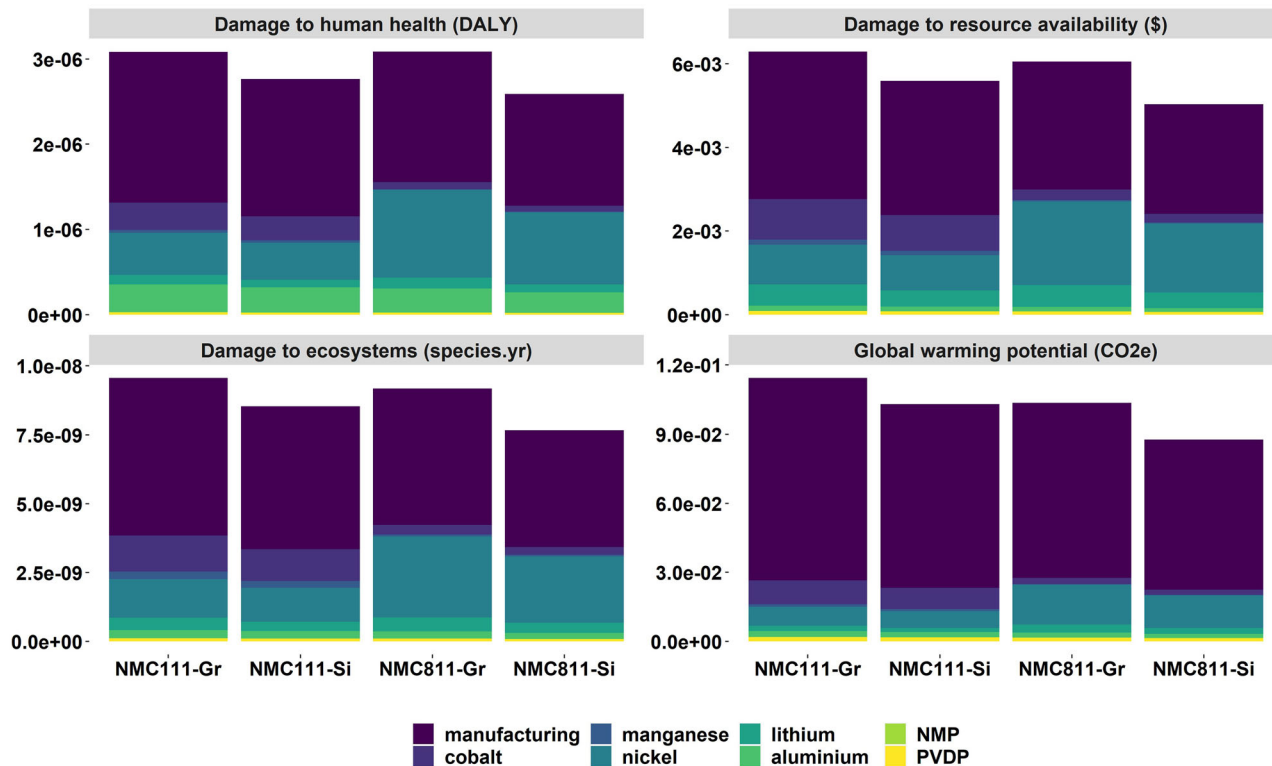


FIGURE 4 The ReCiPe endpoint impacts and GWP per kWh of the cathode in a 253 kg lithium-ion battery modeled. The kWhs of the batteries differ based on energy density. The underlying data for this figure can be found in Supporting Information S2.

Due to the manufacturing of new batteries in the Recycling Pathways, the cathode and the other LIB components represent a large portion of impacts (Figure 4). Cathode production is largest in the Damage to Resource Availability and GWP characterization factors because of impacts from cobalt, nickel, and aluminum. The “other LIB components” category is also significant and represents a larger portion of impacts in the Damage to Ecosystems characterization factor largely due to impacts from the aluminum, copper, and electricity usage in the production.

3.1 | Material substitution

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

The manufacturing of an LIB with an NMC811 cathode, instead of NMC111, results in lower impacts to all endpoint characteristics (Figure 4 and Figure S1), but the substitution of materials does not always result in the decrease of environmental impacts. Results show the NMC chemistry with decreased use of cobalt results in a net increase in some of midpoint impact indicators due to the high impact of nickel. The midpoint indicators where NMC 811 results in higher impacts than NMC111 include: FDP, FETP, IRP, MDP, PMFP, and TAP. NMC 811 has lower impacts in EOPF, GWP, HTP, MEP, ODP, TETP, FEP, LOP, and POFP (Figure S2).

3.2 | Recycled content

The impact of manufacturing a new battery is lessened with higher recycled content. All endpoint categories show the lowest impacts are from a battery produced with 100% recycled lithium, nickel, manganese, cobalt, and copper (Figure S3). In FEP, HTPnc, and TETP, the anode represents a large impact due to the use of copper foil and declines with the use of recycled materials is the result of recycled copper (Figure S5). The increased recycling does not considerably decrease the GWP, a category which has historically been given the most attention in LIB analysis.

3.3 | The impact of capacity fade, efficiency fade, and the electricity source

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

If a used battery is received at a capacity lower than 80%, and then cycled to 50% (i.e., decreased cycling of the repurposed battery), the impacts do not surpass the Recycling Pathways. This is because the largest impacts of the Repurposing Pathway are from the electricity cycling, so as the lifespan (cycles) decrease, so do use-phase-related impacts.

A decline in efficiency makes a much larger difference to the life cycle impacts of the Repurposing Pathway. When a repurposed battery has a very low efficiency, it takes in more electricity than is discharged, resulting in an increase in associated environmental impacts. The point where the repurposing of an NMC111-Gr battery is to be less advantageous than the recycling and manufacturing of an NMC811-Si battery with 100% recycled content, was found for the endpoint indicators and GWP midpoint indicator. For Damage to Ecosystems, the Recycling Pathway is more advantageous at or below a cycling efficiency of 28%. Damage to Resource Availability requires a lower efficiency of 20%. Due to the Repurposing Pathway having much less impact to Damage to Human Health, even with decreased efficiency, this trend continues.

The electricity used for battery cycling also has a large impact on the life cycle impacts of the Repurposing Pathway due to the impacts of electricity sourcing and the declining cycling efficiency. Thus, if the battery charges using the US average grid, the Recycling Pathway becomes preferable at an earlier cycling efficiency. For the Damage to Ecosystems indicator, the Recycling Pathway is preferable if the repurposed battery is at an efficiency of 30% or lower; Damage to Resource Availability is at 38% or lower; and Damage to Human Health at 29% or lower. Outputs can be found in Table S7 of Supporting Information S2.

4 | DISCUSSION

4.1 | The waste hierarchy

Findings show the currently understood waste hierarchy, of reduce, reuse, repurpose, and recycle, should continue to be used for LIBs of normal condition, when considering the material substitutions and impacts reviewed in this paper. These findings also show that it may be more beneficial to recycle batteries which have low cycling efficiency, if the new battery has increased density and is manufactured with recycled materials. Testing of batteries to access this battery health information will be crucial in determining the best EOL route.

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

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

When a truly market disrupting and innovative technology is developed, the EOL for the batteries under production may also need to be reconsidered (Figure 5). Thus, the impact of innovations on life cycle impacts and the optimal waste hierarchy should continue to be analyzed for product systems. While this can be true for all products, batteries using scarce materials are specifically suited for reviewing this relationship because of their long lifespan, expected ramping of demand, and the rate of innovation.

The technology reviewed in this study demonstrates that in most cases technological innovation will not alter the waste hierarchy from an environmental perspective, but a truly disruptive LIB innovation could result in the desire to recycle batteries early for their critical materials.

4.2 | Social impacts

Environmental impacts represent only one facet of the analysis that should be conducted. Literature documenting the humanitarian impacts from mining demonstrate that the review of social impacts is necessary to make a definite statement on the correct EOL path for these batteries. The

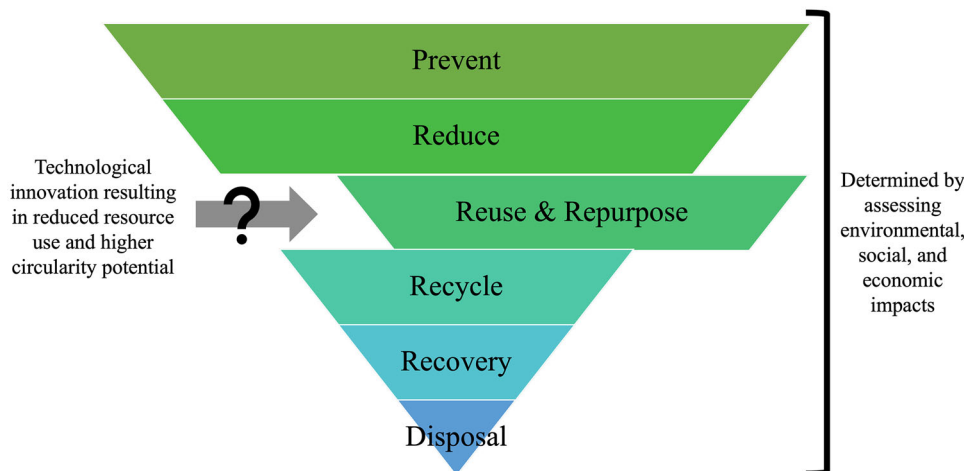


FIGURE 5 Technological development can impact the preferable waste hierarchy.

social impacts here refer more broadly than the socio-economic impacts discussed by Rasmussen et al. (2005), including the treatment and pay of laborers, the displacement of people, conflict, and corruption (Amnesty International & Afrewatch, 2016; Sovacool, 2019). While the environmental impact of cobalt is high, results demonstrate the manufacturing of low-cobalt chemistries also have relatively high environmental impacts, mainly due to the energy requirements and the use of nickel, aluminum, and copper. In addition, research has demonstrated that worker safety from cobalt manufacturing is high, although nickel and copper foil are higher contributors to potential health impacts (Arvidsson et al., 2022). These impacts are the reason using LIBs in a second-life application is better than recycling materials and producing a new LIB for stationary storage. Since this analysis does not consider the social impacts, it cannot speak to the socially preferable EOL route, and a social LCA is needed to lend in the comparison (Petti et al., 2018).

4.3 | Nickel as a substitute for cobalt

The substitution of nickel for cobalt is seen to be advantageous because nickel can provide higher density at a lower weight. In addition, nickel does not have as high a geopolitical risk, or the humanitarian concerns associated with cobalt. The NMC811 battery contains approximately four times less cobalt and nearly two times more nickel than the NMC111 battery (Argonne National Laboratory, 2020). Substitution has several corresponding environmental and economic impacts, pointing to the conclusion that we need to be careful about what materials we use as substitutes.

The refining process of nickel production results in high SO_x emissions (244.18 g kg^{-1}), a pollutant that damages foliage and decreases plant growth (Dai et al., 2019). The majority of material comes from Indonesia and the Philippines, where strip mining leaves little ability for rehabilitation of tropical rainforests and results in widespread biodiversity loss (Barlow et al., 2016; Supriatna et al., 2020). Second, there is a projected shortfall due to nickel class II refining capacity constraints (The White House, 2021). The increased demand for nickel and strain on resources is now leading to rapidly increasing costs.

4.4 | Availability of recycled materials

Using recycled materials results in a large reduction of impacts associated. Prioritization of ensuring recycling of batteries at their EOL to create a circular LIB economy will help materialize these reduced impacts. The increased demand for batteries as EVs begins to replace gasoline vehicles makes it so that all LIBs cannot be manufactured with 100% recycled materials (Dunn et al., 2021). While this is sizeable, it demonstrates a constraint to achieving minimum impacts using the current technology.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The data is available in an online repository: https://datadryad.org/stash/share/At1hpNyDZesuVd_bFif_mO_1qSvnWevoVkXs0W6Dm80.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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