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Comparative Life Cycle Assessment of three Recycling Approaches for Electric Vehicle Lithium-ion Battery after Cascaded Use

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With increasing concerns on environmental impacts of retired lithium-ion batteries (LIBs) and supply risk of critical materials, second life and recycling are considered as promising strategies to mitigate the environmental impacts of retired automotive LIBs. In this life cycle assessment (LCA) study, we investigate environmental benefits of second life and recycling methods on three types of widely adopted, commercialized automotive LIBs, including lithium manganese oxide (LMO), lithium nickel manganese cobalt oxide (NCM) and lithium nickel cobalt aluminum oxide (NCA). The results show that when subjected to second life, LIBs with both superior energy storage capacity and energy density generate more environmental benefits than LIBs that is less competitive in either working performance (31 % - 44 %, 16 % - 21 % and 50 % - 56 % reduction in environmental impacts). This is in line with the current trend towards the development of electric vehicles with larger energy storage capacity and longer battery life. We also found that direct cathode recycling outperforms hydrometallurgical and pyrometallurgical recycling regardless of battery chemistry. Among all recycling methods, only pyrometallurgical recycling is carbon intensive. The recovery of cathode active material and metals, including copper and aluminum, are critical to the environmental performance of batteries.

1. Introduction

Owing to the raised concern in global warming, technology innovation, reduced cost and increased reliability in safety and lifetime, the electric vehicle (EV) market grows exponentially starting from the year of 2010 and was estimated to reach an annual supply of 40 GWh (Ciez and Whitacre, 2011). Given the commonly 8-year warranty of EV, a huge amount of retired LIBs can be expected to be disposed of (Luo & Zeng, 2017). Previous studies showed that lithium-ion batteries lost only around 20 % in their initial energy capacity at the end of their life in EV (Wang et al., 2019), because of the rigid performance standards of EV (Neubauer and Pesaran, 2011). However, less-demanding applications such as stationary energy storage systems (ESS) could serve as an alternative to direct disposal (Zackrisson et al., 2010). Tesla has developed a Megapack made up of LIBs in Australia and successfully facilitate in mediating the local electric grid and resulting in an annual saving of nearly $40 MM (Tesla, 2019). Spent LIBs have the potential to be re-used in ESS to make profits, (Heymans et al., 2014) and alleviate burdens metal supply (MaKinsey, 2018).
Previous work also proved environmental feasibility of cascaded use to offset energy consumption and carbon footprint (Richa et al., 2017). However, the effect of cascaded use on different types of LIBs has not yet been explored. Besides, the impact of cascaded use on different recycling methods has not been studied. Other than global warming potential (GWP) and cumulative energy demand (CED), a more comprehensive environmental profile such as ReCiPe and TRACI is under investigation. In this study, we conduct a comparative LCA to assess the GWP, CED and ReCiPe endpoint indicators for LMO, NCM and NCA LIBs with first use in EV and second use in ESS before subsequently put to one of the three recycling methods, namely hydrometallurgical, pyrometallurgical and direct cathode recycling.

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2. Material and methods

In this study, we conducted a “cradle-to-cradle” LCA to estimate the GWP and CED associated with the production, consumption and end-of-life of three automotive lithium-ion batteries, namely LMO, NCM622 and NCA, after cascaded use in the stationary energy storage system. In addition, to obtain a more comprehensive environmental profile, 3 ReCiPe endpoint indicators from a hierarchal perspective were evaluated to assess damage to ecosystems, damage to human health and damage to resource availability. All values of GWP, CED and ReCiPe scores were sourced from Ecoinvent V3.6 (Wenet et al., 2016).

2.1 Life cycle inventory

The environmental impact associated with cathode active material of LMO was obtainable directly through the Ecoinvent database while the environmental impact associated with avoided $Ni(OH)_2$ production and cathode active material of NCM and NCA production were not available. Instead, the upstream materials and energy for producing $Ni(OH)_2$ sourced from (Majeau-Bettez et al., 2011) and the NCM and NCA cathode active materials were acquired from the GREET model (Dunn et al., 2012). The production of NCM and NCA cathode active material requires material inputs of lithium source, transit metal sources, deionized water, NH_4OH and NaOH, energy inputs and waste sludge treatment (Zhao et al., 2019). It is worth to mention that the product yields are all assumed to be 100 % since the production is modelled based on industrial data which is designed to minimize material loss and result in nearly 100 % product yields. In addition, the by-product of sodium sulfate was excluded from the system boundary using the concept of “recycled content” approach.

Working parameters, such as energy consumption rate in electric vehicle and energy storage capacity, are extracted from the BatPac model. Other working parameters, such as initial battery discharging efficiency, battery discharging efficiency after electric vehicle use and after stationary ESS use, the lifetime of the EV battery pack and stationary ESS battery pack, roundtrip and transmission efficiency for stationary ESS, and Depth-of-discharge (DoD) of stationary ESS battery pack, are extracted from literature (Richa et al., 2017). Automotive use is assumed to be 100,000 miles over an 8-year timeframe for both use scenarios of all types of batteries, and will the discharging efficiency would decrease from 100 % to 80 % (Väyrynen and Salminen, 2012). Starting from an initial discharging efficiency of 80 %, Stationary ESS use is assumed to operate one charge/discharge cycle on a daily basis for 10 years with an end-of-life (EOL) discharging efficiency of 65 %.

After retired from electric vehicles, battery cells will be tested to validate their residual energy capacity and consumed electricity. (Richa et al., 2017) Life-extending battery cells are assumed to be repurposed into new battery packs with an energy storage capacity of 450 kWh, aiming for their second use in stationary ESS. All battery module components and part of battery pack components such as module compression plates and steep straps are reused, while coolant, battery management system (BMS), module interconnects and a tri-layer battery jacket are replaced. The replaced components will be sent to the EOL phase. Notably, BMS is assumed to be 1.5 % of the total mass of one repurposed battery pack, based on the fact that BMS of battery packs designed for stationary ESS is not as demanding as those for the electric vehicles, and less advanced printed wiring board design and less complicated wiring would be applied. A cabinet made of mild steel sheet was used as the pack casing instead of the tri-layer jacket. The life cycle inventory for two use phase scenarios and three EOL scenarios will be detailed in the following two sections.

2.2 Use phase scenarios

Two use phase scenarios are assessed: one is electric vehicle use only, and the other is second use in stationary energy storage system after EV use. We adopt a combination of two functional units, namely functional unit LCED (1 kWh of life-cycle electricity delivered) and functional unit ESC (1 kWh of battery energy storage capacity). Both functional units measure the function of lithium-ion batteries, although from different perspectives. In terms of comparing life-cycle environmental impacts of batteries with EV use scenario and cascaded use scenario, the functional unit LCED delivered focus on evaluating the batteries’ life-cycle performance of delivering electricity. However, in terms of comparing across different types of batteries, the functional unit LCED is considered insufficient due to its incapability to reflect the energy storage capacity of different batteries. The functional unit of ESC, which is widely adopted in previous LCA studies of lithium-ion batteries, is added in addition to the functional unit of LCED to complement the shaded view.

2.3 End-of-life scenarios

EOL of lithium-ion batteries involves dismantling, material production, energy generation, incineration, combustion, waste sludge treatment, energy and material recovery. The environmental impact associated with energy and material recovery is considered as avoided burden and is reported as a reduction in carbon footprint and primary energy consumption. A comprehensive material flow analysis is performed and coupled with LCA (Gao et al., 2018). Three EOL scenarios, namely hydrometallurgical, pyrometallurgical and direct cathode recycling, are investigated. Notably, for all EOL scenarios, the spent LIB pack was first discharged and dismantled. In this step, the plastics were landfilled. The metals such as aluminum, chromium steel and copper are recycled with a loss rate of 5 %, 1.3 % and 8 %. Waste steel is landfilled and waste fiber glasses are incinerated. Printed wiring boards are prepared for further metallurgical treatment. (Wenet et al., 2016) Coolant is incinerated as hazardous waste. After disassembly, cathode, anode and electrolyte are left. Besides, plastics, aluminum and copper, were left for the scenario of pyrometallurgical recycling. Hydrometallurgical recycling aims at recovering metals using aqueous chemistry, which involves steps of leaching, solvent extraction and precipitation. Under this scenario, the leftovers are soaked in N-Methyl-2-Pyrrolidone (NMP) and crushed to dissolve the binder Polyvinylidene fluoride (PVDF) and separate the active materials from the current collectors. (Dunn et al., 2012) Later, solids such as graphite, carbon black, copper and aluminum are filtered out, the carbon black and graphite are completely combusted to CO_2, and the filtrate is calcined and grinded. Leaching and precipitation processes are used to extract the valuable metals and recover the cathode active material. The leaching efficiency is 95 % of Li for LMO, 99.7 % of Li, Ni, Co, Mn for NCM, and 80 % of Li, 99.8 % of Ni, 95.6 % of Co and 99.5 % of Al for NCA.

Direct cathode recycling process focuses on recover the cathode active material and electrolyte with limited processing. After dismantling, cathode, anode and electrolyte are crushed and electrolyte is extracted by liquid CO_2. Experiments showed that the recovery rate of $LiPF_6$, ethylene carbonate and dimethyl carbonate are 100 %, 75.76 % and 81.71 %. (Nowak and Winter, 2017) After the electrolyte was extracted and recovered, the residues underwent steps of size reduction and final separation which both requires electricity. Then, they undergo NMP soaking, hydrothermal treatment and annealing process to recover the cathode active materials.

Pyrometallurgical recycling is developed by Umicore and is designed to recover Nickel and Cobalt by forming an alloy through a three-stage smelting. (Cheret and Santen, 2007) After disassembly, the left spent LIB modules is fed directly into the smelting furnace together with slag-forming materials. First, the electrolyte is evaporated and the plastics are melted. Subsequently, blast furnace slag and alloy are formed. The gaseous phase is post-combusted in a combined heat and power generation (CHP) facility and complete combustion was assumed. The alloy is further leached and precipitated to recover copper, goethite, $Ni(OH)_2$ and cobalt ion solution.

3. Results and discussion

The results of carbon footprint, primary energy consumption and ReCiPe endpoint indicators are presented based on distinguished use scenarios and EOL scenarios.

3.1 Environmental benefits of introducing second life to batteries

To assess the implications of second life application to LIBs from the environmental perspectives, we compare the life cycle environmental impacts between the two use scenarios (with and without second life) across LMO, NCM and NCA LIBs using functional unit LCED and ESC as shown in Figure 1a and 1b. Life cycle environmental impacts are quantified in the form of ReCiPe scores. Environmental scores are calculated for each ReCiPe midpoint indicators by translating complicated life cycle inventories (LCIs) into 18 impact categories. Later, ReCiPe scores of 3 endpoint indicators are summarized into the ReCiPe total scores and are shown as data labels on top of each bar in Figure 1The EOL recycling method is kept the same when this comparison is made (Yang et al., 2017). From the perspective of life-cycle devotion to electricity delivery (Figure 1a), NCM LIBs perform the worst without second life (16 %, 76 % and 83 % more than the total ReCiPe score of LMO LIBs, and 27 %, 29 % and 40 % more than that of NCA LIBs for hydrometallurgical, direct cathode and pyrometallurgical recycling). In the presence of cascaded use, the environmental performance of NCM LIBs become drastically favorable and comparable with those of LMO and NCALIBs.

This notable reduction in the ReCiPe total scores of NCM LIBs can be mainly attributed to the highest amount of electricity delivered during second life. Remarkably, this can be fundamentally attributed to three factors: (1) the superior energy storage capacity of the NCM LIBs (88.17 kWh per EV battery pack) over the LMO and NCA LIBs (23.53 and 52.94 kWh per EV battery pack), which results in a comparable level of electricity delivery during automotive use but a significantly higher amount of electricity delivery during second life; (2) the outstanding battery pack energy density of NCM (181 Wh/kg) that conveys higher energy storage capacity to the same weight of environmentally-expensive LIB pack, compared to the LMO (159 Wh/kg) and NCA (172 Wh/kg) LIBs; (3) the overwhelming environmental impacts associated with a large amount of electricity delivery during the use phase (accounts for 55 % - 62 % and 24 % - 31 % of the total ReCiPe score for NCM LIBs with and without second life). The third factor is also responsible for the increase in ratios of scores for "damage to human health" and "damage to resource availability" and decrease in that for "damage to ecosystems". Since the electricity grid in New York State (NYS) is mainly powered by natural gas, nuclear and hydroelectric,"damage to human health", "damage to resource availability" and "damage to ecosystems" account for 29 %, 29 % and 42 % of the total ReCiPe score associated with the use phase.

When environmental impacts associated with use phase increases, the shares of ReCiPe scores for three endpoint indicators get closer to that of the use phase.

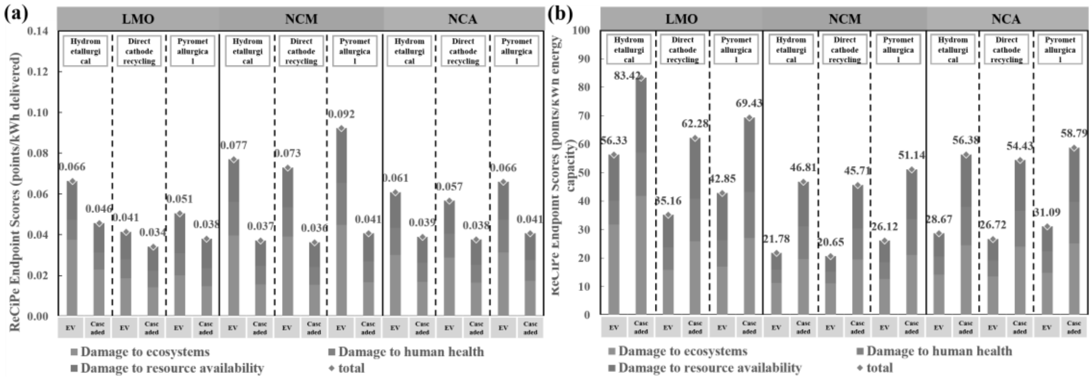

Figure 1: Score of ReCiPe endpoint indicators comparing battery chemistries, use scenarios and EOL scenarios based on a. functional unit LCED and b. functional unit ESC.

Switching to the perspective of energy storage capacity, NCM LIBs achieve outstanding environmental performance while LMO LIBs achieve the worst performance regardless of the use scenarios. This is because of the superiority in both battery pack energy storage capacity and energy density. Notably, the difference between use scenarios is not significant, because functional unit ESC does not address any function related to the second life.

3.2 Environmental impacts of battery recycling methods on second life

The environmental impacts of battery recycling methods on second life are shown in Figure 1. The EOL phase appears to be the leading contributor for the total ReCiPe score. The direct cathode recycling outperforms the other two recycling methods regardless of battery chemistry, while hydrometallurgical recycling outperforms pyrometallurgical recycling only for NCM and NCA LIBs. Due to the utility of environmentally expensive leachate citric acid and the manganese source Mn_2O_3, hydrometallurgical recycling of LMO underperforms pyrometallurgical recycling. Instead, EOL phase with positive ReCiPe score is much less influential than EOL phase with negative ReCiPe score. Hydrometallurgical and direct cathode recycling result in comparable negative ReCiPe scores, while hydrometallurgical recycling leads to a higher positive ReCiPe score compared to the direct cathode recycling. On the contrary, pyrometallurgical recycling leads to both the most positive ReCiPe score and the least negative ReCiPe score for all LIBs.
Both the inter-battery chemistry and the inter-EOL scenario variances in the total ReCiPe scores become less. In particular, for use scenario with and without second life, the inter-EOL scenario standard deviation change from 0.013, 0.010, 0.005 to 0.006, 0.002, 0.002 for LMO, NCM and NCA LIBs. The inter-battery chemistry standard deviation changed from 0.008 – 0.021 to 0.001 – 0.005.

3.3 Life-cycle carbon footprint and primary energy consumption

The GWP and CED are two important metrics to evaluate the climate change mitigation potential and energy performance of introducing second life and recycling into batteries' life cycle. The results of GWP and CED are compared from the perspective of life cycle delivered electricity and energy storage capacity as demonstrated in Figure 2a and 2b using stacked bars and red rhombus.
Figure 2a and 2b show that from the perspective of functional unit LCED, the GWP and CED result in environmental benefits of introducing the second life application to LIBs is consistent with that of ReCiPe results: NCM LIBs without second life are the most carbon intensive among all types of LIBs (the GWP of NCM LIBs is twice more than that of LMO LIBs), while under the cascaded use scenario, carbon footprint of NCM LIBs become comparable to that of NCA LIBs and only 32 % - 51 % more than that of LMO LIBs. In particular, the difference between life-cycle carbon footprint of NCM and LMO LIBs drops from 0.092 – 0.188 kg CO_2 eq. to 0.009 – 0.023 kg CO_2 eq. after adopting second life. Compared with use scenario of only automotive use, LMO, NCM and NCA LIBs with second life achieve 9 % - 15 %, 31 % - 44 % and 19 % - 30 % reduction in life cycle carbon footprint. The reductions in primary energy consumption are less distinct in that LMO, NCM and NCA reduce 3.6 % - 4.8 %, 15.6 % - 21.3 % and 8.9 % - 13.2 % of CED when adopting second life. This suggests

that while LMO remains to be the least carbon and energy intensity, the adoption of second life can pull down the high carbon and energy intensity of LIBs such as NCM and NCA substantially to an acceptable level. In other words, the adoption of second life can relieve many of the concerns towards the commercialization of lab-scale batteries with superior working performance but costly environmental impacts.

Compared to the ReCiPe results, cascaded use accounts for a higher share in the total life cycle GWP (81.4 % - 87.1 %, 74.1 % - 83.6 %, and 74.0 % - 83.3 % for LMO, NCM and NCA LIBs) and CED (93.6 % - 94.9 %, 90.4 % - 92.8 % and 90.2 % - 92.8 % for LMO, NCM and NCA LIBs). Notably, the shares of automotive use in the GWP and CED values associated with cascaded use for LMO, NCM and NCA LIBs is the same as that of the ReCiPe result (46.6 %, 22.5 % and 32.6 %). This revalidates the conclusion that batteries with larger energy density could be more environmentally sustainable if subjected to second life. In consequence of the large proportion of CED related to the use phase, other phases appear not as significant as what is shown in GWP results.

Instead of being the most influential contributor to the total ReCiPe score, the EOL phase appears to be the second leading contributor for both carbon footprint and primary energy consumption. Among the three EOL scenarios, direct cathode recycling appears to be the least carbon intensive for all LIBs. On the contrary, hydrometallurgical recycling of LMO LIBs surpasses direct cathode recycling in terms of primary energy consumption. Notably, only pyrometallurgical recycling results in a net positive carbon footprint, while it appears to be slightly energy saving. This result is different from that of the ReCiPe score, where all three recycling methods can lead to net negative ReCiPe scores that have much larger impacts on avoiding batteries' environmental burdens. The result suggests that instead of using metrics such as carbon footprint and primary energy consumption, the environmental benefits of introducing second life and recycling into batteries' life cycle can be better revealed by the systematic life cycle environmental impact assessment.

By further disaggregating the EOL phase into positive and negative portions, it can be observed that direct cathode recycling results in the least positive GWP, while at the same time avoiding the most GWP for all three types of LIBs. Nevertheless, pyrometallurgical recycling leads to both the most positive GWP and the least negative GWP. Similarly, direct cathode recycling and pyrometallurgical recycling lead to the most and the least negative CED. The positive CED results are more complicated: first, hydrometallurgical recycling has the most positive CED for NCM and NCA LIBs, but LMO LIBs has the least positive CED by using hydrometallurgical recycling; second, direct cathode recycling leads to the most positive CED for LMO LIBs and the least positive CED for NCA LIBs; the pyrometallurgical recycling leads to the least positive CED for NCM LIBs.

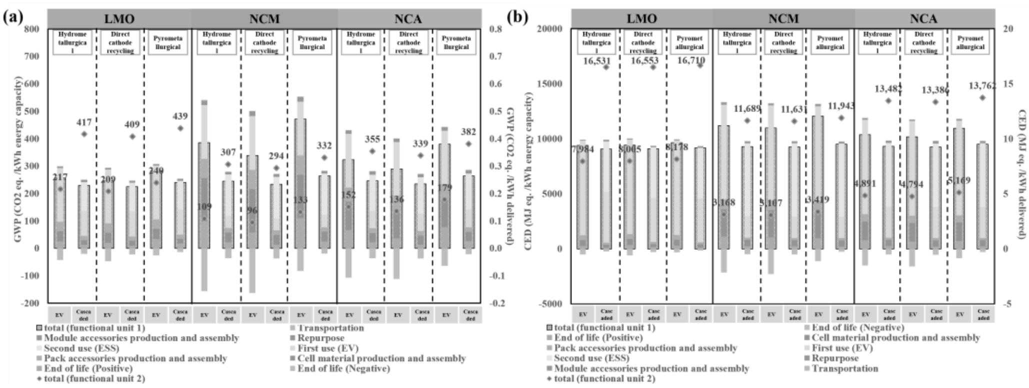

Figure 2: Breakdown of a. carbon footprint and b. primary energy consumption for LMO, NCM622 and NCA LIBs with three EOL scenarios and two use scenarios

As a consequence of the large proportion of GWP and CED related to the use phase, other phases appear not so significant as shown in ReCiPe results. The material production and assembly for cell and pack accessories are ranked third and fourth in their contributions to carbon footprint and primary energy consumption. Among the three types of LIBs, NCM cell production appears to be the most carbon and energy intensive (1 % - 5 % and 108 % - 118 % higher than GWP and CED of NCA and LMO cell production). The pack accessories production and assembly do not show a significant difference in GWP between these LIBs. Module accessories production and assembly, repurpose and transportation account for no more than 5 %, 3.5 % and 1 % of the life cycle GWP and CED. They become relatively negligible.

From the perspective of functional unit ESC, NCM LIBs outperforms LMO and NCA LIBs in terms of life-cycle carbon footprint and primary energy consumption regardless of EOL scenarios. Direct cathode recycling remains to have the smallest carbon footprint and performs slightly worse than hydrometallurgical recycling in terms of primary energy consumption.

4. Conclusions

The comparative LCA illustrated the advantages of second use for spent LIBs from automotive use, especially on those with superior performance such as high energy storage capacity and high energy density, i.e. NCM LIBs in this study. Concerns towards environmental impacts can be relieved in that the adoption of cascaded use reduces 50 % - 56 % of ReCiPe score, 31 % - 44 % of carbon footprint and 16 % - 21 % of primary energy consumption for NCM LIBs and pulled down the environmental impacts of NCM LIBs to a level that is comparable to that of other batteries with less environmental impacts. Comparison of recycling methods points out the necessity to recover cathode active material that is shown to be the most environmental expensive process throughout the batteries' life cycle. Direct cathode recycling outperforms hydrometallurgical and pyrometallurgical recycling in all impact categories, however, fell short of expected advantages due to the overwhelming energy consumption of liquid CO_2 production, suggesting that efficiency improvement and the alternatives to the current environmental expensive process or materials could be further studied in the future battery recycling research.

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