



Research Paper

Life cycle assessment of secondary use and physical recycling of lithium-ion batteries retired from electric vehicles in China

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ABSTRACT

With the rapid development of the global new energy vehicle industry, how to minimize the environmental impact of the recovery has become a common concern and urgent concern. China is a major production and consumption market for electric vehicles, there are no specific and extensive resource and environmental assessment system for batteries. In this paper, the retired Electric vehicles lithium-ion batteries (LIBs) was the research object, and a specific analysis of the recycling treatment and gradual use stages of power batteries were based on life cycle assessment. Different battery assessment scenarios were established according to the development of battery recycling in China. The results showed that the secondary use has the optimal performance compared to the full-component physical, pyrometallurgical and hydrometallurgy recycling. The results showed that direct recycling has a GWP of 0.037 kg-CO₂ eq/kg LIB⁻¹, which is lower than others. Secondary use of LIB accounts for the most emission reductions with Global warming (GWP) as 12.134 kg-CO₂ eq/kg LIB⁻¹. The secondary use has the greatest impact on the assessment results, especially in dynamic scenarios. Through a comprehensive comparison of different recycling technologies, the secondary use, increasing the recycling rate, reducing resource, energy consumption and pollution emissions.

1. Introduction

As the core component of electric vehicles (EVs), lithium-ion batteries (LIBs) are widely used and the amount of LIB materials that needs to be extracted, produced and disposed of has increased dramatically (Diouf and Pode, 2015; Liu et al., 2022; Son et al., 2021). When a battery's capacity falls below 80 %, it is retired from the vehicle (Porzio and Scown, 2021). Decommissioned LIBs are environmentally harmful and resource intensive, and will cause serious environmental pollution and resource waste if not recycled (Kang et al., 2013; Kang et al., 2010). Recycling decommissioned LIBs is important for the sustainable development of the new energy vehicle industry (Liang et al., 2020). With the introduction of China's carbon peak and carbon neutral targets, there is also increasing concern about the environmental impact of retired LIBs

(Lai et al., 2022; Larcher and Tarascon, 2015; Oliveira et al., 2015; Peters et al., 2017).

The current treatment methods for used lithium batteries are mainly pyrotechnically recycling, hydrometallurgy recycling and direct recycling (Gaines, 2018; Zhang et al., 2018b). Thermal recycling has high energy consumption and wet recycling produces large amounts of wastewater to pollute the environment, and both methods are not effective in mitigating pollution (Ciez and Whitacre, 2019; Hendrickson et al., 2015). Direct recovery can retain the original chemical structure while recovering the active material (Shi et al., 2018). Direct recycling maximizes the retention of the battery itself, requires minimal addition of new materials to assemble a new battery for secondary use, allows for large-scale recycling and processing, and realizes a fully automated production process that will significantly reduce the energy

Abbreviations: EVs, Electric vehicles; LIBs, lithium-ion batteries; LFP, lithium iron phosphate; NCM, lithium nickel cobalt manganese oxide; LCA, Life Cycle Assessment; LCIA, Life Cycle Inventory Analysis; LCI, life cycle inventory; ESS, energy storage systems; EOL, end-of-life; LAB, lead-acid batteries; CBS, communication base stations; GWP, Global Warming Potential; EF, Environmental Footprint; EC-JRC, European Commission's Joint Research Centre; GWP, Global warming; SOD, Stratospheric ozone depletion; PMF, Fine particulate matter formation; TAP, Terrestrial acidification; FEP, Freshwater eutrophication; FTP, Freshwater ecotoxicity; HTP, Human carcinogenic toxicity; MD, Mineral resource scarcity; FFD, Fossil resource scarcity; FWC, Water consumption.

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consumption and economic costs of the secondary use battery remanufacturing process (Liu et al., 2021; Quan et al., 2022). Therefore, in this paper, the mechanical grinding direct recycling method is chosen as the recycling method for used batteries. Combining the requirements of different application scenarios on battery capacity and safety and economy, the domestic retired electric vehicle batteries are divided into static energy storage systems and dynamic energy storage systems according to the use scenarios when secondary utilization is carried out (Crenna et al., 2021). The battery sub-use scenarios in this paper are also set up based on this classification.

The Life Cycle Assessment (LCA) approach is designed to quantify the environmental impact of a product's impact categories across a range of processes from materials, manufacturing, use and disposal (Finnveden et al., 2009). In recent years, LCA has been widely applied to quantify the carbon emissions and various environmental burdens associated with the manufacturing, use and recycling of LIBs (Feng et al., 2022; Li et al., 2022), and is used to guide battery manufacturing and recycling processes and process improvements, product development and macro-level decision-making. However, most studies have focused on the evaluation of carbon emissions and environmental indicators in the production phase of batteries, or the LCA of batteries throughout their life cycle, but less research has been conducted on the environmental impacts of material recycling and secondary use of used batteries (Lai et al., 2022).

Most of the research related to the evaluation of power battery life has focused on the environmental impact of the battery production process, while not much research has been done on the battery recycling process, and some studies have even ignored the environmental impact of the recycling process. At present, power battery recycling technologies at home and abroad are not yet mature, making it difficult for researchers to obtain actual production data, and therefore no in-depth studies have been conducted. However, the reasonable recycling of power batteries can largely reduce the environmental burden of the entire life cycle of batteries. Wang and Yu (Wang and Yu, 2021) speculated on the environmental impact of LIBs using a life cycle evaluation method and found that the environmental impact of LIBs is well controlled over the life cycle when they are properly recycled. Quan et al. (Quan et al., 2022) also quantified and compared the environmental impact of lithium iron phosphate (LFP) batteries and lithium nickel cobalt manganese oxide (NCM) batteries over the life cycle and Dewulf et al. (Dewulf et al., 2010) compared the environmental impact of using virgin materials versus recycled materials as production materials and found that the use of recycled materials could reduce energy consumption by approximately 50 %. The findings of Jiang et al. (Jiang et al., 2022) showed that the extraction, processing and use stages of battery materials played a dominant role in the overall environmental performance impact. Meanwhile the environmental impact of batteries during the manufacturing and transportation stages was relatively small, but battery recycling could significantly increase the environmental benefits.

Low-cost recycling methods for direct LIB recovery are highly environmentally competitive for specific battery types (Ciez and Whitacre, 2019; Larouche et al., 2020; Weber et al., 2018). With the development of recycled lithium technology and the increasing demand for virgin materials for battery production, high prices and depleted virgin resources are becoming a huge problem. As a result, there is a need for greater use of recyclable materials in batteries (McManus, 2012). In addition, potential solid waste residues, wastewater contaminants and secondary toxic emissions from primary battery manufacturing can all be released through recycling. There are also a few barriers to current battery recycling, including: inadequate collection networks, unsatisfactory processing capacity, and incomplete recycling methods, which hinder the recycling of waste LIBs, and improper disposal of waste LIBs can lead to explosions (Gu et al., 2017; Huang et al., 2018; Wang and Wu, 2017). Improving the environmental efficiency of the battery manufacturing process through LCA analysis can show the high

environmental feasibility of using waste EV LIBs as ESS (energy storage systems) to replace LAB (lead-acid batteries) in CBS (communication base stations) (Sanfelix et al., 2015; Wu and Kong, 2018; Yan et al., 2020). The reuse of retired LIBs in less demanding systems such as peaking, renewable energy storage and exchange power plants is also a favorable option for end-of-life (EOL) management (Zhang et al., 2018a).

In the literature based on LCA studies, batteries with high energy density and long life have been found to have a low negative impact on the environment (Majeau-Bettez et al., 2011; Yu et al., 2014). Although various LCA studies have evaluated LBS, these analyses have found differences from previous studies (Hawkins et al., 2013; Sanfelix et al., 2015; Wang et al., 2020). Most of the available studies focus on a limited type of battery and all utilize their own techniques to assess their impact, with significant uncertainty associated with the data and results (Dai et al., 2019; Ellingsen et al., 2017; Peters et al., 2017). First, for background data, most of these studies used secondary Life Cycle Inventory (LCI) databases, non-uniform LCI databases or literature publications as data sources. Furthermore, for foreground data, most studies were based on previous literature publications, engineering calculations and secondary data, and therefore did not reflect current commercial-scale automotive LIB production. Furthermore, for life cycle stages, most studies focus only on production (cradle to gate), while only a few studies explicitly assess the EOL stage. It is therefore important in the Chinese context to assess the life-cycle environmental impacts of retired LIBs using primary life-cycle data and to identify the potential for reducing the environmental impacts of LIBs.

The aim of this study is to address the environmental aspects of recycling LIBs throughout their life cycle by presenting an innovative life cycle model of retired electric vehicle lithium batteries. The regenerative manufacturing phase, the secondary use phase and the recycling phase are all considered within the system boundary. For the reuse and recycling stages, prospective data are obtained from typical Chinese companies. In this study, retired EV batteries were defined as standard commercial type LIBs and recycling was judged based on the degree of battery capacity decay. At the same time, two different recycling processes were considered for the retired batteries, direct recycling of all of them and for secondary use. The aim is to further explore the environmental performance of secondary use recycling technologies. In addition, a complete and transparent life cycle inventory is provided for all life cycle stages. The results of the LCA will help to make a judgement on secondary use in the ESS. The comparative analysis of the environmental impacts associated with the recycling stages will help governments and businesses to select and promote recycling technologies and provide decision-making and technical support for related green manufacturing and smart recycling of materials to help the LIBs industry develop more sustainably.

2. Materials and methods

2.1. Scope and functional unit

The purpose of this study was to conduct an LCA study of battery recycling to determine the environmental impact of the LIB recycling process in China. The Life Cycle Inventory Analysis (LCIA) was analyzed and calculated using SimaPro, one of the most commonly used LCA software. It enables easy modelling in a way that the analysis is systematic and transparent, and the topical issues in the results are visible (Sinha et al., 2016). The software not only uses fast algorithms that are flexible and efficient, but also allows customized additions to the background databases. With these powerful computational features and a comprehensive coverage of background databases, SimaPro can ensure the accuracy and reliability of the LCA calculations.

The functional unit is defined as processing 1 kg of decommissioned batteries and performing an analysis from the recycling plant to the recovered material. According to ReCiPe 2016 midpoint method, Global

Warming Potential (GWP) (kg-CO₂ eq) is used to compare environmental impacts between different processes and stages (Rey et al., 2021). In this paper, retired batteries of electric vehicles are selected as the research object, and all data collected need to be converted into successful energy units to ensure comparability of results. It directly affects the accuracy of calculation results.

The data needed for the calculations are mainly from publicly available data from companies within China. The processes of battery recycling, remanufacturing, secondary use, and material recovery are modeled based on the real material required by the companies. EV battery recycling companies in China whose recycling processes comply with the national environmental standards and have an average annual treatment capacity of more than 1,000 tonnes of waste batteries were selected. Specific recycling methods and processes are described in the supporting materials. The calculations use data from the public reports of Hefei Guoxuan Gaoke Power Energy Co. and Guangdong Jiecheng New Energy Environmental Protection Technology Co. The specific values of substances and energy used during the treatment of batteries were shown in the tables of the supporting materials (Table S4–S6), and the process of battery treatment was shown in the pictures of the supporting materials (Fig. S1, S2). Fig. 1 showed the main components of the batteries used in the calculations and the specific proportion of each material.

2.2. System boundary

In this study, waste batteries are post-use decommissioned products and the production and primary use of the batteries are not included in the system. The system includes not only the material recycling process of the batteries, but also the secondary use process of the batteries. Therefore, the scope of this study is not only to recycle waste electrode materials from batteries, but also plastics and materials such as copper foil, aluminum foil, and steel cases. Sensitivity analysis was used to assess the effect of the quantity ratio of the secondary batteries on the results. The boundaries and diagrams of the battery recycling process are shown in Fig. 2. The treatment process for retired batteries requires an external supply of 333,000 kWh of electricity per year to ensure that the process can always continue. 10,000 tons of retired batteries from electric vehicles can be collected each year, of which only 4,000 tons

meet the capacity requirements for secondary use and can be reassembled for secondary use in ESS, the rest of the batteries are directly recycled for material recovery. This paper is based on calculations and assumptions based on the annual recycling levels of retired domestic EV batteries at this stage and actual data from the last three years of market conditions, and the data for battery recycling has been collected for actual usage within China.

2.3. Life cycle inventory analysis

2.3.1. Libs remanufacturing

The energy required for the remanufacturing of the batteries is measured based on the actual situation in the factory. After the used batteries have been tested for battery health, batteries with more than 60 % of their capacity were newly assembled and manufactured to become energy storage batteries ready for use in the ESS. During the reassembly process, new materials such as connecting tabs, wire harnesses and shell were added (Fig. S1). To suit different ESS scenarios, the finished batteries were manufactured in packs and modules.

2.3.2. Libs recycling

The data for direct mechanical handling of physical recycling was derived from actual production data from domestic companies and the process was showed in Figure S2. Decommissioned batteries, which could not be subjected to secondary use, are automatically dismantled in the factory to recover metal parts, plastics and electrode materials. The batteries were thoroughly discharged by immersion in a 5 % sodium chloride solution for 12 h to ensure the safety of the subsequent processing. After the batteries have been crushed at various levels, the electrode material of different sizes was collected by sorting. After the above system, the waste LIBs were dismantled into waste electrode material, waste copper foil, waste aluminum foil, waste plastic diaphragm and waste metal shell. The recovered graphite, spacer and electrolyte conditioning components could also be reused after repair to produce new batteries, enabling the recycling and reuse of all LIBs components. Table S5–6 detailed the input and output inventory data for materials, energy and emissions.

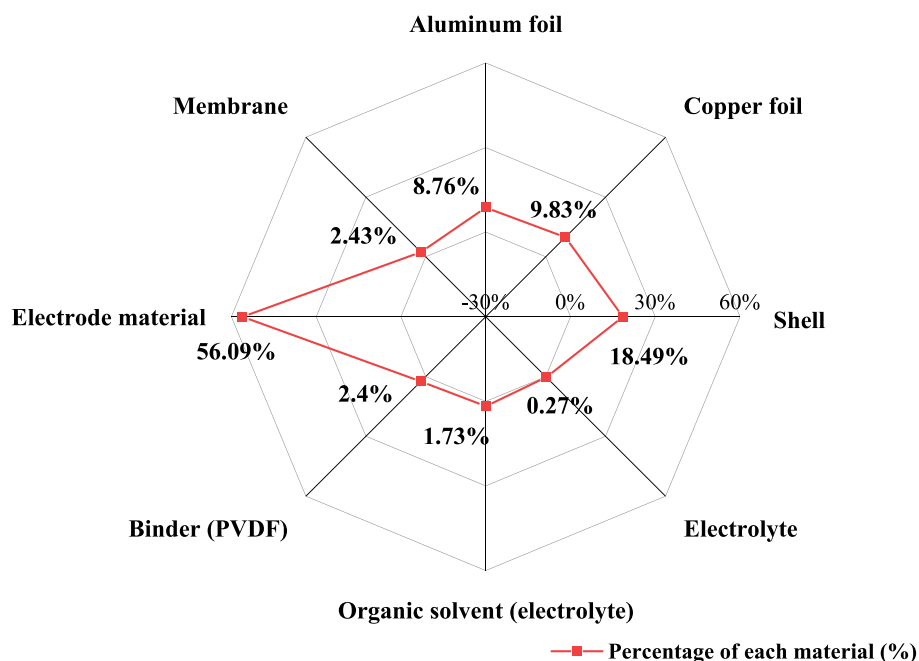


Fig. 1. Components and percentages of the battery.

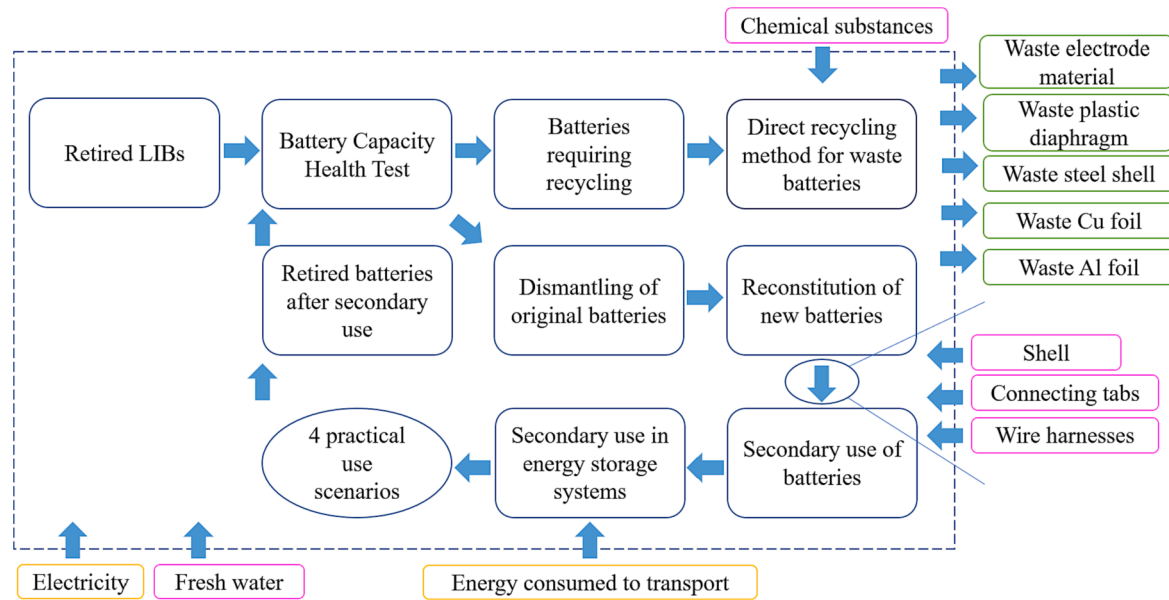


Fig. 2. Technical system boundary used in the study.

2.4. Secondary use management scenarios

In the secondary use phase of batteries, five scenarios were considered in this study to assess the environmental benefits of recycling decommissioned batteries under different use paths in ESS. The secondary use batteries in this study are all state uniform batteries that have been processed by recycling plants according to a hierarchy of battery states. Due to the different requirements of safety and capacity for different scenarios of secondary utilization of batteries, the use scenarios are mainly divided into two categories: stationary scenarios and mobile scenarios (Cusenza et al., 2019; Hiremath et al., 2015). According to the actual situation in China, four scenarios of gradient use were selected to ensure smaller capacity of batteries, safe and reliable use environment, better economy and better commercialization prospect (Geng et al., 2022). The stationary use scenario chose light storage charging station and communication base stations (Scenarios 2 and 3), and the mobile scenario chose mobile charging vehicle and courier tricycles (Scenarios 4 and 5). Recycling without secondary use served as a reference case (Scenario 1) and was compared with the cases that include secondary use before recycling. According to what has been carried out in China now in the ESS using secondary batteries is the actual situation, setting up 4 situations for comparison, where the address of the battery recycling plant is in Hefei, while the 100 KWh optical energy storage charging station base is in Nanjing, 30 KWh communication base station is located in Kunming, 109 KWh of electricity in mobile charging vehicles comes from secondary batteries, and express tricycle vehicles currently with the storage capacity of 3 KWh are in operation. These data have been analyzed and calculated using a LCA analysis model.

Scenario 1 (SCE-1): It is assumed that all retired batteries entering the recycling plant are tested to less than 60 % capacity and are all discarded for direct recycling by mechanical grinding, with the resulting electrode powder being recycled for disposal.

Scenario 2 (SCE-2): The retired batteries in the recycling plant that meet the conditions for secondary use are reassembled and manufactured into new energy storage batteries, and according to the actual production data we can get that the batteries that can be used for secondary use account for 40 % of the total number of batteries (Gu et al., 2018; Gu et al., 2021; Yoo and Park, 2019).

Scenario 3 (SCE-3): Under the condition that the various indicators of the recovered retired batteries remain unchanged, it is assumed that the batteries that can be used for secondary use are all used in the energy

storage system of the communication base station.

Scenario 4 (SCE-4): Assuming no change in battery recycling, all batteries that can be reused are used in mobile charging vehicles, replacing half of the original battery pack in the vehicle's powertrain.

Scenario 5 (SCE-5): If the situation of retired batteries received is the same as in Scenario 2, the batteries that can be reconstituted for secondary use are used in the courier tricycle as a supplement to the power supply.

In order to investigate the environmental impact of retired power batteries more fully in different scenarios, both mobile and stationary, a sensitivity analysis was carried out by varying the proportion of batteries used in scenarios 2 and 4. In order to make the difference between the use of reconstituted batteries in stationary and mobile scenarios as clear as possible, a floating range of $\pm 25\%$ was chosen as the adjustment.

2.5. Impact assessment categories

In the LCIA, the midpoint model quantifies environmental impacts by linking the use and emissions of natural resources to impacts on the environment. This effect is calculated using specific factors known as representational factors. Here, we applied the Environmental Footprint (EF) method v.3.0, provided in SimaPro v.9.0.0.48, developed by the European Commission's Joint Research Centre (EC-JRC) (Golsteijn and Vieira, 2020). The midpoint level effects of the following 10 impact categories were analyzed: Global warming (GWP), Stratospheric ozone depletion (SOD), Fine particulate matter formation (PMF), Terrestrial acidification (TAP), Freshwater eutrophication (FEP), Freshwater ecotoxicity (FTP), Human carcinogenic toxicity (HTP), Mineral resource scarcity (MD), Fossil resource scarcity (FFD), Water consumption (FWC).

3. Results and discussion

3.1. GWP of LIB recycling

Midpoint LCA provides a broad understanding of the environmental impact of the battery recycling process. We first focus on the impact of GWP as it is a simple and effective way to make cross-sectional comparisons with previous studies. The results in Fig. 3(a) are reported based on 1 kg of recycled waste LIBs. In SCE-1, a maximum value of

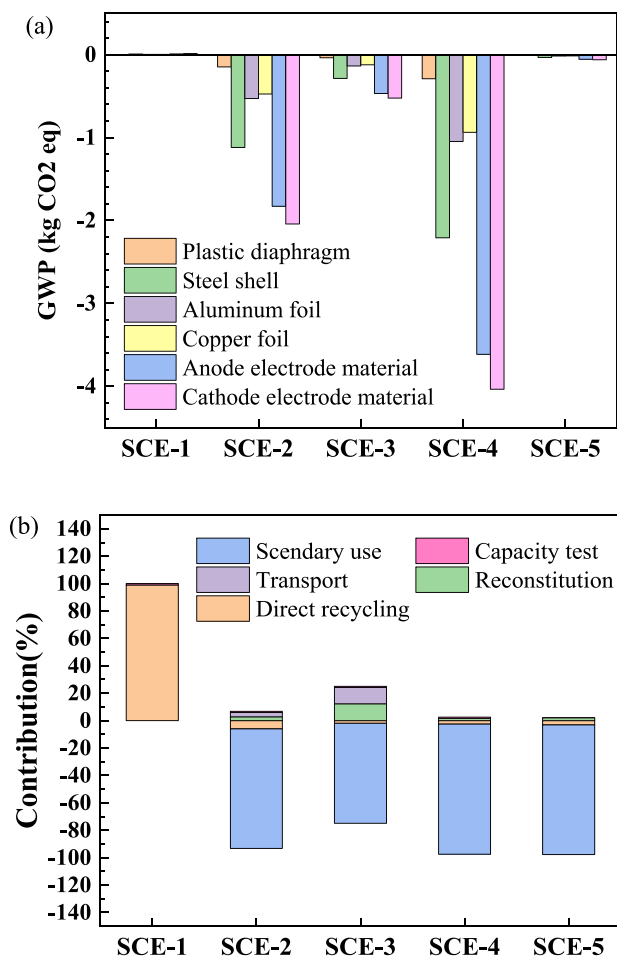


Fig. 3. Comparison of environmental emissions per kg of retired battery: (a) GWP for 5 scenarios, (b) comparison of GWP for direct disposal and secondary use for stages.

0.037 kg-CO₂ eq·kg LIB⁻¹ was obtained for GWP, while all other scenarios recovered negative GWP values with a negative impact on global temperature rise, which certainly indicates that the secondary post-recycling approach is environmentally competitive compared to direct LIB recovery. SCE-4 has the lowest GWP value of $-12.14 \text{ kg-CO}_2 \text{ eq·kg LIB}^{-1}$ due to the secondary use without the energy consumption of transporting the batteries over long distances to the stationary scenario, as well as the high battery use of the rechargeable vehicle, which generates the highest energy efficiency and can avoid manufacturing the highest number of brand-new LIBs used.

In the published literature (Fig. 4), batteries are pyrotechnically recycled by the emerging direct current plasma pyrometallurgy technology, which emits 1100 kg-CO₂ eq per ton of LIB, while the use of recycled material avoids the emission of 1220 kg-CO₂ eq per ton of LIB (Rajaeifar et al., 2021). Other researchers have assumed that the batteries are all wet recycled and that the recovered material is used in the manufacture of new batteries, the EOL stage avoids the emission of 30.91 kg-CO₂ eq·kWh LIB⁻¹ to the outside environment (Sun et al., 2020). Other researchers' analyses have shown that using the more advanced hydrometallurgical technologies available at this stage, without using the recovered material for battery production, would still result in 79.90 kg-CO₂ eq·kWh LIB⁻¹ (Wang et al., 2022). Tao et al. (Tao et al., 2023) studied the environmental effects of pyro-hydro metallurgical methods for battery recycling and showed that direct treatment of the retired batteries would have an impact of 34.3 kg-CO₂ eq·kWh LIB⁻¹ and that recovery of the material would reduce the impact of emissions to the environment by 19.6 kg-CO₂ eq·kWh LIB⁻¹.

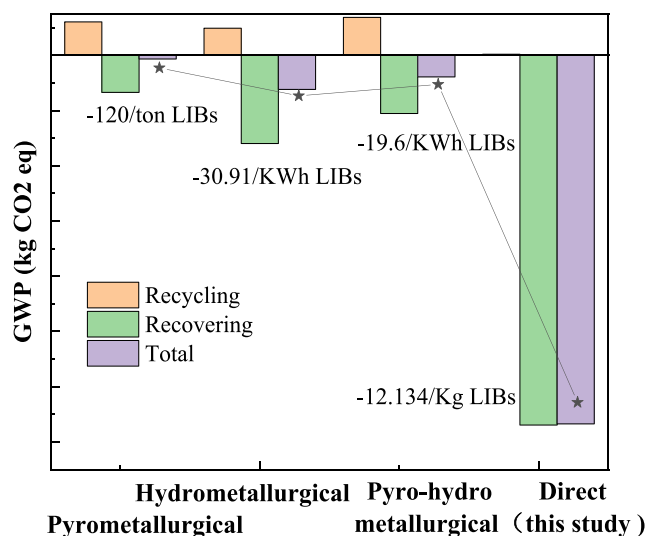


Fig. 4. Comparison of the results of treating batteries using different methods with recycling and recovering material.

A comparison of pyrotechnically and hydrometallurgy recycling of battery materials shows that the recycling process alone does not reduce energy consumption and that secondary use of the recovered material is required to achieve a reduction in carbon emissions. At the same time direct recycling is more environmentally friendly as it emits less CO₂ into the environment than the other two methods. The mechanical grinding process for waste batteries is highly automated, energy efficient, has a high recovery rate and is a cleaner treatment method with a lower GWP than other battery recycling methods.

As can be seen in Fig. 3(b), the secondary use of batteries in the ESS is key to reducing carbon emissions. The large difference in energy consumption from battery transport in SCE-2 and SCE-3 is due to the large difference in transport distance, but the energy consumption from transport has little impact on the total emissions. Compared to SCE-4 and SCE-5, where there are no emissions from transport, the GWP values for the whole process are much lower, suggesting that secondary use can be used more often as an energy storage device in battery packs for electric vehicles in the future. In each case a certain amount of energy is consumed for battery testing and the combination of automated testing and automated recovery processes will reduce CO₂ emissions.

It can be concluded that the use of recycled batteries in ESS is the best way to maximize the use of battery resources and that the use of secondary use batteries in electric vehicle battery packs is a new direction that can be further developed in the future.

3.2. Other impacts in 5 scenarios

In the comparison of the other environmental evaluation indicators in Fig. 5, the SCE-1 scenario has a smaller value for the environmental impact indicator, but also places a burden on the external environment. In contrast, the environmental burdens in SCE 2–5, where secondary use is carried out, are all negative, with no output to the environment but rather new output avoided, reducing the generation of various pollutants. The secondary use of batteries reduces the generation of various pollutants during the production and manufacture of the original battery, while maximizing the use of the battery and making better use of the battery storage resources, reducing the waste of resources.

The environmental evaluation of the secondary use in 4 scenarios can be found to be in line with the order of $4 > 2 > 3 > 5$. SCE-2 and SCE-4 have a greater generation of electrical energy from battery use than the other two, indicating that secondary battery substitution of electrical energy is the main influencing factor in avoiding environmental impacts. The efficiency of the generation of replacement electricity is in

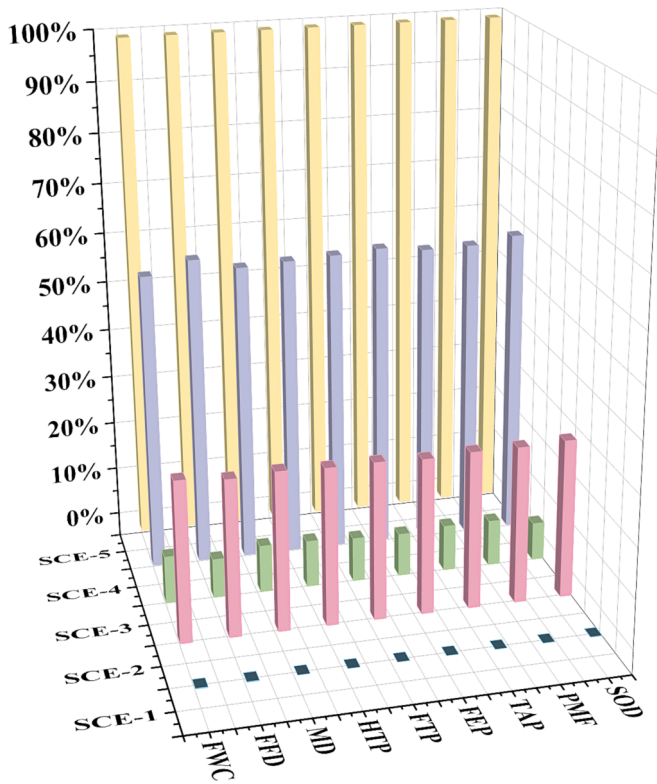


Fig. 5. Comparison of different environmental impact indicators in 5 scenarios.

turn very relevant to the battery technology of the scenarios used, so it is vital to vigorously develop the technology of secondary use batteries in the ESS. Secondary use batteries not only reduce pollution emissions and protect the environment but are also very effective in protecting soil and

water quality. The use of chemical products and the landfilling of metals in the waste phase of batteries can cause non-negligible environmental pollution. At the same time the toxic substances in the batteries are disposed of and then buried, and the effects on the human body cannot be avoided. The secondary use of batteries can reduce the production and emission of toxic substances, which has a very good protective effect on human health.

The most significant reduction in environmental impact is shown for SCE-2 and SCE-4, indicating that the use of secondary batteries in both dynamic and static ESS systems can be a good way to avoid the use of new materials and reduce emissions of harmful substances and energy consumption, but the environmental benefits are better in SCE- 2. The environmental impact results for SCE-1 to SCE-5 are shown in Table 1 of the supporting material, with FFD, MD and HTP having the most significant impact, indicating that the secondary use of batteries can be a good resource saver. Much of the other literature focuses on ozone pollution and arithmetic pollution from the recycling process, but not enough on human health (Li et al., 2021; Reinhardt et al., 2019). In the calculated results, the secondary use of batteries in ESS can largely reduce the emission of toxic substances in the batteries, which is more beneficial to human health.

3.3. Sensitivity analyses

The results of the sensitivity analysis are shown in Fig. 6 for a $\pm 25\%$ variation in secondary use battery mass for SCE-2 and SCE-4. It was found that the number of batteries used in different scenarios was sensitive to the environmental impact results. Compared to other studies (Ellingsen et al., 2014; Kim et al., 2016), the energy consumption of the waste disposal phase of the batteries recovered in this paper is low (Dai et al., 2019). Under current conditions, a $\pm 25\%$ change in the mass adjustment of the secondary use battery can significantly change the energy intensity of the recycled battery process. The reduction in environmental impact is greater with the increased mass of batteries used in SCE-4 compared to SCE-2 only. Used alone in SCE-4 compared to SCE-2

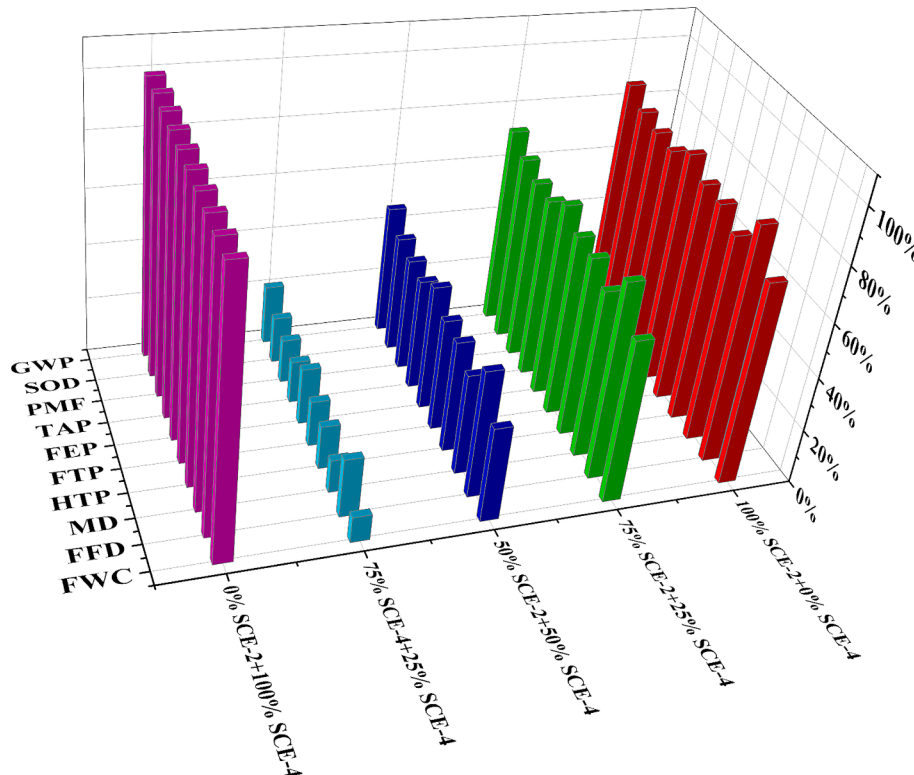


Fig. 6. The effect on battery recycling for SCE-2 and SCE-4 with different percentages.

The change is almost 90 % with a ± 25 % change in mass and approximately ± 30 % change in environmental assessment. By strongly promoting the use of secondary use batteries in mobile charging vehicles, it may be possible to successfully reduce the environmental burden of LIBs. The strong development of the use of secondary batteries in the power supply of LIBs could well avoid the incomplete consumption of battery energy. Also, the use of retired batteries as EVs in EVs could avoid the mismatch of battery packs or battery modules, also as the power output of trams, and the energy consumption of point eating would be more complete. Secondary recycling and collection will also be a new issue as communication base stations and charging pads require the batteries to be transported to a fixed location. There is therefore a need to increase research into the secondary use of lithium batteries in the power supply of low-speed electric vehicles. The life cycle impact can be significantly reduced by improving battery technology and increasing the efficiency of charging and discharging during the use phase.

4. Conclusion

A comparative life cycle assessment study was conducted on the environmental impacts of standard commercial lithium batteries from battery collection to powder recycling based on retired electric vehicles in the Chinese market. The life cycle lists studied for waste battery disposal and secondary battery assembly are disclosed for future comparison. Environmental impacts are grouped into 10 indicators, such as global warming potential, human toxicity or acidification potential, because they provide more detailed information on the corresponding environmental burden associated with the final disposal of waste batteries and the corresponding environmental burden associated with the secondary use of LIBs.

The results show that secondary use of decommissioned batteries in the ESS can reduce the environmental impact of the entire battery by a minimum of five times (e.g., global warming potential between 53 and 248 kg-CO₂ eq/kg LIB⁻¹). Interestingly, the use of secondary batteries to replace some of the batteries in mobile charging vehicles has the least environmental impact, suggesting that the use of secondary batteries can effectively reduce the use of new batteries. Partial use of secondary batteries has less impact on the environment than using brand new LIBs. In addition, although the technology of using secondary use batteries in fixed communication base stations or light-energy storage and charging stations has reached the popularization level, the obtained LCA results clearly show that the use of secondary use batteries in the battery power system is more able to make full use of the battery's own energy and generate more electric energy instead. Accordingly, a sensitivity analysis of reducing battery usage in fixed scenarios was carried out and it was revealed that the environmental burden could be reduced by up to 90 % simply by limiting the number of batteries used in fixed base stations. For the battery recycling industry, it should not simply be about recycling raw metal materials, but about increasing the secondary use of decommissioned batteries for electric vehicles. A stable supply chain and testing standards for secondary use batteries should be formed within the industry as soon as possible to facilitate more companies to recycle batteries and manufacture batteries with the same fixed specifications and quality as the original batteries. For the government, it should strongly advocate and promote the use of secondary batteries to replace some of the original brand-new batteries and ensure that the remanufactured batteries can be used. The public should also respond positively to the government's call so that the battery recycling industry can continue to grow.

The results are particularly significant considering that the technical maturity of secondary batteries for power batteries is relatively low compared to that of secondary batteries for fixed base stations, leaving room for substantial improvement in environmental performance. Overall, this study will support further follow-up studies focusing on the key aspects to be considered for low environmental impact LIB recycling

to provide guidance for promoting the commercialization of green LIBs.

CRediT authorship contribution statement

Hanxue Yang: Data curation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **Xiaocheng Hu:** Formal analysis, Methodology, Writing – review & editing. **Guanhua Zhang:** Conceptualization, Funding acquisition, Project administration, Resources, Supervision, Writing – review & editing, Writing – original draft. **Binlin Dou:** Writing – review & editing. **Guomin Cui:** Writing – review & editing. **Qiguo Yang:** Writing – review & editing. **Xiaoyu Yan:** Conceptualization, Funding acquisition, Project administration, Software, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.wasman.2024.02.034>.

References

- Ciez, R.E., Whitacre, J.F., 2019. Examining different recycling processes for lithium-ion batteries. *Nat. Sustainability* 2, 148–156.
- Crenna, E., Gauch, M., Widmer, R., Waeger, P., Hirschier, R., 2021. Towards more flexibility and transparency in life cycle inventories for Lithium-ion batteries. *Resour. Conserv. Recycl.* 170.
- Cusenza, M.A., Guarino, F., Longo, S., Mistretta, M., Cellura, M., 2019. Reuse of electric vehicle batteries in buildings: An integrated load match analysis and life cycle assessment approach. *Energ. Buildings* 186, 339–354.
- Dai, Q., Kelly, J.C., Gaines, L., Wang, M., 2019. Life Cycle Analysis of Lithium-Ion Batteries for Automotive Applications. *Batteries-Basel* 5.
- Dewulf, J., Van der Vorst, G., Denturck, K., Van Langenhove, H., Ghyoot, W., Tytgat, J., Vandepitte, K., 2010. Recycling rechargeable lithium ion batteries: Critical analysis of natural resource savings. *Resour. Conserv. Recycl.* 54, 229–234.
- Diouf, B., Pode, R., 2015. Potential of lithium-ion batteries in renewable energy. *Renew. Energy* 76, 375–380.
- Ellingsen, L.-A.-W., Majeau-Bettez, G., Singh, B., Srivastava, A.K., Valoen, L.O., Stromman, A.H., 2014. Life Cycle Assessment of a Lithium-Ion Battery Vehicle Pack. *J. Ind. Ecol.* 18, 113–124.
- Ellingsen, L.-A.-W., Hung, C.R., Stromman, A.H., 2017. Identifying key assumptions and differences in life cycle assessment studies of lithium-ion traction batteries with focus on greenhouse gas emissions. *Transportation Research Part D-Transport and Environment* 55, 82–90.
- Feng, T., Guo, W., Li, Q., Meng, Z., Liang, W., 2022. Life cycle assessment of lithium nickel cobalt manganese oxide batteries and lithium iron phosphate batteries for electric vehicles in China. *J. Storage Mater.* 52.
- Finnveden, G., Hauschild, M.Z., Ekvall, T., Guinee, J., Heijungs, R., Hellweg, S., Koehler, A., Pennington, D., Suh, S., 2009. Recent developments in Life Cycle Assessment. *J. Environ. Manage.* 91, 1–21.
- Gaines, L., 2018. Lithium-ion battery recycling processes: Research towards a sustainable course. *Sustainable. Mater. Technol.* 17.
- Geng, J., Gao, S., Sun, X., Liu, Z., Zhao, F., Hao, H., 2022. Potential of electric vehicle batteries second use in energy storage systems: The case of China. *Energy* 253.

- Golsteijn, L., Vieira, M., 2020. Applicability of the European Environmental Footprint (EF) methodology in Southern Mediterranean countries—learnings and recommendations for enabling EF-compliant studies in regions outside of Europe. *Int. J. Life Cycle Assess.* 25, 2407–2416.
- Gu, F., Guo, J., Yao, X., Summers, P.A., Widiyatmoko, S.D., Hall, P., 2017. An investigation of the current status of recycling spent lithium-ion batteries from consumer electronics in China. *J. Clean. Prod.* 161, 765–780.
- Gu, X., Ieromonachou, P., Zhou, L., Tseng, M.-L., 2018. Developing pricing strategy to optimise total profits in an electric vehicle battery closed loop supply chain. *J. Clean. Prod.* 203, 376–385.
- Gu, X., Zhou, L., Huang, H., Shi, X., Ieromonachou, P., 2021. Electric vehicle battery secondary use under government subsidy: A closed-loop supply chain perspective. *Int. J. Prod. Econ.* 234, 108035.
- Hawkins, T.R., Singh, B., Majeau-Bettez, G., Stromman, A.H., 2013. Comparative Environmental Life Cycle Assessment of Conventional and Electric Vehicles. *J. Ind. Ecol.* 17, 53–64.
- Hendrickson, T.P., Kavvada, O., Shah, N., Sathre, R., Scown, C.D., 2015. Life-cycle implications and supply chain logistics of electric vehicle battery recycling in California. *Environ. Res. Lett.* 10.
- Hiremath, M., Derendorf, K., Vogt, T., 2015. Comparative Life Cycle Assessment of Battery Storage Systems for Stationary Applications. *Environ. Sci. Tech.* 49, 4825–4833.
- Huang, B., Pan, Z., Su, X., An, L., 2018. Recycling of lithium-ion batteries: Recent advances and perspectives. *J. Power Sources* 399, 274–286.
- Jiang, S., Hua, H., Zhang, L., Liu, X., Wu, H., Yuan, Z., 2022. Environmental impacts of hydrometallurgical recycling and reusing for manufacturing of lithium-ion traction batteries in China. *Sci. Total Environ.* 811, 152224.
- Kang, D.H.P., Chen, M., Ogunseitan, O.A., 2013. Potential Environmental and Human Health Impacts of Rechargeable Lithium Batteries in Electronic Waste. *Environ. Sci. Tech.* 47, 5495–5503.
- Kang, J., Sohn, J., Chang, H., Senanayake, G., Shin, S.M., 2010. Preparation of cobalt oxide from concentrated cathode material of spent lithium ion batteries by hydrometallurgical method. *Adv. Powder Technol.* 21, 175–179.
- Kim, H.C., Wallington, T.J., Arsenault, R., Bae, C., Ahn, S., Lee, J., 2016. Cradle-to-Gate Emissions from a Commercial Electric Vehicle Li-Ion Battery: A Comparative Analysis. *Environ. Sci. Tech.* 50, 7715–7722.
- Lai, X., Chen, Q., Tang, X., Zhou, Y., Gao, F., Guo, Y., Bhagat, R., Zheng, Y., 2022. Critical review of life cycle assessment of lithium-ion batteries for electric vehicles: A lifespan perspective. *Etransportation* 12.
- Larcher, D., Tarascon, J.M., 2015. Towards greener and more sustainable batteries for electrical energy storage. *Nat. Chem.* 7, 19–29.
- Larouche, F., Tedjar, F., Amouzegar, K., Houlachi, G., Bouchard, P., Demopoulos, G.P., Zaghib, K., 2020. Progress and Status of Hydrometallurgical and Direct Recycling of Li-Ion Batteries and Beyond. *Materials* 13.
- Li, Y., Lv, W., Huang, H., Yan, W., Li, X., Ning, P., Cao, H., Sun, Z., 2021. Recycling of spent lithium-ion batteries in view of green chemistry. *Green Chem.* 23, 6139–6171.
- Li, P., Xia, X., Guo, J., 2022. A review of the life cycle carbon footprint of electric vehicle batteries. *Separation and Purification Technology* 296.
- Liang, Q., Yue, H., Wang, S., Yang, S., Lam, K.-H., Hou, X., 2020. Recycling and crystal regeneration of commercial used LiFePO₄ cathode materials. *Electrochim. Acta* 330.
- Liu, W., Liu, H., Liu, W., Cui, Z., 2021. Life cycle assessment of power batteries used in electric bicycles in China. *Renew. Sustain. Energy Rev.* 139.
- Liu, Y.-K., Zhao, C.-Z., Du, J., Zhang, X.-Q., Chen, A.-B., Zhang, Q., 2022. Research Progresses of Liquid Electrolytes in Lithium-Ion Batteries. *Small*.
- Majeau-Bettez, G., Hawkins, T.R., Stromman, A.H., 2011. Life Cycle Environmental Assessment of Lithium-Ion and Nickel Metal Hydride Batteries for Plug-In Hybrid and Battery Electric Vehicles. *Environ. Sci. Tech.* 45, 4548–4554.
- McManus, M.C., 2012. Environmental consequences of the use of batteries in low carbon systems: The impact of battery production. *Appl. Energy* 93, 288–295.
- Oliveira, L., Messagie, M., Rangaraju, S., Sanfeli, J., Rivas, M.H., Van Mierlo, J., 2015. Key issues of lithium-ion batteries - from resource depletion to environmental performance indicators. *J. Clean. Prod.* 108, 354–362.
- Peters, J.F., Baumann, M., Zimmermann, B., Braun, J., Weil, M., 2017. The environmental impact of Li-Ion batteries and the role of key parameters - A review. *Renew. Sustain. Energy Rev.* 67, 491–506.
- Porzio, J., Scown, C.D., 2021. Life-Cycle Assessment Considerations for Batteries and Battery Materials. *Adv. Energy Mater.* 11.
- Quan, J., Zhao, S., Song, D., Wang, T., He, W., Li, G., 2022. Comparative life cycle assessment of LFP and NCM batteries including the secondary use and different recycling technologies. *Sci. Total Environ.* 819.
- Rajaeifar, M.A., Raugel, M., Steubing, B., Hartwell, A., Anderson, P.A., Heidrich, O., 2021. Life cycle assessment of lithium-ion battery recycling using pyrometallurgical technologies. *J. Ind. Ecol.* 25, 1560–1571.
- Reinhardt, R., Christodoulou, L., Gasso-Domingo, S., Amante Garcia, B., 2019. Towards sustainable business models for electric vehicle battery second use: A critical review. *J. Environ. Manage.* 245, 432–446.
- Rey, I., Vallejo, C., Santiago, G., Iturrondobetia, M., Lizundia, E., 2021. Environmental Impacts of Graphite Recycling from Spent Lithium-Ion Batteries Based on Life Cycle Assessment. *ACS Sustain. Chem. Eng.* 9, 14488–14501.
- Sanfeli, J., Messagie, M., Omar, N., Van Mierlo, J., Hennige, V., 2015. Environmental performance of advanced hybrid energy storage systems for electric vehicle applications. *Appl. Energy* 137, 925–930.
- Shi, Y., Chen, G., Liu, F., Yue, X., Chen, Z., 2018. Resolving the Compositional and Structural Defects of Degraded LiNi_{0.8}Co_{0.1}Mn_{0.1}O₂ Particles to Directly Regenerate High-Performance Lithium-Ion Battery Cathodes. *ACS Energy Lett.* 3, 1683–1692.
- Sinha, R., Lennartsson, M., Frostell, B., 2016. Environmental footprint assessment of building structures: A comparative study. *Build. Environ.* 104, 162–171.
- Son, D.-K., Kim, J., Raj, M.R., Lee, G., 2021. Elucidating the structural redox behaviors of nanostructured expanded graphite anodes toward fast-charging and high-performance lithium-ion batteries. *Carbon* 175, 187–201.
- Sun, X., Luo, X., Zhang, Z., Meng, F., Yang, J., 2020. Life cycle assessment of lithium nickel cobalt manganese oxide (NCM) batteries for electric passenger vehicles. *J. Clean. Prod.* 273.
- Tao, Y., Wang, Z., Wu, B., Tang, Y., Evans, S., 2023. Environmental life cycle assessment of recycling technologies for ternary lithium-ion batteries. *J. Clean. Prod.* 389, 136008.
- Wang, L., Hu, J., Yu, Y., Huang, K., Hu, Y., 2020. Lithium-air, lithium-sulfur, and sodium-ion, which secondary battery category is more environmentally friendly and promising based on footprint family indicators? *J. Clean. Prod.* 276.
- Wang, Y., Tang, B., Shen, M., Wu, Y., Qu, S., Hu, Y., Feng, Y., 2022. Environmental impact assessment of second life and recycling for LiFePO₄ power batteries in China. *J. Environ. Manage.* 314.
- Wang, W., Wu, Y., 2017. An overview of recycling and treatment of spent LiFePO₄ batteries in China. *Resour. Conserv. Recycl.* 127, 233–243.
- Wang, S., Yu, J., 2021. A comparative life cycle assessment on lithium-ion battery: Case study on electric vehicle battery in China considering battery evolution. *Waste Manag. Res.* 39, 156–164.
- Weber, S., Peters, J.F., Baumann, M., Weil, M., 2018. Life Cycle Assessment of a Vanadium Redox Flow Battery. *Environ. Sci. Tech.* 52, 10864–10873.
- Wu, Z., Kong, D., 2018. Comparative life cycle assessment of lithium-ion batteries with lithium metal, silicon nanowire, and graphite anodes. *Clean Techn. Environ. Policy* 20, 1233–1244.
- Yan, W., Cao, H., Zhang, Y., Ning, P., Song, Q., Yang, J., Sun, Z., 2020. Rethinking Chinese supply resilience of critical metals in lithium-ion batteries. *J. Clean. Prod.* 256.
- Yoo, M.J., Park, H.B., 2019. Effect of hydrogen peroxide on properties of graphene oxide in Hummers method. *Carbon* 141, 515–522.
- Yu, Y., Chen, B., Huang, K., Wang, X., Wang, D., 2014. Environmental Impact Assessment and End-of-Life Treatment Policy Analysis for Li-Ion Batteries and Ni-MH Batteries. *Int. J. Environ. Res. Public Health* 11, 3185–3198.
- Zhang, X., Li, L., Fan, E., Xue, Q., Bian, Y., Wu, F., Chen, R., 2018b. Toward sustainable and systematic recycling of spent rechargeable batteries. *Chem. Soc. Rev.* 47.
- Zhang, N., Song, Y., Du, Y., Yuan, Y., Xiao, G., Gui, Y., 2018a. A Novel Solid-Solid Phase Change Material: Pentaglycerine/Expanded Graphite Composite PCMs. *Adv. Eng. Mater.* 20, 1800237.