Recycling Solid-State Batteries: Challenges and Innovations for Sustainable Energy Solutions

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Abstract. The increasing use of electric vehicles (EVs) has led to a growing demand for battery technology, particularly for lithium-ion and solid-state batteries (SSBs). This paper investigates the complexities and potential strategies for recycling SSBs, with a focus on electrolyte materials such as oxides, sulphides, and polymers. Given that conventional recycling methods are limited in their applicability to the unique components of SSBs, specific processes like hydrometallurgical and dissolution techniques show promise, especially for sulphide electrolytes. For oxide-based SSBs, dry processing proves more suitable due to their material characteristics. Multi-stream recycling systems that allow tailored treatment for different battery types are identified as optimal, as they enhance material recovery, yield, and economic viability. The study also discusses legislative and manufacturing considerations, advocating for standardized battery forms, increased collection rates, and energy-efficient production processes to support widespread SSB adoption. Finally, the paper emphasizes that to achieve a sustainable, closed-loop recycling process, further research is needed to assess SSBs' full life cycles and adapt facilities to process emerging battery materials, like lithium-sulfur and sodium-ion technologies.

Keywords: Solid-State Batteries, lithium-ion batteries, electrolyte materials, closed-loop recycling process.

1. Introduction

As a result of the massive population growth over the past few years, the demand for travelling has increased, and the number of vehicles on the roads has become more and more numerous. With this comes a lot of vehicle emissions, which pollute the environment, and the world is becoming increasingly concerned about the environmental pollution caused by vehicle exhaust. In 1951, the total number of vehicles in India was 310000, and by the end of 2005-2006, the total number of vehicles in India increased to 8900000 (Dey & Mehta, 2020). Most conventional automobiles use internal combustion engines as power machinery, whether it is an ignition internal combustion engine represented by petrol engines or a compression ignition internal combustion engine represented by diesel engines, they work on the principle of converting the chemical energy of the fuel into thermal energy, then using the thermal cycle to convert the thermal energy into mechanical energy, and finally through the mechanical system for the effective output of power (Liu & Wang, 2001). Pollution from internal combustion engines mainly consists of carbon monoxide, hydrocarbons, nitrous oxide, and small amounts of sulphur oxides, lead and particulate matter. Motor vehicle emissions account for two-thirds of urban air pollution, which is a large part of the cause of respiratory diseases, asthma, cancer, and other serious life-affecting health disorders. At the same time, vehicle pollution is also responsible for serious problems such as acid rain and global warming (Dey & Mehta, 2020). In this scenario, people have turned to cleaner energy sources that are more environmentally friendly, which has led to the success of the battery market, mainly in the electric vehicle industry. Today, a large amount of research is going into the field of battery technology, and the battery market is worth up to a billion dollars annually. According to a market research report, the global battery hourly rate was $62 billion in 2014 (Mohammadi & Saif, 2023).

Lead-acid, nickel-metal hydride and lithium-ion batteries are common battery types used in electric vehicles. These batteries can be used not only for automotive and traction applications but also for backup and emergency power for electrical equipment, as well as for submarines and uninterruptible power supplies. It can be said that they are very popular types of batteries, and they have low cost and high durability (Mohammadi & Saif, 2023). Among them, lithium-ion batteries have become the main power source for new energy vehicles because of their high energy density, low self-discharge rate, long cycle life and no memory effect. Due to the rapid development of new energy vehicles, the global shipment of lithium batteries has increased nearly 20 times in the past five years. Global demand for lithium-ion batteries in electric vehicles is expected to reach 680 GWh and 1,525 GWh in 2025 and 2030, respectively, from 120 GWh in 2019. To ensure safety, lithium batteries must be decommissioned when they have been used in electric vehicles for five to eight years or when the battery capacity has degraded to between 70 and 80 per cent of its initial capacity. It is expected that from 2021 to 2030, the total weight of lithium batteries retired from electric vehicles will be as high as 12.85 million tonnes globally. These decommissioned lithium-ion batteries are an abundant energy carrier, but they may also place a heavier burden on the environment. In particular, global reserves of key elements such as lithium, cobalt, and nickel in Li-ion are very limited and unevenly distributed, and Li-ion faces resource shortages and supply chain disruptions (Lai et al., 2022). At the same time, the manufacturing and assembly of lithium-ion batteries also require a large amount of energy and release toxic gases, finally, how to recycle these used batteries in a way that minimizes the environmental impact has become a worldwide concern.

Used batteries may have high voltage, which may cause the risk of electric shock. Volatile organic electrolytes and other additives in used batteries can form toxic or corrosive substances, posing a risk to human health, and if used batteries are not handled properly, they can lead to external short-circuiting, which can result in serious fires (Lai et al., 2022). Batteries buried in the soil in the traditional way can cause serious pollution of soil and water bodies. For these reasons, lithium batteries cannot be mass-produced at low cost, and the performance of lithium batteries themselves is approaching its limits (Janek & Zeier, 2023). Potential successor batteries are being investigated, and solid-state batteries are expected to offer higher energy density, faster-charging performance, and greater safety than lithium-ion batteries. However, there is a limited understanding of solid-state batteries in terms of recyclability (Ahuis et al., 2024). So this article focuses on promising SEs and analyses whether recycling solutions at this stage for various solid-state batteries are robust and energy-efficient, with minimal impact on the environment, and at the same time can provide high recycling rates and good quality secondary materials.

2. Case Description

Today, the business, social, and environmental benefits of closed-loop supply chains are widely acknowledged. Companies are increasingly recognizing the importance of managing the entire product life cycle, from manufacturing to use and recycling, through effective closed-loop supply chain management (Liu et al., 2023). BYD has implemented an innovative closed-loop recycling system for electric vehicle (EV) batteries, working in partnership with Shenzhen-based startup Pandpower and Japanese trading giant Itochu. Through this collaboration, used EV batteries are recycled and repurposed into large-scale energy storage systems, which can support industrial backup power and renewable energy projects, such as solar farms. Under this strategy, old batteries from BYD cars—like buses and taxis—are gathered, examined, and converted into power storage units the size of containers. With a capacity of around one megawatt, these units are being offered primarily in Southeast Asian, Australian, and Japanese markets, with aspirations to grow internationally. These recovered battery systems are an affordable option for energy storage since their projected cost is 20–30% less than that of newly manufactured storage units (Crompton, 2021). However, the establishment of a full closed-loop management system requires a high initial investment at the beginning, which is costly and technically demanding, as it requires testing and managing the performance and status of each battery. Moreover, BYD's recycling model is mainly concentrated in China, making it difficult to meet global demand growth. For achieving the U.S. energy transition goal of net-zero carbon emissions by 2050, the Biden Administration has emphasized the critical importance of supporting emerging battery recycling technologies and improving supply chain resilience for key battery materials (CGEP, 2024). Former Tesla CTO JB Straubel established

Redwood Materials, which effectively extracts precious metals like lithium, nickel, and cobalt from spent EV batteries using sophisticated recycling procedures like hydrometallurgy. Redwood Materials is a proponent of the circular economy concept, which aims to minimise environmental consequences and reduce reliance on raw material extraction by recovering these metals and reintroducing them into the supply chain. To guarantee a consistent supply of end-of-life batteries for recycling, the firm has also forged alliances with significant manufacturers such as Ford, Toyota, and Volvo. This supports Redwood's objective of establishing a completely closed-loop system for battery manufacturing and reuse in the United States. However, Redwood Materials faces the risk of an unstable supply of materials, as it relies on the supply of used batteries, but the U.S. market has a limited number of end-of-life batteries and may face a shortage of resources, especially as it expands in the new energy vehicle market. Recycling technologies such as hydrometallurgy have a high rate of return but high initial investment costs, and Redwood Materials also faces greater international competitive pressure. BYD's advantage lies in its total life-cycle management and localised recycling efficiency, while Redwood Materials' advantage lies in its strong government support and high recycling rate. However, both technologies are challenged by high costs and an unstable supply of resources.

3. Analysis of the Recycling of solid-state batteries

Currently known processes for recycling lithium-ion batteries include mechanical, pyrometallurgical and hydrometallurgical processes. In almost all cases, one combines at least two of these three processes to obtain a higher recovery rate. Mechanical methods are designed to separate the battery into two parts, a pure part and a recyclable part, thereby increasing the recovery rate. Pyrometallurgy is a process that recovers valuable fractions like cobalt, nickel, and copper18 by selectively heating and lowering an environment. Battery-grade precursors are recovered and made available by hydrometallurgical procedures, which can be used to the synthesis of novel battery materials (Ahuis et al., 2024). Solid-state batteries hold the promise of higher energy density, faster charge/discharge performance, and greater battery safety than lithium-ion batteries. Since there is limited understanding of the recyclability of solid-state batteries, this section will focus on reviewing the various current recycling methods and policies for solid-state batteries, including re-synthesis technologies as well as direct recycling strategies. The focus will be on valuable solid-state electrolytes, the introduction of which can lead to changes in battery materials, manufacturing processes, and battery characteristics. Different recycling routes for solid-state electrolytes are also considered, including pre-treatment, mechanical and metallurgical processes (Ahuis et al., 2024). The main differences between solid-state and lithium-ion battery recycling are their anode design and the fact that solid-state batteries use a solid electrolyte rather than a liquid electrolyte. There are several conceivable designs for the anodes in SSBs.

Due to polymer SSBs’ low toxicity and less strict safety standards, they enable simpler recycling process equipment. However, due to their strong adhesion between cell components, disassembly down to the electrode level and dry mechanical treatment are not promising. Because the cheap cost of SE makes recycling less viable for industrial use, recycling procedures need to be both economical and energy efficient. However, as the electrolyte makes up between 20 and 25 per cent of the weight of the cell, new methods must be developed to recover it due to the high recycling rates necessary, especially in the EU. Both polymers and conductive salts decompose at relatively low temperatures and may react with active materials at high temperatures. Pyrometallurgy as a method is therefore impractical and can lead to low recovery of materials and the possible formation of toxic compounds. For polymer SSBs, coarse shredding combined with metallurgical processing of the resultant foil fraction is a feasible approach. Using parts that can be divided into monomers and repolymerized is one way to recycle battery-specific polymers (Ahuis et al., 2024). This technique entails adding cleavable links or degradable monomers to the polymer, which are then split or broken down by heat or chemical processes (reversible radical polymerization). The link can be broken, and the monomers repurposed by heating the polymer to a temperature of up to 300 °C. In the end, creating recyclable polymers rather than recycling them is the holy grail of polymer recycling.

Lithium salt and polymer-based electrolytes can be reconditioned or resynthesized using various approaches. Firstly, pyrometallurgical and hydrometallurgical methods may be used to recover lithium from lithium salt in the form of Li2CO3, or a hydrometallurgical method can be used to recover Li3PO4. A solid polymer electrolyte's thermal breakdown can recover 70–80% of its LiTFSI (Ahuis et al., 2024). Furthermore, upon depolymerization, the polymer can be recovered by dissolving. However, it is important to take into account how the polymer solution affects (metal) leaching since inadequate leaching outcomes would arise from excessive viscosity and low liquid-solid mass ratio. Various resynthesis techniques, such as crosslinking or copolymerization, can be used, depending on the shape of the polymer ingredients that are recovered. Polymer electrolytes have a low value, which makes it difficult to create sophisticated, expensive recycling procedures. This makes industrial direct recycling of polymer electrolytes exceedingly challenging (Ahuis et al., 2024). The performance of the directly recycled electrolytes can also be significantly impacted by impurities in the recovered polymer and potential degradation processes of the polymers during operation. For a closed-loop material cycle, our general recommendation is to produce novel degradable SE polymers where new polymers may be synthesized from the resultant monomers.

The primary obstacle to recycling sulfur-based battery waste is the instability of sulfur-based electrolytes in ambient air or humidity. Initially, when they react with water, hazardous H2S is produced, which makes recycling safe. Second, the difficult synthesis of the sulphone or thiophosphate electrolyte in an inert environment account for a significant portion of the material cost since the reaction results in the electrolyte losing its electrolytic function and material value. Solvent-based stabilization and dry mechanical processing in an environment devoid of water are promising alternatives for secure recycling.

The benefit of solvent-based pretreatment/deactivation is that it dissolves the SE and lowers possible risks. On the other hand, pyrometallurgical processing is not advised due to the possibility of hazardous gas and SE breakdown, challenges in regaining lithium from the slag, and the consequently reduced material recycling efficiency. The electrolyte material is destroyed during pyrometallurgy, which further lowers the recycling value. Sulphone SSB recycling, in general, necessitates the highest technical and safety criteria, yet it is seen financially feasible due to the high-quality and costly components involved (Ahuis et al., 2024). Sulfide-based SSBs have the potential to yield comparatively high recycling rates with cost-effective process design by integrating solvent-based resynthesis with mechanical and hydrometallurgical processes.

It makes sense to resynthesize or directly recondition sulphide electrolytes in an appropriate solvent. Numerous synthesis strategies based on solvents have previously been disclosed. The recovery of electrolytes from the liquid phase is determined by the solubility of the materials and can be based on undissolved electrolyte particles (reconditioning/direct recycling) or complete dissolution of the reactants (resynthesis), depending on the electrolyte material and its condition after recycling. With strong (electro) chemical oxidation stability and ionic conductivities of >1 mS cm−1, halides are a relatively recent family of SE. However, because of their instability with metallic lithium, further recycling work will be required to create a hybrid cell with a sulphide separator, for example, or a protective layer on the anode side. After the cell has been mechanically broken down, the procedures must be carried out in a dry-room environment due to the instability of halides in moisture and the ion-exchange reactions that occur. Basically, there are two good choices. The first is solvent-based direct resynthesis or direct dry reconditioning in conjunction with lithiation, which is like direct recycling in that no elements are separated. Reconditioning via relithiation is the second method; it is followed by thermal treatment (co-melting) or high-energy ball milling. Because this process approach requires less energy due to its superior mechanical pretreatment and high levels of halide purity, it is advantageous from both an economic and environmental standpoint.

Because solid oxide electrolytes are hard and ceramic in nature, recycling them is difficult. To avoid ion exchange and the consequent loss of electrolyte function, an atmosphere with the least amount of moisture is required for processing oxide solid solid beads (SSBs). Oxide SSBs can be mechanically handled in a variety of ways after discharging in order to get the most effective procedure. To put it briefly, the goal of oxide-based system processing is to yield a fraction that resembles black material and can be treated using hydrometallurgical techniques or direct reconditioning procedures. Present-day solid-state battery (SSB) recycling methods include a variety of strategies to improve material recovery and economic viability. High-temperature thermal treatment has the ability to directly restore high ionic conductivity in materials such as composite cathode materials and LLZO oxide electrolytes. Another technique that shows promise is the removal of contaminants using solid-state reactions or hydrothermal processes, and then the relithion of materials with the addition of lithium salts in warm water (Ahuis et al., 2024). Because hybrid SSBs are composed of composite materials, they must be processed progressively, much like wet-chemical procedures, in order to separate the polymers from the inorganic electrolytes. Customized pretreatment procedures are needed for structural SSBs in order to extract reinforcing fibers directly. When combined, these techniques are recycling SSBs with high material yield and purity, improving both environmental and economic sustainability.

4. Suggestion

Solid-state batteries (SSBs) have a complicated composition and a wide range of components, which makes recycling difficult. This is especially true since different battery designs are required to coexist with different electrolyte types (polymers, sulphides, and oxides). In contrast to traditional LIBs, SSBs frequently have a larger percentage of metals, transition metals, and halogens than carbon. Because SSBs have a wide range of materials and require certain electrolyte conditions, recycling them poses both economic and environmental issues. A "one-size-fits-all" recycling strategy might result in reduced purity and recovery rates, especially for high-value elements like lithium, but it might also save pretreatment expenditures and initial investment costs. A more specialised strategy, such multi-stream recycling systems, could be able to manage various SSB kinds more skillfully. This strategy might use a number of processing techniques, including thermal and hydrometallurgical procedures, to separate and recondition electrolytes. To optimize material recovery rates and purity, process-specific treatment may be beneficial for SSB materials such as oxide and sulphide SEs, which are best suited for dry procedures. Legislative actions to increase collection rates and standardize battery forms are also essential for sustainability, as is the use of non-critical materials in SSB manufacture and energy-efficient manufacturing techniques (Azhari et al., 2020). These developments might facilitate the widespread implementation of SSBs while streamlining recycling and improving material recovery.

5. Conclusion

This paper focuses on the feasibility and challenges of recycling solid-state batteries (SSBs) through specialized processes tailored for various solid electrolytes, such as oxides, sulphides, and polymers. Key findings suggest that hydrometallurgical treatment, particularly dissolution methods, is highly effective for sulphide electrolytes, whereas oxide based SSBs may be better suited for dry processing techniques. Efficient material recovery requires the pre-separation of different SSB types, with multi-stream recycling systems offering flexibility and economic advantages by accommodating various battery compositions. The study also highlights the potential of direct recycling to enhance material yield while minimizing environmental impact. In order to achieve complete sustainability and closed-loop recycling, future research should focus on life cycle evaluations, improve the flexibility of recycling facilities to accommodate novel battery materials like lithium-sulfur and sodium-ion, and optimize multi-stream processing processes.

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