Environmentally friendly recycling and rejuvenation of used lithium-ion battery cathode materials

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Abstract. With more electric cars and the use of LIBs for green energy, the market has to accept a lot of batteries, which are now harming the environment and resources. Although LIBs rely on lithium, cobalt, and nickel to function well, their cathodes may become hazardous when not disposed of correctly. The study examines and evaluates technologies to restore LIB cathode performance for battery use. A range of techniques is discussed in the article by covering their role in improving usefulness, scaling capacity, and their effect on the environment. Producing new regenerative processes is better for the environment, uses resources more efficiently, and requires less energy than traditional pyro and hydrometallurgy. At this time, delivering direct regeneration to the market is difficult, as arranging the different materials and selecting the proper sorting and regeneration processes have proved challenging. Efforts should be made to connect or combine automated, set modules with different diagnostic systems for the large-scale creation of organ replacements. Applying this technology will support the circular economy and allow us to use less raw material in the future.

Keywords: Lithium-ion batteries (LIBs); Cathode material regeneration; Direct recycling technology; Hydrometallurgy and pyrometallurgy; Circular economy and resource recovery.

1. Introduction

As the basis of human survival and social development, energy and environmental issues have become hot spots of world concern. Tan et al. concluded that catalytic production of high-value chemicals and clean energy are regarded as the research frontiers.[1] The world consumes massive amounts of energy, which is expected to expand quickly throughout the coming years. Today, fossil fuels are still the world's primary energy source.[2] Renewable energy, wind power, solar energy, etc., generally develop (i.e., comply with the opposite historical development phase of cities). Still, its development is regionally conditioned in time and space and intermittently conditioned regionally. Thus, energy storage technology is needed to efficiently and effectively utilize renewable energy. The greenhouse effect has thus become the most pressing environmental issue confronting the international community. The development of transportation systems restructures an increase in energy storage grids, for which lithium-ion batteries are in high demand. Due to the special characteristics of lithium-ion batteries, such as high working voltage, small dimensions, low weight, long cycle time, wide operating temperature range, and low self-discharge rate, they are widely used in portable electronic devices. Since these batteries have become an ever-more critical need, the ability to manage the expected volume increase of those batteries has also been expanded, particularly as the first wave of EV batteries approaches their end-of-life cycle.

Between 1996 and 2020, the average annual growth rate of global lithium battery sales was always over 16%, and the average annual growth rate of lithium batteries was 24% from 2021 to the present, twice that of a national integration planner. However, the theory of the service life of a power lithium battery is about 8~10 years, which includes the recycling and disposal of waste lithium-ion batteries as a part of the whole construction planning. To ensure that when the recycling needs to reach a certain scale, there are the corresponding technical conditions and supporting infrastructure, to avoid wasting directly. As there are higher requirements for the battery-forming system upgrading and for different environmental and social benefits, the industrial model and technical requirements of battery recycling will also face new changes and tests. In general, a good-performance cathode material should possess: (1) high redox voltage; (2) safe; (3) stable discharge voltage; (4) low structure change in charge and discharge process; (5) high electronic and ionic conductivity; (6) rich source and easily prepared, as shown in Fig. 1[3] Gao et al. concluded that ion-conductive coating necessitated the cathode material exhibiting exceptional ionic and electronic conductivity.[4] Yamaguchi et al. confirmed that LiCaF-dominated cathodes result in the exceptional electrochemical performance of the oxide fuel cells.[5] Currently, the commercial LIB can be classified into four categories according to the material system of cathodes, namely, the Lithium Cobalt Oxide (LCO), the Lithium Manganese Oxide (LMO), the Lithium iron phosphate (LFP), and the ternary material — represented by nickel cobalt manganese (NCM) oxide. LCO has high working voltage, large volumetric energy density, and good electrochemical characteristics, which were mainly utilized within 3C industries, as illustrated in Fig.2[6]. However, this cobalt (Co) is limited to the global reserves, which increases the battery cost. LMOs have low energy density and short cycle life, and they are mainly applied to small power and new energy specialty vehicles, etc. The most used cathode materials in power batteries are LFP and ternary. Ternary batteries are used, which have a higher capacity but worse safety and stability than LFP batteries. Lots of research on sodium-ion batteries (SIBs) is primarily focused on the specific cathode materials, such as transition metal (TM) oxides, polyanionic compounds, Prussian blue (PB), and their derivatives (PBAs). Currently, the best cathode materials are layered oxides. Yet, their research has been given more attention because ternary or multiple oxide cells are structurally unstable and their efficiency is low. A substance that exemplifies ion-conductive compounds with NASICON-type structure is called Na, M2(PO4)3, and phosphate fluoride (Na3V2O2(PO4)2F3- 2) has a high working voltage. Unlike PBAs, the presence of crystalline water and vacancies in crystals results in limited battery capacity and poor performance, so batteries using crystals do not achieve much in energy storage.

Because so many batteries are now retired, recycling them as soon as possible is vital to allow the economy to use lithium, cobalt, nickel, and manganese. If these precious metals are handled badly, they are hazardous to people and the planet by hurting nearby places. Ironically, the momentum of environmental consciousness and the growing fear of turning resources into panaceas is inevitable in LIB. Disposing of these abandoned batteries has a significant environmental impact. More specifically, spent LiBs that are not recycled would create toxic byproducts that increase the danger of them by leaking soil and water pollution and heavy metals (cobalt and nickel).

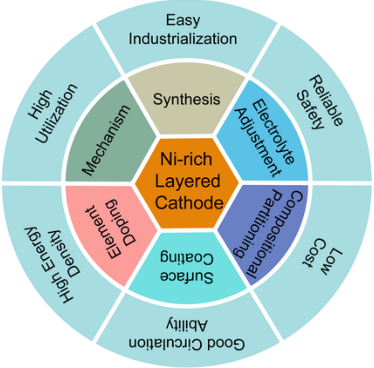

Fig.1 Ideal properties of Ni-dominant cathode materials.[3]

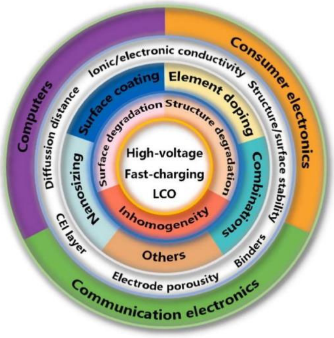

Fig.2 Structure, application, and advantage of Lithium Cobalt Oxide (LCO) in batteries.[6]

In terms of industrial application, because the positive electrode material is also the most valuable component of the entire battery, in light of economic interests, the recycling of precious metals represented by cobalt through dissolution-precipitation-recycling has formed a certain recycling business model. In response to a further drop in the cost of batteries, enterprises have widely begun to switch battery-positive electrode materials to chemical systems of high nickel, low cobalt varieties of ternary system, or lithium iron phosphate system, as illustrated in Fig.3[7] Because so many batteries are now retired, recycling them as soon as possible is vital to allow the economy to use lithium, cobalt, nickel, and manganese. If these precious metals are handled badly, they are hazardous to people and the planet by hurting nearby places. Different research teams at home and abroad have different targets for recycling, different reagents used in the recycling process, different methods, and even their conclusions. We sort out different technical methods to help people understand recycling and give a clear and organized technical route. Recycling waste lithium-ion batteries refers to separating useful components in the battery according to their physical and chemical properties, to realize the harmless treatment of pollutants and reuse of resources. According to one of the studies, a recurring treatment of waste lithium-ion batteries is categorized into three steps: battery pretreatment, separation of active substances and collectors, and recovery and reuse of valuable metals. Currently, pyrometallurgical and hydrometallurgical processes are most commonly used, as illustrated in Fig.4[8] Pyrometallurgical technology relies on the heat treatment of high temperatures to speed up the separation of battery components, and undergo chemical changes (oxidation, reduction, or combination) of metals or metal oxides in battery materials, and finally, through the subsequent treatment to recover waste materials in the form of alloys or metal compounds. The recycling process for waste lithium batteries has a wide range of applications. The high-temperature treatments provided in this novel approach can effectively remove the organic binders from carbon-based catalysts (in the form of char) and release them due to the thermal cracking of the organic binding material. Many commercial industrial recycling processes of waste lithium-ion batteries rely on pyrometallurgical technology. This is mainly because of the following two reasons: One is the high-temperature reaction and relevant chemical conversion rate are fast, accompanied by the short processing time as well as the strong material adaptability; Secondly, the recycling industry of waste lithium-ion batteries is still in the initial stage, many process flows are still exploring. Consequently, the industrial application becomes promising due to fully utilizing existing metallurgical technology and equipment. Li-ion battery recycling is a complex system of engineering tissue. The pyrometallurgical process is an essential step in recycling waste lithium-ion batteries. However, classification and treatment still rely upon traditional physical and chemical methods such as condensation, screening, magnetic separation, and leaching to enable the metal elements to be eventually reused. In a strict sense, hydrometallurgy means the recovery of other products with the chemical reactions of metals in aqueous solution. Hydrometallurgical technology for recycling waste lithium-ion batteries generally comprises a series of processes, including pretreatment of the waste lithium batteries, leaching, and separating valuable metals in the leachate. Thus, metallurgy operates on top of hydrometallurgy, whereby metabolic processes and metabolites of organic and inorganic acids are used by microorganisms (bacteria and fungi) to get the suitable elements oxidized and leached into solution as an ionic compound, which is then further treated to extract the useful element from the reagent. Due to its high efficiency, low cost, and low resource consumption, it is receiving more and more attention.

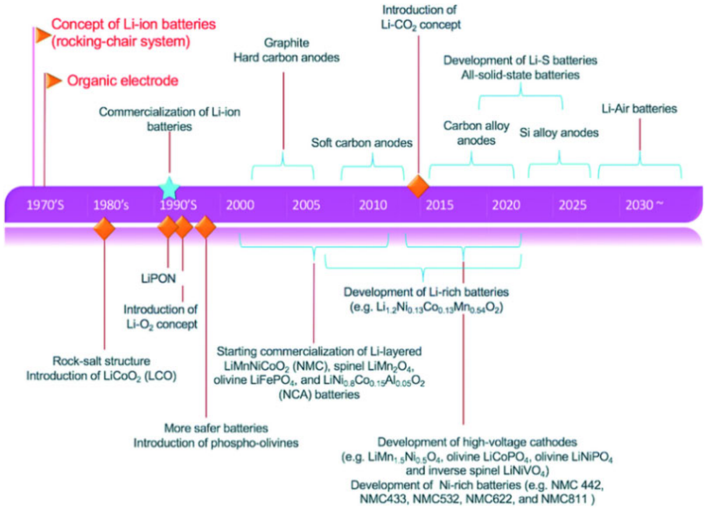

Fig.3 Development of cathode materials for LIBs.[7]

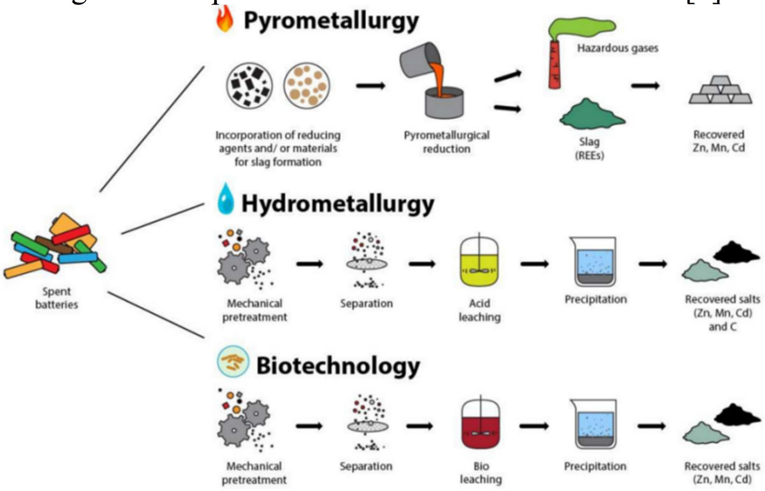

Fig.4 Conventional direct, pyrometallurgical, and hydrometallurgical recycling processes for the recovery of valuable components of Li-ion battery active materials.[8]

Because electric vehicles and renewables use more lithium-ion batteries (LIBs), a higher amount of these batteries is being thrown away without treatment, causing various problems (resource and environmental). [9] Vendors use essential lithium, cobalt, and nickel when producing new batteries found in the LFP and NCM materials. Still, these materials must be handled carefully because they harm the environment. Recycling technology is improving, but certain problems exist, such as having few people do it, difficulty recycling the metal, and negative effects from older methods. [10] Currently, the recycling and reuse field faces several challenges that new advanced technologies for cathode materials can handle. It looks into how advancements in cathode direct regeneration are achieved through four approaches aimed at rebuilding their original properties without disintegration. Using the described techniques, the major challenges of lithium loss, phase changes, and degradation on the material surface can be addressed. [11] Direct regeneration allows for greater efficiency in creating materials, costs less, and triggers less environmental damage. Restoring the cathode in this way leads to less waste in the production and makes the disposal of batteries safer. Using machine learning and solvents developed by engineers enables us to process large quantities of battery waste.

2. Conventional recovery methodologies

LFP batteries have critical components such as a positive electrode, a negative electrode (LFP, graphite, electrolyte, collector, diaphragm, etc.), auxiliary materials (binders for both electrodes, conductive agents, etc.), and pre-lithiation substances. Following the charged crush, the retired battery packs are converted into black powder and processed to separate the positive and negative electrode components.

2.1 Advanced Separation Techniques for Spent Electrode Materials

Old LIBs no longer work but still hold a lot of energy that could lead to short circuits, overheating, or fire.[12] Deactivation of the battery using salt solutions is required initially because it reduces the danger involved in handling and recycling processes. Before you start separating the tightly pressed parts of the battery, you must disassemble the battery by mechanical or hand methods. Due to the way electrode materials are attracted, it is challenging to separate them. The anode made from copper foil can be easily separated from graphite. However, detaching the aluminum cathode becomes difficult after graphene and its binder bond greatly, even after many charges. [13] Because batteries come in various sizes, types, and chemical combinations, their sorting and pre-processing should be designed for specific results. The most accurate approach is to document disassembly, but it takes significant time to set up for large companies. Rinsing and wet crushing materials offer fire and dust protection, though they lead to waste since the water carries away some particles. The best way to reduce environmental risks and secondary pollution is to balance the processes with effective systems for capturing dust and gas.[14] Fig.5[15] shows the standard process for recycling lithium-ion batteries. This diagram details how recycling is handled concerning spent lithium-ion batteries. Initially, used batteries are sorted, and they are only discharged mechanically. After leaching comes the part where lithium (Li), nickel (Ni), manganese (Mn), and cobalt (Co) are obtained from the material. Separating the material's coating, applying pyrolysis, and sieving leads to improved refinement.

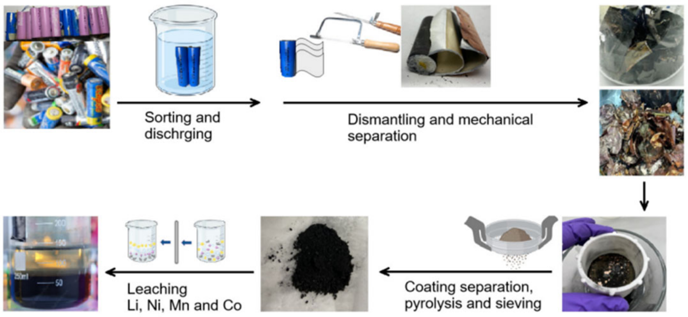

Fig.5 Pretreatment process and leaching of Li, Ni, Mn, and Co from spent Li-ion battery electrode coatings.[15]

Many methods have been invented to assist in the separation and recovery of electrode materials. Nowadays, mechanochemical activation is a more effective alternative to chemical leaching. It involves using forces to cause redox reactions and damage the electrode structure. The soluble lithium salt is recovered from spent lithium iron phosphate (LFP) when the material is co-milled with NaClO or $Na_2S_2O_8$ and then leached without using concentrated acids.[16] Better selectivity for metals is achieved when using fewer chemical solutions and applying various recovery methods. Carrying out large-scale processes with high-speed mills and expensive oxidizing agents becomes difficult and is very energy-intensive. Using the right pH, frother, and collector can result in a significant rise in the performance of the separation. Researchers have found that using pyrolysis pretreatment with LFP is beneficial for flotation as it removes PVDF and restores the material's surface with water affinity. Removing the $-(CH_2CF_2)$ n-segments in PVDF using heat treatment allows water to enter, making PVDF less waterproof and making LFP and graphite easier to separate in flotation. When combined, these methods allow for separating the cathode and graphite parts. High-gradient magnetic separation works well for LFP separation and reduces the usage of graphite. As a result of implementing magnetic separation and flotation, the graphite in the magnetic fraction was brought down from more than 12% to less than 5%. Even with better technologies, PVDF still appears in the processing system. When physical elements are removed, water - material interaction is not stopped because the binder films in the thin residue still impact each phase of flotation and leaching. The industry normally uses a calcining process to split decomposing PVDF, yet this process requires too much energy and makes some metals evaporate. Wang et al. improved the yield and purity of cathode materials by using a large-scale version of pyrolysis-assisted flotation technology.[17] PVDF melting is achieved with an

NMP solvent because it matches the natural structure of the active ingredients. When PVDF dissolution uses organic solvents, there are difficulties with toxicity issues, higher costs, and problems relating to scale. An alkaline NaOH treatment is a potential method for aluminum foil dissolution, since it preserves most items in the cathode. Various steps must be taken to recycle spent LIBs, including draining the batteries for safety, separating necessary parts controlled by a brain-inspired computer, crushing, screening, and using chemical or physical pretreatment to separate the materials. Higher levels can be achieved by combining mechanochemical activation, flotation, thermal process, and selective leaching. Further studies are required to boost these techniques for factories since they are now costly, use more energy, and create environmental issues.

2.2 Pyrometallurgy: High-Temperature Smelting to Recover Metals

Heavy metal volumes in the industry are recycled using the well-established and time-honored pyrometallurgical approach. Heating spent LIBs between 900 and 1200° C (lots of letters) separates the different metals into alloys and slag.[18] balt (Co), nickel (Ni), copper (Cu), and aluminum (Al) on a large scale from leftover cathode materials. Badminton shoes should be treated in a furnace at a certain temperature, but the process starts with breaking the shoes into components by shredding them first. Because melting points differ among metal groups, we can easily separate them. Because lithium-based battery materials are easily oxidized, the metallic ingredients pass into the molten layer as the lithium becomes slag. One outcome of waste management processes is scattered materials that can be reused due to the metals they contain. Processing waste batteries at a large scale is achieved with Pyrometallurgy using simple equipment. This approach allows for the efficient processing of a lot of materials, meeting the demands of the recycling sector. There are many significant disadvantages to using Pyrometallurgy. The main difficulty must be overcome because the process consumes excessive energy. When metals are melted this way, it takes a great deal of energy and releases lots of carbon.[19] The sulfur dioxide (SO2) and hydrogen fluoride (HF) emissions are very hazardous to the environment and are released during the whole process. Cathode material cannot heal its microstructure after it is destroyed by pyrometallurgical treatment. Because of this, restoring the original cathode function cannot be done using recovery techniques. It is important because it supports the processing of metals from large stockpiles at an economical price. Fig. 6[20] presents how high-temperature smelting is applied to spent lithium-ion batteries (SLIBs). A high-temperature smelter is designed to use and smelt all raw materials, namely cathode and anode, binders, electrolytes, and slag-forming agents that rely on charcoal. The first stage in slag and alloy separation is to clean the gas. The smelting of valuable metals leads to slag production and the required metals. After extracting the metal alloys, further handling of the remains allows them to be used again, which helps make lithium-ion battery recycling more efficient.

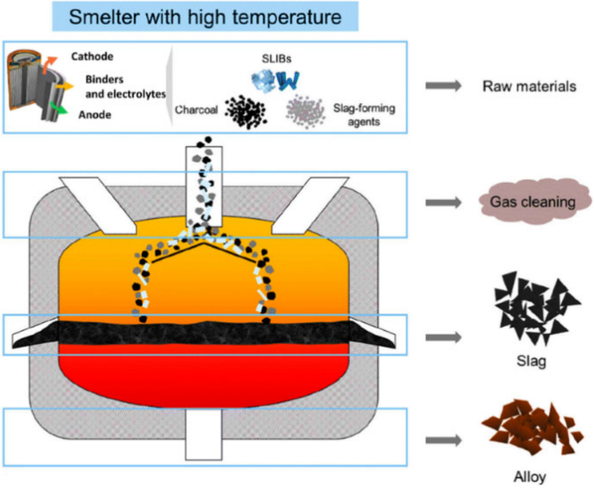

Fig.6 Typical pyrometallurgical processes of spent LIBs.[20]

2.3 Hydrometallurgy: Metal extraction using aqueous solutions

This method can potentially replace pyrometallurgy by extracting metals from used cathodes and treating them with chemical solutions of various pH levels.[11] Removing metals from the mixture begins with solvent leaching. The solution is made up of individual metals, and these metals are recovered by using solvent extraction, chemical precipitation, or the electrowinning technique. This field has an advantage because it is more energy-efficient than pyrometallurgical methods. The process requires no energy since chemical reactions are performed at normal temperatures in hydrometallurgy.[21] With hydrometallurgy, only lithium, cobalt, nickel, and manganese can be removed from the waste mixture left behind after spent batteries are used. Sulfuric acid retrieves cobalt and nickel, but hydrofluoric and nitric acids are needed to successfully recover lithium. When metals are removed from an organic solution by electrolysis, solvent extraction can be used to obtain the metals afterward. Hydrometallurgy brings several disadvantages to the process. The chemical compound used creates one of the most serious environmental hazards. Metallic compounds form large molecular structures with either sulfuric acid or sodium hydroxide bases or strong acids. Released wastewater or acidic liquid could result in polluted underground and surface water, eventually leading to secondary pollution.

The leaching of metals (lithium and Ni in particular) functions adequately through hydrometallurgy, but lithium recovery requires additional development since solution-based metal separation ranks among the most complicated operations. Lithium recovery through existing recycling protocols demonstrates high efficiency levels. However, suppose you seek to recover materials at small and medium capacities while keeping costs low. In that case, performing this task becomes challenging since hydrometallurgy is the main technology. Permanent improvements in chemical use alongside lithium extraction maximization continue at the formation stage. The electrochemical recycling method for spent $LiCoO_2$ (Lithium Cobalt Oxide) batteries appears in Fig.14[22] At first, the separated $LiCoO_2$ with graphite-based products are put through an electrochemical process in a hot NaCl and Na_2CO_3 saline solution. Connecting the anode and cathode with the outside power supply makes it possible for ions to move. When $LiCoO_2$ undergoes the process, it changes into lithium oxide (Li_2O), lithium carbonate (Li_2CO_3), and cobalt (Co). At the cathode end, oxygen gas is given out by the anode, while carbon dioxide forms at the other end. Since metals are restored by this method, there are fewer environmental consequences because it reduces the use of raw materials in LIB batteries.

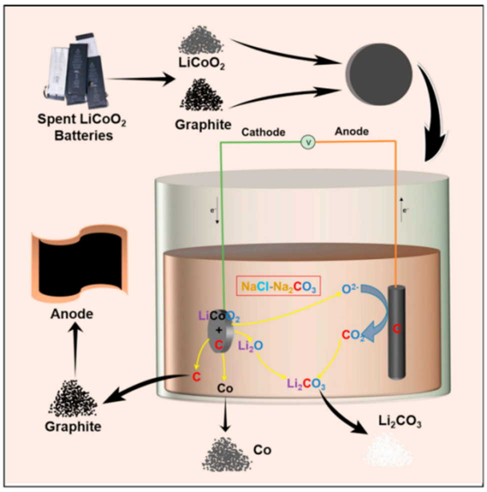

Fig. 7 Recycling of Spent $LiCoO_2$ Batteries through Electrochemical Process.[22]

2.4 Direct intensifying technologies

Strategies to intensify the recycling of spent lithium-ion batteries (LIBs) should contribute to an efficient, environmentally sustainable, and safe outcome. For this issue, techniques are grouped into external application methods, using new solvents, harvesting the product directly, and new tools in analytical chemistry. Magnetic, electric, and ultrasonic fields proved to be effective tools for boosting the speed of solids and liquids while also helping separate and remove the binder and active material from the positive electrode.[23] Acid leaching has discovered better alternatives using special solvents that are safe for the environment. When metal separation uses deep eutectic mixtures and organic ligands as novel solvents, it becomes more efficient and reduces environmental problems. Lithium salts and transition-metal oxides are more efficiently separated from each other using hydrophilic organic systems instead of strong liquid acids.[24] When direct methods such as lithiation, mechanical processing, and thermal reactivation are used, the cathodes can be restored to their original form and used again. Recycling reduces the activity's negative impacts on the environment and raw material resources. Two new technologies have been introduced to the LIB recycling field: biosorption and sorting assisted by machine learning. [25] Applying new technology in the industry leads the way to effective circular models in battery management, ensuring both the protection of nature and economic benefits.

3. Direct Regeneration of Cathode Materials

Now, more attention must be given to how the materials in LIBs are managed after these batteries are no longer used in electric vehicles or storage systems. LFPs can be transformed into various useful things, without changing how they are built, and they are beneficial for the environment. The change in shape is achieved by electrochemistry, while crystal structures stay unchanged. LFP loses function because lithium is immobile, iron converts into a higher oxidation state, and block and antisite defects appear on the olivine's surface. Now, more attention must be given to how the materials in LIBs are managed after these batteries are no longer used in electric vehicles or storage systems. The main benefits of lithium involve repairing the battery's structure and outer layers, removing defects, and handling lithium impurities. To collect lithium in lithium batteries, Li_2CO_3, LiOH, and different organolithium compounds are used in the solid-state technique. With temperatures between 700 °C and 900 °C, the system completes two processes: it allows lithium to be incorporated and, simultaneously, removes organic matter and changes Fe^{3+} into Fe^{2+}. When using –CN and –OLi groups, along with lithiation, the energy cost decreases and particle conductivity improves in the electrodes at easier temperatures.[26]

Both hydrothermal and solvothermal methods rely on performing chemical reactions with water under high pressure. LFP particles A can be reconstructed in uniform size with their original electrochemical qualities using a solution of lithium and iron. One advantage of using hydrothermal methods is that they protect the batteries from the damaging effects of high temperatures. Compared to other agents, ascorbic acid and oxalic acid effectively process the redox chemistry of Fe on the nanoscale by acting as reducing agents and chelators. When pressure is high and the reaction takes time, it becomes difficult for the industry to meet throughput goals. They assist the process by restoring the phase condition and removing PVDF, Al oxide particles, and carbonaceous dust from the surface. Using molten salts to decompose fluorinated polymers successfully ensures the safe disposal of HF gas. These molten salts, called ionic liquids, make it possible to recycle multi-element NCM cathodes for cathodes with high quality and strong capacities.[27]

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NCM cathodes for cathodes with high quality and strong capacities. There has been success with several regeneration techniques, but scientists still have to handle some problems that stand in their way. The capability of a material to be renewed largely hinges on its type of breakdown, which is affected by battery usage, the different conditions it experiences during use, and how the cells are built. In laboratory experiments, the reasons for failure are properly identified using neutron diffraction, Raman mapping, and synchrotron XRD. During the selective regeneration of mixed batteries, various cathode materials cause trouble because they could lead to chemical shifts and more phase impurities. It is vital to use LIB chemistry-based systems and intelligent methods for sorting before recycling begins. These methods are impossible in real situations because of the lack of automated, industrial-scale systems.

4. Conclusion and Future Perspectives

Because so many LIBs are in use, managing the trash from them and getting vital cathode elements have become the biggest challenges. Recycling LIBs on a large scale depends on traditionally used pyrometallic and hydrometallic processes, but those techniques create various environmental, energy, and material issues. With new direct regeneration, the cathode materials can restore themselves in place without being turned into a powder. These methods assist organizations in using resources wisely so that an economy following circularity principles can be created. Several problems make it difficult to adapt direct regeneration on an industrial scale. Different compositions in old batteries should be identified using particular classifications, and specialized methods for regenerating them will exist. Regenerative medicine is tested at the small scale of laboratory research and has not yet been developed with modern production methods. To use direct battery regeneration, it is necessary to set the same baseline for evaluating performance, greenhouse gases, and economic factors for all new technologies. The issue is still left unaddressed when it comes to reducing environmental harm, and there are effective ways to manage and dispose of harmful binders and any remaining organics, such as PVDF. With the help of advanced technology, we should create systems that can perform on-site XRD and spectroscopy to control regeneration processes. When policies for green recovery are implemented and technological research is repeated, it will help improve the recycling of lithium batteries at larger production factories.

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