In-depth research report

Explores the lithium recycling industry and provides ideas for process optimization

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China is currently the world's largest producer and consumer of lithium-ion batteries. Additionally, the installed capacity of power batteries is expected to reach 294.6GWh, which is a 90.7% increase from the previous year. According to GGII (Gao Gong Industrial Institute in Shenzhen), China's new energy vehicle sales volume is expected to reach 6.887 million in 2022, which is a 95.6% increase from the previous year. It is projected that China will produce 769,000 tons of scrapped lithium-ion batteries in 2025 and 4,198,000 tons in 2030 due to the 5-7 year effective lifespan of these batteries. This will result in the first large-scale scrapping of power batteries in 2026 and an increased demand for the lithium recycling industry in China. This paper focuses on optimizing the lithium recycling industry in China. Specifically, it explores ideas for improving the recycling process.

In the case of power batteries for new energy vehicles, once the battery capacity declines to 80% of the rated capacity, it is no longer suitable for use. This means that the battery has an actual effective life of approximately 5-7 years. The decommissioned power battery can be directly recycled or repurposed for low-performance requirement scenarios. The cascade utilization technique is used for power batteries that have decayed to less than 70-80% of their rated capacity. While these batteries may not meet the standards for use in electric vehicles, their remaining capacity can still meet the energy demands of other equipment. Batteries can be disassembled, screened, reorganized, and systematically integrated into small battery packs. These packs can be used in fields that do not require high energy density, such as low-speed electric vehicles (electric bicycles, courier trucks, etc.), solar street lamps, and communication base stations. Power batteries with more than 40% battery capacity degradation will undergo the disassembling and recycling process.

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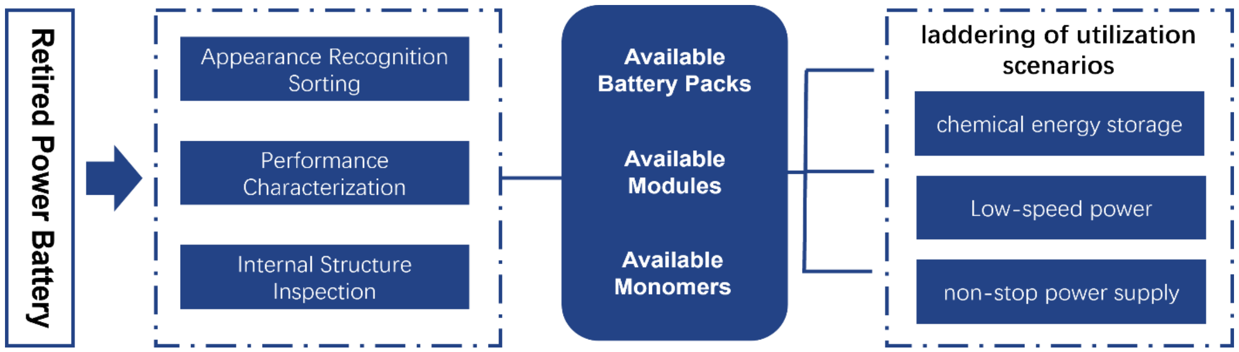

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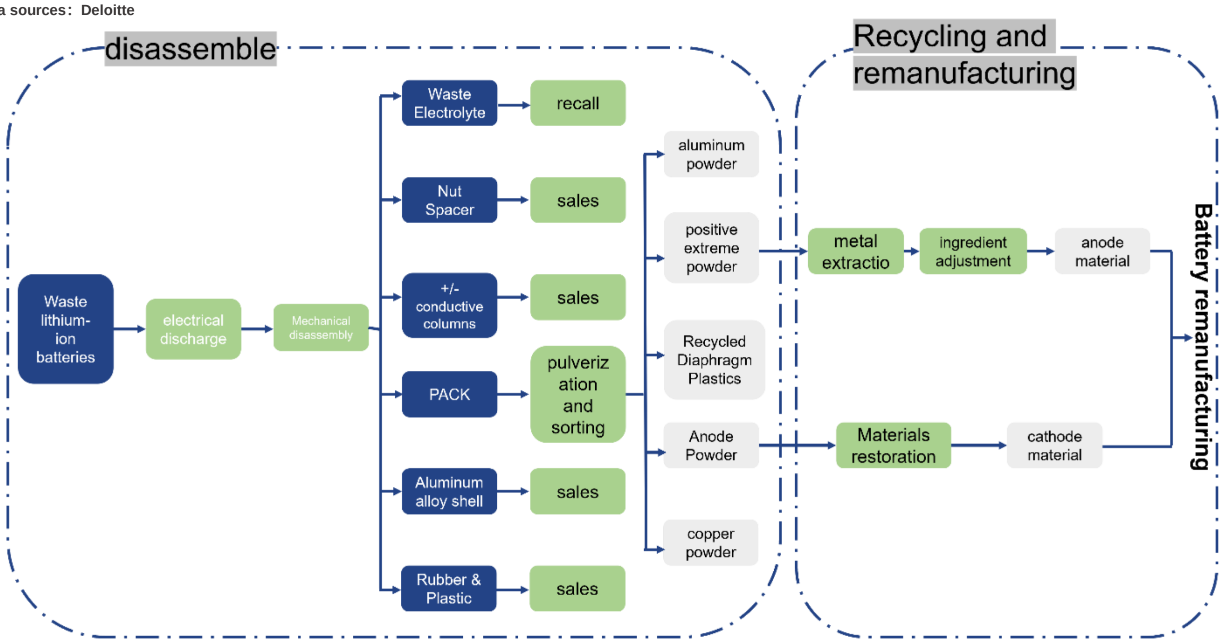

1

RECYCLING PROCESS ANALYSIS AND COST ESTIMATION

The recycling of used lithium power batteries is necessary for both environmental protection and economic reasons. From an environmental protection standpoint, lithium-ion batteries contain various heavy metals, organic and inorganic compounds, and other toxic and hazardous substances. If leaked into the soil, water, or atmosphere, they can cause serious pollution. Additionally, metals such as cobalt, nickel, copper, aluminum, and manganese have a cumulative effect and can be enriched in the human body through the food chain, causing harm. Therefore, it is necessary to centralize the harmless treatment of waste lithium-ion batteries and recover the metal materials therein to ensure sustainable development of human health and the environment. Additionally, according to Hao Siyue, Zhang Wei et al.'s 'Overview of recycling of used power batteries', recycling used power batteries can effectively reduce carbon emissions from raw materials by over 40%. From an economic perspective, the cathode materials in used lithium-ion batteries often contain valuable metals such as Li, Co, Ni, and Mn. In fact, the metal content in these materials is often higher than that of some natural ores. Extracting valuable metals from ores requires high costs and energy consumption. However, recycling these metals from used batteries not only yields high-purity products but also reduces costs effectively, generating considerable economic benefits.

The mainstream recycling process of lithium-ion battery is divided into four types: wet process, thermal process, combined process and repair / regeneration process.

The traditional recycling process is mainly wet and thermal recycling. Waste lithium-ion batteries are discharged, disassembled, crushed, sorted and other pre-processing steps to separate the positive electrode, negative electrode, and separator from the collector, and then crushed, sieved, and magnetically selected to obtain high-value failed positive electrode powder. The cathode material will be processed by thermal or wet method to regain the raw-material of cathode material, mixed with a certain amount of lithium salt, and sintered to generate new cathode material again. The two recycling processes extract the elements from the battery by completely destroying the original composition and structure of the materials in the battery, and use them as raw-material for the synthesis of new raw materials.

Emerging direct recovery technologies generally start from the composition and structure of the used material, make structural regeneration and restore the electrochemical activity of the material without destroying the inherent structure of the material. Mainstream technologies for direct recycle of cathode materials include solid-phase method, molten salt method, hydrothermal lithiation, atmospheric pressure lithiation with low eutectic solvent, etc.

Solid-phase method is simple and widely used, but with high energy consumption; molten salt method has low reaction temperature, but it requires strict requirements on lithium salt dosage and heat treatment time; hydrothermal method has lower lithiation temperature, shorter time, and more uniform reaction, but there are certain safety hazards in the high-pressure environment; low-eutectic solvent method can regenerate the failed cathode at atmospheric pressure and the DESs (Deep eutectic solvents) are green and can be recycled, which can greatly reduce the recycling cost, and is expected to be used for large-scale recycling, but the relevant research is still ongoing. It is expected to be used for large-scale recycling, but there are fewer related researches at present, and the DESs systems applicable to different cathode materials are still to be developed. At present, the direct regeneration process of waste batteries is still in the experimental stage of research and development, and has not been realized to be used on a mass scale for the time being.

Existing battery recycling in China is dominated by the traditional process of disassembly + hydrometallurgy. Recycling enterprises firstly disassembly and crush recycle batteries manually/mechanically into different materials, and enterprises with recycle as their main business will sell different materials such as shell plastics, aluminum powder, copper powder, anode powder, etc. to downstream affiliates, and the downstream company refine the materials; enterprises with a high degree of integration directly smelt the waste powders accordingly, which can be made into sulphates such as cobalt sulphate and nickel sulphate, and also precursors such as nickel hydroxide and cobalt hydroxide.

Referring to the oriented recycling process of recycling battery from BP: firstly, disassembly the waste power battery to get the metal shell, then heat to remove the organic solvents, the process is absorbed by cyclone dust removal and alkali spraying, and finally, after the mechanical crushing and sorting, sort out the materials such as the plastic shell, positive electrode, negative electrode and separator, etc. After acid leaching, the positive electrode material is used in the acid leaching process. After acid leaching of cathode materials with P204, P507 extraction to remove copper, iron and aluminum, the purified liquid is passed into the ammonia alkaline precipitation to generate nickel-cobalt-manganese hydroxide, and then add the recycled lithium carbonate, sintering preparation of ternary materials.

Opposite to China, there are some countries where the recycling process is based on thermal-metallurgy, where the pre-treated active material is placed in an incinerator at a high temperature to remove organics, smelted to obtain metal alloy, and then obtained metal compounds through the leaching/extraction process.

Specific operation, used battery recycling treatment is mainly divided into three processes: pretreatment, first process and main process. Pre-treatment is mainly for deep discharge, crushing and physical sorting. First process is to make positive and negative electrode active materials and substrate separation occurs, mainly heat treatment method, organic solvent dissolution method, alkali dissolution method and so on. Main process includes leaching and separation and purification, and the extraction of valuable metal materials is the key to the recovery process.

2

PRETREATMENT AND FIRST PROCESS: DISCHARGE, DISASSEMBLY AND MATERIAL SEPARATION

2.1 Discharge

In order to prevent short-circuiting and spontaneous combustion of used batteries, it is necessary to discharge the batteries first before disassembly them. The mainstream practice is to immerse the positive and negative terminals of the battery into a conductive salt solution to discharge by short-circuit; this method is highly efficient and stable, low-cost, and suitable for small-scale discharge treatment of used batteries. For lithium-ion battery packs for electric vehicles, due to the high residual capacity, a charger/discharger can also be used to collect the residual power and detect the residual voltage in the safe range before entering the disassembling and crushing stage.

For high-capacity battery mass industrialization application, it can also use low-temperature freezing method, that is, the waste battery frozen to a very low temperature (such as liquid nitrogen freezing) inactivation and safe crushing; but the method of equipment requirements is higher, the initial construction cost is higher, the current use of the method of the United States Umicore and Toxco companies.

2.2 Sorting and disassembling

Due to different active materials, different purposes of use, different battery manufacturers produce batteries in the volume, packaging, material components, etc., sorting and disassembling is aimed at removing the shells and packaging of used batteries (groups), reducing the battery volume, and targeting the classification of different types of waste batteries.

In laboratory research and enterprises that have not yet recycled on a large scale, manual operation is the main disassembling method. Operators use knives, saws and other tools to manually disassembly the used cell, remove the plastic or metal shell, and separate the shell, positive and negative electrodes, and separator. The purity of the active material obtained in this way is relatively high, and the battery cell enters the subsequent processing stage as a whole with less impurity content. However, the manual disassembly efficiency is low, the processing capacity is small, and it can only be used as a research stage or small workshop production, and it is difficult to reach large-scale industrial application.

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In the face of large-scale recycling process, mechanical disassembly is more advantageous in terms of economy and industrial application. Mechanical operation of the plastic or metal container open the battery with hacksaw cutting to remove the ends and containers, access to the internal material of the battery, and then process the chemical composition of the positive electrode material using a more targeted recycling process for batch processing. In terms of battery sorting, Philips designed sensors can detect the magnetic field of each battery and measure the corresponding response frequency, with an accuracy of 99%, but the batteries must pass through the sensor inspection one by one, which makes the sorting speed slower. In Germany, according to the shape and size of the battery after a simple manual picking, the use of X-ray inspection method to further subdivide the different components of the battery, placed on the conveyor belt after X-ray scanning, real-time analysis of the battery type, mechanical sorting to different containers, sorting speed of up to 12 blocks / sec.

2.3 Crushing. sieving & separation of active materials from collectors

After sorting and disassembling, metals contained in the materials need to be further sorted out, by dry or wet processes. Dry recycling process is not through the solution and other media, direct recovery of valuable metals, the main use of heat treatment + mechanical separation of multiple processing. Mechanical separation method is to take advantage of the differences in density, magnetism and other physical properties of different components of the battery, using flotation, vibration screening, magnetic separation and other ways to screen the classification of crushed battery materials, to achieve the initial separation of plastics, metal shells, copper foil, aluminum foil and electrode materials. However, the battery active material is adhered to the collector through the binder, and the separation speed of mechanical treatment directly separating the active material is low: if the crushing intensity is small, many active materials can not be completely recovered, and the intensity is too large, and some copper and aluminum will be crushed to fine particles into the active material. Therefore, the removal of organic binders is a necessary step before mechanical crushing, firstly, through the heat treatment method of discharge, disassembled electrode waste calcined at high temperature to remove the separator, binder and carbon materials, etc., and then through a series of mechanical processing (such as crushing, screening, etc.) to achieve the separation of the active material and the collector fluid. Dry recycling process is simple, rapid reaction at high temperature conditions, suitable for dealing with a large number of batteries or more complex structure; but high energy consumption and easy to cause atmospheric pollution, the initial investment in equipment is also higher.

Wet recycling process is to dissolve the metal ions in the waste lithium batteries through acid and alkali solution, and then use precipitation, adsorption, extraction and other methods to re-extract the ions in the solution, so that they are separated in the form of oxides, salts and so on. In the active material and collector separation section, the main wet process has organic solvent dissolution method, alkali dissolution method and so on. Organic solvent immersion method uses the principle of similar solubility, the use of organic solvents with strong polarity to dissolve the binder, destroying the adhesion of the interface between the collector and the contact of the active material to result the separation of the active material; the method does not destroy the structure of the material and does not change the components of the active material, the recovery efficiency is high; however, most of the organic solvents are expensive and toxic and volatile, which is not suitable for large-scale industrial applications.

Alkali dissolution method is to use the amphoteric nature of aluminum, the use of alkali dissolved aluminum foil, and the active material is not dissolved in the solution, to achieve the separation of the two; the method is simple to operate, separation efficiency is high, but the generation of sodium aluminate is more difficult to recycle and handle, strong alkali is also easy to corrode the equipment. Wet recycling process is more complex and delicate, but the purity of the recycled products is higher, so it is the first choice of the current waste battery recycling process.

3

DEEP TREATMENT: LEACHING ,SEPARATION AND EXTRACTION OF TARGET METALS

The deep treatment of battery recycling is to dissolve and leach the electrode materials after

pretreatment discharge, dismantling, crushing and separation, so that the
The metals and compounds therein enter into the leaching solvent in the form of ions, and then the separation and recovery of the corresponding metals are carried out respectively;
It can be mainly divided into two stages: leaching and extraction.

3.1 Leaching

Leaching is a key step in the wet process of used battery recycling, which is mainly to convert the metal elements in the active material of the positive electrode after pretreatment into ions in the solution, so as to facilitate the subsequent separation and recycling process, and the commonly used acids include inorganic acid (HCl, H SO, HNO, H PO, etc.) and organic acid (oxalic acid, citric acid, malic acid, etc.). Among the traditional inorganic acids, hydrochloric acid has the best leaching effect, but it is easy to be volatile, and Cl_2 will be generated during the reaction process; nitric acid is not only volatile, but also has strong oxidizing property, which is easy to generate toxic nitrogen oxides, and the price is higher than that of hydrochloric acid and sulfuric acid; sulfuric acid is inexpensive and easy to obtain, with higher boiling point, and it can be used to improve the leaching rate and solubility by adopting a higher leaching temperature. However, the leaching efficiency of sulfuric acid is relatively low, so the actual operation process is often added in the sulfuric acid solution reducing agent H_2O_2, and the need for higher leaching temperature and a larger liquid-solid ratio. However, due to the poor stability and easy decomposition of H_2O_2, a lot of research agent has been devoted to finding more efficient and stable substitutes.

In addition to inorganic acids, environmentally friendly organic acids are also the current direction of attention: organic acids do not produce toxic gases compared to inorganic acids, and the waste liquid is not as acidic as inorganic acids.

gas, the waste liquid is not strongly acidic, easy to deal with, less corrosive to the equipment; but the price of organic acid is higher, the leaching speed is slower, and the liquid-solid ratio is higher than the liquid-solid ratio.

However, the price of organic acid is higher, the leaching speed is slower, the liquid-solid ratio is higher than that of inorganic acid, and the processing capacity of corresponding positive electrode materials is much smaller than that of inorganic acid, so it is difficult to be applied to large-scale processing.

Therefore, it is difficult to apply to large-scale treatment. If it is necessary to recover metal Al first, two-step method can be used to dissolve and leach metal materials with alkaline solution first, and then leach other metals by acid leaching.

Acid leaching of other metals. Overall, the core of the acid leaching process is the reaction temperature, time, acid concentration, solid-liquid ratio and reductant content, which directly affect the metal ionization process.

content of reducing agent, which directly affects the leaching rate of metal ions.

3.2 Extraction

After the leaching of anode active material, the key recovery of cobalt, lithium, nickel, manganese and other metals are in the form of ions in the leaching solution, which need to be further processed in depth for thorough separation, purification and recovery, and the mainstream processes include chemical precipitation, solvent extraction, electrochemical deposition, etc. Chemical precipitation is the process of leaching metals into the leaching solution.

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Chemical precipitation method is to add appropriate precipitant to the metal leaching solution, and metal ions precipitation reaction to achieve the separation effect. The separation mechanism of chemical precipitation method is the different solubility of metal compounds under a certain pH, and different metal ions such as Ni, Co, Mn, Li, etc. are separated by gradient precipitation under different pH values; this method has a high extraction rate, low cost, and low requirements for equipment, but due to the harsh conditions of precipitation of some metals, a variety of metals may be precipitated at the same time if the dosage of precipitant and the acidity and alkalinity of solution are not well controlled phenomenon, difficult to separate, resulting in unnecessary waste of resources.

The extraction method is to choose a specific extractant or a mixture of several extractants, through the formation of stable complexes with the target metal ions, separate from the leaching solution in the organic phase, and then back-extract the complexes to achieve the separation and purification of metal ions. The advantage of extraction method is that the target metal ions are extracted accurately, low energy consumption, easy to operate, and the recovery rate and purity are relatively high; however, the disadvantage is that a large number of chemical reagents are required, and there is the risk of polluting the environment, and the solvents will be lost to some extent during the extraction process, and the cost is high. Usually, mixed extractant has better synergistic effect, and the extraction effect is better than single extractant.

At present, China battery recycling plants mainly use step-by-step extraction method to recover metal ions in the leach solution: first of all, Cu, Al, Fe and other impurities are removed by chemical precipitation, the purification solution is extracted manganese with P204, and the loaded organic phase is back-extracted with sulfuric acid solution and purified to get pure manganese sulfate solution; the residual liquid is then extracted with P507 under different pH conditions to extract Co and Ni, and the loaded organic phase is back-extracted with sulfuric acid solution to get pure manganese sulfate solution; and the residual liquid is then extracted with P507 in different pH conditions to extract Co and Ni, respectively. Co and Ni were extracted by P507 under different pH conditions, and the loaded organic phase was back-extracted by sulfuric acid solution to pure cobalt sulfate and manganese sulfate solution. This method can prepare pure Mn, Co, Ni sulfate products, but the extractants used in the extraction process, P204 and P507, need to be saponified, and the alkali consumption is large; in addition, the whole extraction process is relatively long, and the number of stages is large.

Electrochemical deposition method refers to the method of obtaining metals by electrochemically reducing the target metal ions in the leaching solution at the cathode through the difference in electrode potential under the action of an external electric field. The method is simple and easy to implement, no chemical reagents is required in the operation, the introduction of impurities is small, not only to make the product purity and recovery is very high, but also to avoid the complexity of the subsequent treatment process. The method is simple and easy to implement, no chemical reagents required in the operation, the introduction of impurities is small, not only to make the product purity and recovery rate is very high, but also to avoid the complexity of the subsequent treatment process. However, the disadvantage is required to consume more electricity, in addition to avoid the co-deposition of other metal ions, to purify the active material in the pre-treatment process.

Despite the relative simplicity of the dry process, lower recovery costs, but the product of more impurities, more pollution in the treatment process, the target product recovery rate is lower than the wet method, there are certain process deficiencies; therefore, the current battery recycling production line in China to the wet method is dominated.

4

BATTERY RECYCLING PRODUCT VALUE ANALYSIS

For lithium iron phosphate battery, the current main recycling products are waste copper, lithium carbonate, iron phosphate. Referring to the calculation in Chapter 2 of this paper, in the lithium iron phosphate power battery pack, the cells weight is about 60%, and the weight of anode materials in the cell is about 32.1% (the active material in the anode material accounts for 88-89% of the weight), and the weight of copper foil is about 10.8%; in accordance with the recycling rate of 98% of the copper foil, 90% of the lithium carbonate, 95% of the iron phosphate assumptions, a single ton of lithium iron phosphate battery packs can be recycled to collected the scrap of copper 63.5kg, lithium carbonate 35.9kg, iron phosphate 154.8kg, corresponding to the value of the main recycled products 17,000 yuan / ton of lithium iron phosphate battery pack.

For ternary battery, the main recycling products are nickel sulfate, cobalt sulfate, manganese sulfate and lithium carbonate. In the ternary power battery pack, the weight of cells is about 68.2%, the weight of anode materials in the cell is about 39% (the weight of active materials in anode materials accounts for 88-89%), the weight of copper foil is about 13%; according to the assumption that the recovery rate of copper foil is 8%, nickel sulphate 98%, cobalt sulphate 98%, manganese sulphate 98%, and lithium carbonate 85%, a single ton of different ternary battery packs can be extracted to collect 86.9 kg of waste copper, 123.4 kg of nickel sulphate, 123.4 kg of nickel sulphate, 123.4 kg of manganese sulphate, and 85% of lithium carbonate, kg, nickel sulfate 123.4~293.6kg, cobalt sulfate 36.8~123.6kg, manganese sulfate 5.8~120.4kg, lithium carbonate 84.9~85.7kg, which corresponds to the value of the main recycled products of 42-45,000 yuan/ton of ternary battery pack.

5

SOME REFLECTIONS ON THE FUTURE

In summary, for the battery recycling industry chain participants, the future should be more focused on the stable collection of raw materials, recycling links to simplify the cost, as well as product yield enhancement.

Waste battery source channel standardization, the extension of producer responsibility

From the perspective of raw material collection, the lack of regulation and standards in the early stage of the industry has caused disorder in the front-end waste battery recycling system, with a large number of informal enterprises competing for high prices and squeezing the space of formal enterprises. For waste battery resource channel operators, choosing formal channels for recycling means paying higher costs, for example, formal enterprises need invoices for recycling batteries in order to offset VAT in later sales, causing additional costs for small-scale recyclers.

This causes additional costs for small-scale recyclers, so raw material suppliers prefer small workshops and used car markets.

After the formal implementation of the pilot of the extension of producer responsibility in China in 2023, automobile enterprises assume the main responsibility to complete the battery recycling with a third party. For example, in the first batch of extended producer responsibility pilots, Dongfeng Corporation, as the main unit, jointly with Lantu Automobile and Dongfeng Hongtai, as the joint reporting unit; Chery Automobile, as the main unit, jointly with United Xuan Hi-Tech, Jinfa Science and Technology and eight other enterprises as the joint reporting unit. Geely Automobile/Geely Commercial Vehicle, as the main unit, has joined hands with Huayou Resources, Saidmei and other enterprises as the joint reporting unit. At present, Geely Automobile has opened up various links of recycling, dismantling, manufacturing and sales, and plans to set up a recycling system to recycle 20,000 end-of-life vehicles annually within two years in six cities and regions, such as Hangzhou, Ningbo, Taizhou, Jinhua, Ningguo and so on. By setting the pilot target of extended producer responsibility for automobile products in 2023, the company plans to achieve the target of 75% comprehensive utilization rate of automobile resources for key models by 2024.

Pre-processing: Intelligent dismantling and cost reduction is a major issue to be tackled by the industry

In the front-end dismantling part, manual labor is still needed. In China alone, there are more than 5,000 kinds of power battery packs, with many varieties, different brands and models, complex structures and uncertain decommissioning status, so the shells of battery packs and the outer packaging of single batteries are still mainly dismantled manually. When dismantling large quantities of batteries, there are many problems with manual dismantling: high voltage battery packs, complex internal wiring harnesses, there is the risk of electrocution, short circuit; battery packs with a large amount of internal adhesive, the need to rely on brute force to dismantle; at the same time, we also need to pay attention to improve the dismantling efficiency and reduce labor costs. Therefore, intelligent dismantling is a major issue that the industry needs to focus on. For the intelligent and flexible dismantling of used power batteries by machine instead of manual labor, the main steps include: the establishment of data acquisition system from 3D camera, collaborative dismantling of cover screws by multiple robots, handling of cover, handling of battery modules, intelligent sorting of dismantled products, and milling of modules & batteries, and other steps. The main breakthroughs involved in intelligent dismantling stem from the diverse appearances of different battery models, the intelligent recognition and grasping of components, and the deformation generated after years of operation, which requires the dismantling system to be dynamically adjusted according to the specific conditions.

Hydrometallurgy: yield improvement & process simplification become the focus of attention

The limitation of lithium recovery rate mainly comes from the nickel-cobalt-manganese solution extraction, the waste residue in the process of removing impurities will adsorb the loss of 10% of the lithium ion. In the ternary battery black powder acid leaching solution, lithium is the smallest radius, the most active metal; although in the front-end calcination reduction process, can be extracted 80% of the lithium carbonate, but in the second section of the nickel cobalt manganese extraction process, the formation of the dregs will be adsorbed 10% lithium ions, resulting in the third section of the lithium carbonate crystallization of the reduction of the recovery rate, so it is difficult to exceed the recovery rate of lithium for the ternary batteries of the existing process 90%. In order to improve the recovery rate of lithium, the domestic high efficiency led by the Central South University began to research and development of new lithium extractant, aimed at the front end of the extraction instead of calcination reduction, the full extraction of lithium ions, to avoid the impact of the back end of the dregs adsorption.

In addition to the front-end lithium extraction process optimization to improve yields, the end of lithium recovery also has the space to reduce costs. At present, the end of the nickel and cobalt extraction of lithium recovery, the main use of MVR evaporation process concentration, through the evaporation of water in the solution so that the concentration of lithium from the original 1g / L to more than 10 g / L, and then complete the back end of the precipitation of lithium. MVR evaporation process has the advantage of mature technology, widely used, but its operation requires the consumption of large amounts of electricity (production of 1 ton of lithium carbonate MVR equipment needs to consume 9000 degrees of electricity), the cost is relatively high. The MVR evaporation process has the advantage of mature technology and wide application. On this basis, some companies in the industry also try to use adsorption + membrane, extraction and other more economical solutions to realize the concentration of lithium, reducing the use of MVR equipment. Adsorption + membrane is mainly through the process of adsorption and desorption to increase the concentration, while extraction is realized through extraction and reverse extraction. Compared with the MVR process, the consumption of electricity during the operation of these two processes is greatly reduced, and only part of the consumption of reagents such as adsorbents, membranes, extractants, etc.; the upfront investment is also smaller than that of the MVR process, which is conducive to the enterprise's cost reduction.

The original intention of the development of clean energy is to realize the sustainable development of low carbon and environmental protection, with the first batch of power battery end-of-life, end-of-life battery recycling is a challenge that we must face, we will pay close attention to the latest information and technological development of the lithium recycling industry, if you're also interested in this welcome to subscribe to us, we will share the industry from time to time cutting-edge.

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