Recycling of Rechargeable Batteries: Insights from a Bibliometrics-Based Analysis of Emerging Publishing and Research Trends

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rechargeable batteries contain heavy metal elements, such as nickel (Ni), cobalt (Co), and lead (Pb), which are carcinogenic and mutagenic substances. Improper disposal will cause harm to human health and the environment. In addition, organic and strong acid/alkaline electrolytes of rechargeable batteries can also cause pollution. Green, efficient, and low-cost recycling of these wastes can only avoid potential threats to the environment and human health, but also provide raw materials for the production of rechargeable batteries, reduce dependence on primary mineral resources, and promote sustainable development of the batteries industry. Growing environmental sustainability concerns have been continuously encouraging governments and global organizations to implement efficient and environmentally benign recycling technologies.

Countries continue to lay out in the field of rechargeable battery recycling for more than 20 years, resulting in a surge in the number of patent applications and literature publications. At present, most attention is given to the recycling of LIBs. However, lower recycling efficiency and serious secondary pollution hindered the large-scale application of most of the recycling technologies. Researchers are highly unified on this but are baffled. Therefore, a quantitative analysis of the recycling field to understand the pain points and difficulties will help to improve recycling technology and promote the development of the industry. Bibliometrics is a field of study that attempts to use bibliographic data of publications and their citation relations to evaluate and reveal the structure of research. It has been applied in many fields, such as CO2 conversion, the global situation of COVID-19 research, Nanomedicine. Current literature reviews on the recycling of rechargeable batteries mostly focus on evaluating different recycling technologies and making qualitative comparisons. To the best of our knowledge, however, there is a dearth of information concerning research patterns and publishing in the field of rechargeable batteries recycling. Therefore, the purpose of this study is to conduct a comprehensive analysis of relevant patents and scientific literature since the 21st century based on bibliometrics, quantitatively and qualitatively. From the perspective of industrial application, the research status and future of recycling technology are systematically sorted out to provide a quantitative reference for the development of the technology and efficient policy guidance.

2. Results and Analysis

2.1. Analysis on the Development Trend of Patents

As an important part of intellectual property with high technological content, patents have an important influence on the implementation of intellectual property strategies and the research and development (R&D) of enterprises. Effective use of patent information can shorten R&D time by 60% and save R&D costs by 40% on average. In the world’s average R&D output, the economic value of patents exceeds that of other activities by about 90%. In this section, we analyzed and compared rechargeable battery recycling-related patents worldwide to reveal the characteristics of the patent application layout in this technical field.

2.1.1. Regional Distribution and Trends of Patent Applications

The number of patent applications, to some extent, can reflect the strategic deployment, technological level, and market conditions of different countries in this field. The geographical distribution of global rechargeable batteries recycling patent applications is shown in Figure 1. The data showed that the technology concentration in this field was relatively high, mainly in China, South Korea, Japan, and the USA. China ranks first in the world with 2723 patent applications, while South Korea, Japan, and the USA have 127, 70, and 16 respectively, ranking second, third, and fourth, respectively. Countries and regions such as Russia, India, and France filed relatively few patent applications.

Figure 2 shows the trend of global rechargeable batteries recycling patent applications over the past two decades. This field has experienced three important stages: the initial stage, the slow growth stage, and the development stage. From 1999 to 2006, the battery recycling technology was still in its infancy with a small number of patents. With the rapid development of science and technology and the in-depth understanding of recycling technology, the number of batteries recycling patents increased slightly since 2007. The number of patent applications in China rose sharply by 2013, accounting for more than 69% of the total global patent applications. Since 2014, the number of annual applications has increased exponentially, indicating that the recycling technology has produced breakthroughs and has entered a development period. During this period, China’s patent applications account for more than 88% of the global total, reflecting China’s great enthusiasm for research and development in the field of power batteries recycling and reuse, while maintaining a high degree of research activity. This phenomenon was not only related to China’s huge rechargeable batteries consumer market but also closely related to increasing emphasis on environmental protection, resource recycling, and sustainable development. Figure 2 shows a “zero-high-surge” trend, which indicates that the batteries recycling industry has become a new sunrise industry that is currently widely concerned in the world. More R&D personnel from enterprises and research institutions have sprung up in related research and development, indicating that batteries’ recycling and comprehensive resource utilization have greater space for exploration and development potential.

The trend of patent applications was closely related to the development of the industry. Before 2000, as a major electronic product manufacturing country, the global power batteries recycling industry was basically controlled by Japan and South Korea (Figure 3). Since 2001, China’s gradual emphasis on the batteries recycling industry has broken this situation, forming a trio of China, Japan, and South Korea. The number of patent applications in China has been far ahead of other countries and regions, showing an exponential increase due to the increasing demand for key resources and the high emphasis on environmental protection. In addition, there are huge market opportunities for batteries recycling and utilization, which encourage scientific research institutions and battery recycling companies to actively carry out production, education, R&D in this field, and further stimulate the output of rechargeable battery recycling and utilization technology.

Through classification statistics and system analysis of rechargeable batteries recycling technologies, the development
trend of recycling patents for different rechargeable batteries systems is shown in Figure 4. As many as 1548 patents were filed for LIBs recycling technology, accounting for 48% of the total. The patents related to the recycling of LIBs cathode, anode, and electrolyte (such as N-methyl pyrrolidone (NMP)) materials accounted for 29.6%, 11.5%, and 4.1%, respectively. In addition, there are a small number of patents related to the treatment of separators for LIBs, because the recycling of cathode materials for LIBs has high economic returns, and related scientific research institutions and R&D companies invest more in it, leading to a large number of patent applications. Similarly, the recycling and resource utilization of LIBs have experienced three consecutive periods: the initial period, the slow growth period, and the development period.

Hybrid batteries (HBs, without specifying the batteries systems) had the second-highest number of patent applications, mainly for disassembling and shattering waste batteries. There were 391 patents on lead-acid batteries recycling technology, ranking third. Due to the early start of development and considerable progress in theoretical research, product types, product performance, etc., lead-acid batteries have played an indispensable role in the fields of transportation, communications, electricity, military, navigation, and aviation. The main contaminants involved in lead-acid batteries were heavy metal lead and

Figure 1. Regional distribution analysis of authorized patent applications.

Figure 2. Patent application trends for rechargeable batteries recovery.

Figure 3. Patent application trend of rechargeable batteries recycling in South Korea and Japan.

Figure 4. Development trends of different rechargeable batteries recycling patents.
electrolyte sulfuric acid solution pollution. Lead metal can cause neurasthenia of the nervous system, numbness of hands and feet, indigestion of the digestive system, blood poisoning of the blood circulation system, and kidney damage of the urinary system. Therefore, research on the recycling of lead-acid batteries was carried out earlier. From the application cycle analysis, the number of patents from 1999 to 2011 was relatively small, accounting for about 30% of the total annual patent applications for rechargeable batteries recycling. The resource utilization of waste batteries was maintained with great enthusiasm. Since 2012, the annual total number of applications has remained above 30. However, due to the strong development of LIBs recycling technology in recent years, the proportion of lead-acid batteries recycling patents is far below 30%.

2.1.2. Important Patent Applicants and Layout Situation

The analysis of patent applicants and application trends can indirectly reflect the research attention and technical level of different applicants or main R&D personnel and reflect the distribution of scientific research institutions, scientific research levels, and technical research differences. Figure 5 shows the classification and statistical analysis of the distribution of main applicants.

Companies with a large number of patent applications in China include Hefei Gotion High-tech Power Energy Co., Ltd (GOTION), Ganzhou Highpower Technology Co., Ltd (GANPOWER), Guizhou Zhongwei Resources Recycling industry development Co., Ltd (CNGR), etc. Central South University (CSU), Sichuan Normal University (SICNU), Beijing Institute of Technology (BIT), Kunming University of Science and Technology (KUST), and University of Chinese Academy of Sciences (UCAS) have a large number of patent applications. They all invested early in the research and development of battery recycling technology or recycling devices and have always maintained innovation and attention in the field of technology. Among the foreign patentees, Sumitomo Metal Mining Co. (Japan) and Korea Institute of Geoscience and Mineral Resources (KIGAM) had more patent applications, 47 and 32, respectively, accounting for 67.1% and 25.2% of the total number of applications in Japan. Sumitomo Mining mainly develops the pyrometallurgical recycling process to deal with retired rechargeable batteries. The South Korea Institute of Geology and Mineral Resources mainly carried out research work in four fields: geology, mineral resources, petroleum and marine, and geological environment. Its research on the recycling of rechargeable batteries resources mainly includes hydrometallurgy leaching, pyrometallurgy technology, recovery device, etc. Before 2013, there were many inventors engaged in research on waste battery recycling technology every year, and personnel turnover was frequent. However, the number of inventors working on the technology has changed little since 2014, but new core inventors have emerged in different periods. Moreover, the objects of patent processing also have a certain degree of dispersion, indicating that the technology in this field is developing vigorously, and more inventors have a good prospect in this field of technology and pay attention to their intellectual property protection.

2.1.3. Technical Composition of the Patent Application

The research distribution of the technology in different International Patent Classifications (IPC) can be revealed by analyzing the research field of the technology. As can be seen from the retrieval results (Figure 6), the rechargeable batteries recycling technology involved many fields of IPC classification technology, which were H01M (basic electrical components), C22B (metallurgy; ferrous or nonferrous metal alloys; alloy or nonferrous metal treatment), B09B (solid waste treatment), B03C (separation of solid materials by liquid or by wind shaker or wind jig; magnetic or electrostatic separation of solid materials from solid materials or fluids), C01G (inorganic chemistry); and B02C (crushing, grinding or pulverizing; grain milling pretreatment), mainly distributed in the H (electrical), C (chemical; metallurgy),
Part B (operation; transportation) several categories, and a small amount of section F (machinery, heating). It mainly involved the regeneration of rechargeable batteries components, extraction, and separation of metal elements, disassembly, or pretreatment of rechargeable batteries. As can be seen from Figure 6, the number of patent applications for secondary batteries resource recovery technology in the field of H01M, namely the battery field, far exceeds the research and application in other fields, which was caused by the rapid development of the LIB industry. In addition, the number of patents in the field of C22B occupied second place, indicating that the battery recycling technology mostly adopts the metallurgical method. The two were closely connected and have a strong combination. The research and application of B09B, B03C, C01G, and other aspects also showed more applications and the fields involved increased year by year, which indicated that this technology has a certain field crossover. The research of basic electronic components industry combined with batteries recycling technology has also derived the development of other related aspects of technology and technology fields. In general, the technical fields involved in waste rechargeable batteries recycling technology show an expanding trend, and the research heat in all fields shows a rising trend.

The process of rechargeable batteries recovery and comprehensive resource utilization included echelon utilization of decommissioned batteries, collection of waste batteries, pretreatment, extraction, separation of valuable resources, and products preparation. Due to the inconsistency of batteries, only individual battery packs or single batteries in the decommissioned batteries packs usually reach the state of being scrapped, while other batteries (groups) were still in the normal service period and have a high echelon utilization value. There are a lot of theoretical and technical problems to be studied and solved in the aspects of performance evaluation, sorting and grouping, integrated control, safety and economy, and business model. For retired rechargeable batteries’ state of health, the safety threshold value, the salvage value assessment, low-cost fast and efficient separation group, system control, large-scale collaborative optimization scheduling, and the key technical difficulties, we have to carry out the research from the following aspects. 1) Research and development of low-cost, extensible quick separation devices, batteries’ energy exchange system, and key equipment such as a large-scale distributed grid device. 2) Setting up of platform systems such as batteries energy management and control and automatic inspection the cloud platform, energy storage system grading collaborative management and control platform, and energy operation platform of cascade energy storage system; 3) Development of software systems such as health state assessment and life prediction systems of retired rechargeable batteries and multiscale collaborative optimization operation management system. It will effectively improve the efficiency, reliability, safety, life, and economic benefits of decommissioned power batteries’ cascade energy storage system. 4) Solving a series of industrial pain points existing in batteries, EVs, batteries energy storage, and other industries and providing technical support for the large-scale utilization of decommissioned power batteries and the connection of the whole power batteries industry chain and value chain.

By December 2020, there were 25 patents related to the ladder utilization of decommissioned batteries, covering the screening and recombination system of decommissioned LIBs modules for energy storage power stations, the classification method of decommissioned EV power batteries, the diagnostic method of decommissioned EV batteries packs, and the method of evaluating and detecting the utilization rate of discarded EV batteries. The earliest one was the actual capacity assessment method of decommissioned batteries modules applied by the Shanghai University of Electric Power in 2016.

There were 96 patents related to the waste batteries collection process, covering the recycling box and collection device. The pretreatment stage usually included physical methods, such as disassembly, crushing, screening, magnetic separation, washing, heat treatment, etc. and separation and recovery of shell, plastic, fluid collection, etc. The applied invention patents involved a total of 382 pieces of discharge devices, dismantling devices, shell-breaking devices, and crushing devices. In the process of extraction and separation of valuable metals, pyrometallurgy, hydrometallurgy, biological metallurgy, or a combination of various technologies are mainly used to extract valuable elements from cathode materials, anode materials, or electrolytes, including physical, mechanical, biological, electrochemical, and other recovery technologies. Hydrometallurgical technology has been widely used in various metal element recovery processes due to the simple process, low environmental pollution, and high selectivity of recovery, making the patent proportion relatively high. In addition to the use of physical and chemical technology to extract valuable metal elements, they can also be separated by magnetic separation device, cracking device, sorting device, separation device. A total of 1318 patents were applied, accounting for 54% of the total patent applications, indicating that researchers pay high attention to this field. The product preparation stage involves recovery device and regeneration technology, with a total of 722 related patents, as shown in Table 1.

2.2. Analysis of Published Literature on Rechargeable Batteries Recycling

It is impossible to classify and analyze each paper for each research field; nevertheless, one can highlight the major international research institutions, research directions, and funding status. This work aims to reveal the development trend and research
hotspots of the international rechargeable batteries recycling research and provide some reference for the overall layout of the national and related research institutions. The research progress and development direction in the field of rechargeable batteries recycling were clarified through statistical sorting and analysis of academic papers on rechargeable battery recycling from 1999 to 2020, aiming to promote the large-scale application of rechargeable battery recycling technology and the construction of the system.

2.2.1. Trend Analysis of Global Literature Publication

The number of articles published in a given field in a country is an indicator of the level of interest in that field. The trend of global publications in the past 20 years on spent rechargeable batteries recycling are shown in Figure 7. The number of literature on the recycling of spent rechargeable batteries has been on the rise since 1999 concentrated on lead-acid batteries and Ni–Cd batteries,[11] and the growth rate has been significantly accelerated after 2012 with more attention to LIBs.[12] The number of relevant literature published in 2018 increased by 60% compared with 2017.[13] It is expected that the number of publications related to rechargeable batteries recycling and cascade utilization will continue to maintain steady growth in the next two decades, and there is still a large space for exploration. Rechargeable batteries recycling started to develop in China in 2000 and has dominated this field since 2003.[14] In particular, the number has reached 134 in 2020, accounting for 45.27% of the total published articles in the world.[15] The scientific research strength of rechargeable battery recycling and utilization in China is gradually improving, and its attention is also rising steadily.

China received a total of 492 articles published, far higher than other countries and regions (54.30% of total global literature published), and ranked the first, second, three, four, five countries/regions, respectively, are Brazil[16] (53, accounted for 5.85%), India[17] (46, accounting for more than 5.08%), the USA[18] (41, 4.53%), and South Korea[19] (37, accounted for 4.08%), as shown in Figure 8. These five countries are the main publications of relevant literature in the world, while other countries/regions have relatively few publications. This may be related to the fact that China attached great importance to the development of new energy vehicles and has the largest new energy vehicle sales market in the world.[20] The large-scale production and use of new energy vehicles have largely driven the rapid expansion of the spent rechargeable batteries recycling market, which was conducive to driving new profits and industrial growth

![Figure 7. Rechargeable batteries recycling literature publication and trends (Global and China).](image1)

![Figure 8. Trends of publications in China, Brazil, India, the USA, and South Korea.](image2)

| Technological process | Specific technology | Application | Proportion [%] |
|----------------------|--------------------|-------------|----------------|
| Ladder utilization technique process | Ladder utilization | 25 | 0.6 |
| Waste batteries collection process | Waste batteries recycling bin | 76 | 3.5 |
| Pretreatment process | Waste batteries cleaning device | 2 | 13.2 |
| | Discharge device | 52 |
| | Pretreatment plant | 8 |
| | Dismantling device | 121 |
| | Hatching device | 11 |
| | Crushing plant | 121 |
| | Cutting device | 37 |
| Valuable metal extraction and separation process | Hydrometallurgy | 651 | 54.3 |
| | Pyrometallurgy and hydrometallurgy | 218 |
| | Physical method | 27 |
| | Mechanical method | 56 |
| | Biological methods | 7 |
| | Electrochemical process | 16 |
| | Magnetic separation equipment | 6 |
| | Segregation apparatus | 88 |
| | Cracking unit | 11 |
| | Sorting device | 22 |
| Product preparation process | Recovery device | 615 | 29.7 |
| | Renewable technologies | 107 |
points of related enterprises. The high number of papers in India may be related to the fact that it was used as an electronic waste disposal site.\cite{11} The decommissioned rechargeable batteries inevitably flow to India, which accordingly stimulates the research progress of rechargeable battery recycling in India.\cite{12} South Korea has been actively committed to rechargeable batteries industry technology study to ensure the competitive advantage in this field, thereby driving the rapid development of rechargeable batteries recycling research.\cite{19} The comprehensive and systematic incentive policies and recycling mechanism of waste batteries in the USA give advantages to the research and market application of power batteries and stimulate the development of waste battery recycling.

Brazil and South Korea began to conduct rechargeable battery recycling research in 2002.\cite{23} In addition, Brazil was in an active research period from 2007 to 2017 and has been growing steadily. India was a late starter, with the number of publications gradually increasing after 2013.\cite{24} As a “rising star,” India has shown great potential and development space in the utilization of spent LIB resources in recent years.\cite{22,25} Research on rechargeable battery recycling in the USA has been gradually evolving since 2014,\cite{26} with a significant increase in the number of articles published by 2020. The period of research activity was 2002 to 2006 and 2012 to 2018 in Korea, while the fault line occurred between 2007 and 2015.

2.2.2. Statistical Analysis of Major Global Institutions and Authors

The major high-yield institutions committed to battery recycling were mostly scientific research universities according to the statistical analysis of major institutions (Figure 9). Among the top 12 scientific research institutes with the largest number of publications, nine seats are in China, the rest in Espirito Santo Federal University\cite{27} (Brazil), Council of Scientific & Industrial Research\cite{28} (Indian), and Inst Geosci & Mineral Resources\cite{29} (South Korea). These institutions accounted for 30.91% of the global amount of published literature published in the literature, for the global new energy vehicles research in cutting-edge technologies in the field of power batteries recycling provides a bellwether.

The top five universities in the number of publications included Central South University\cite{30} (CSU, 52, accounting for 18.57% of the number of major institutions relevant literature published, in Chinese), Beijing institute of technology\cite{31} (article 45, 16.07%), Tsinghua University\cite{32} (THU, 28, accounting for more than 9.89%), Chinese Academy of Sciences\cite{33} (CAS, article 27, accounted for 9.64%), and Federal University of Espirito Santo\cite{34} (Brazil, 24, accounted for 8.57%). Central South University has accumulated many research achievements in the recycling of cathode materials for lead-acid batteries and LIBs.\cite{35} Beijing Institute of Technology has been engaged in the research of green and efficient recycling technology of LIBs since 1999. It has made outstanding contributions in the field of LIB recycling with natural green biodegradable organic acid system recycling and material high-value recycling technology.\cite{36} Tsinghua University has made great contributions to the separation and reuses technology of electrode materials for LIBs.\cite{37} The Chinese Academy of Sciences was a rising star in the field of battery recycling, with remarkable achievements in hydrometallurgical, mechanical, and chemical recycling.

Figure 9. High-yield organization of rechargeable batteries recycling literature.
technologies. Since 2005, the Federal University of Espirito Santo, Brazil, has been developing steadily in the field of battery recycling research, focusing on electrochemical recycling methods and the comprehensive utilization of electrode materials.

2.2.3. Analysis of the Composition of Literature Publishing Techniques

The recycling literature was divided in detail according to the types of batteries, and the composition diagram of the types of rechargeable batteries recycling literature was obtained, as shown in Figure 10a (Note: when literature involved the recycling of two or more retired rechargeable batteries, it was counted into the classification of HBs). The literature related to the recycling technology of retired LIBs accounted for the greatest proportion (669 articles, accounting for 73.84% of the total published articles, the same as below). There were 9.7%, 75.1%, and 0.7% references related to LIB anode, cathode, and electrolyte recovery, respectively. In addition, review literature on LIBs recycling accounted for 14.5% of the total published literature.

Therefore, research on the recycling technology of LIB cathode materials was intensive, which may be related to the retired LIB cathode materials that are rich in valuable metals and have high environmental and economic benefits. In addition, the number of literature on lead-acid batteries recycling ranked second (87 articles, accounting for 9.60%), while the numbers of literature on Ni-MH batteries (66 articles, accounting for 7.41%), Ni-Cd batteries (45 articles, 4.97%), and mixed batteries materials (39 articles, 4.30%) were less.

The types and compositions of rechargeable battery recycling literature are shown in Figure 10b. Studies on the recycling of spent rechargeable batteries from 1999 to 2003 are mainly focused on the recycling of lead-acid batteries and Ni-Cd batteries. The number of publications on LIBs recycling increased gradually after 2004, and it became the main body of rechargeable batteries’ recycling literature in 2006, which indicates that LIBs’ recycling has attracted more attention. In 2012, LIBs grew rapidly and gradually surpassed other types of batteries, which was attributed to the fact that LIBs gradually became the preferred power batteries for new energy vehicles.

Figure 10. a) Type composition of rechargeable battery recycling literature. b) Type composition and annual publication of rechargeable battery recycling literature. c) Documented composition of different battery systems and recycling technologies.
Therefore, the value of its precious metals promoted the research progress of LIBs’ recycling technology. In addition, the number of literature on the recovery of waste Ni–Cd batteries was very small. The number of literature published was zero in 2019 and 1 in 2020, indicating that the research and development of related technologies of Ni–Cd batteries has basically withdrawn from the attention of researchers.

The related literature mainly included two aspects: recovery technology research (711 papers) and recovery benefit and status analysis (169 papers). The recycling technology mainly includes pretreatment,[53] valuable metal recovery,[54] reuse, and comprehensive utilization of resources.[55] The number of literature related to the recycling of valuable metals was the highest (474 articles, accounting for 56.03% of the literature related to recycling technology). The recycling process mainly adopted hydrometallurgy,[56] pyrometallurgy,[35,57] high-efficiency composite technology,[58] electrochemical process,[59] mechanical chemical process,[60] physical process,[61] and biological metallurgy process.[62] Due to its advantages of simple process, high recovery rate, low environmental pollution, and high product purity, hydrometallurgical technology has become the most widely used recycling technology and the most published recycling technology at present.

Leached by hydrometallurgy process. The metal ions have excellent performance either in the resynthesis of electrode materials or in the comprehensive utilization of capacitors, catalysts, and other fields, so they have high research value. At present, more studies are inclined to recover metal ions from electrode materials and synthesize various alloys, metal oxides, metal-organic frameworks (MOFs), and other materials with high commercialization potential, such as supercapacitors, various catalysts, and adsorbents.[63]

The technical system composition was statistically analyzed according to the relevant literature of different batteries recycling systems (Figure 10c). The research on the recycling technology of spent LIBs was mainly based on hydrometallurgy[40] (257 papers), followed by recycling (117 papers), pyrometallurgy[65] (68 papers), and comprehensive utilization (67 papers). The reason was mainly related to the physical and chemical properties of key materials of LIBs. Lithium, nickel, cobalt, manganese, aluminum, copper, and iron in LIBs with the most valuable recovery can be efficiently leached by hydrometallurgy process. The metal ions have excellent performance either in the resynthesis of electrode materials or in the comprehensive utilization of capacitors, catalysts, and other fields, so they have high research value. At present, more studies are inclined to recover metal ions from electrode materials and synthesize various alloys, metal oxides, metal-organic frameworks (MOFs), and other materials with high commercialization potential, such as supercapacitors, various catalysts, and adsorbents.[63]

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The research on hydrometallurgical technology started early, and the number of relevant literature published showed a rapid growth trend from 2012 to 2018, indicating that hydrometallurgical technology experienced a rapid development process in the field of batteries resource recycling. After 2012, the research on the reuse and comprehensive utilization of waste rechargeable batteries increased gradually, which was the inevitable result of the steady development of the recycling technology of waste rechargeable batteries. Since 2018, high-efficiency composite technology has gradually attracted the attention of researchers due to the high metal recovery rate and short recovery process. The proposed high-efficiency compound technology provided a new idea for researchers, which can improve the recovery efficiency of valuable metal elements in the subsequent recovery steps through effective and meticulous pretreatment and simplify the complex separation and extraction process of each metal component in the subsequent solution. The number of literature related to hydrometallurgy decreased by 2019, while the number of literature related to pretreatment increased significantly, which was closely related to the development of efficient combination technology. The pretreatment process generally combined discharge, crushing, grinding, screening, and physical separation. The purpose of the pretreatment was to initially separate and recover the valuable part of the LIBs, to enrich high value-added materials such as electrode materials efficiently and selectively, and to facilitate the subsequent recovery process to be more efficient. In the relevant literature published in 2020, various recycling technologies all showed different growth trends; hydrometallurgy, pyrometallurgy, reuse, and comprehensive utilization showed rapid growth. Hydrometallurgy is mainly based on organic acids, while pyrometallurgy focuses on low-temperature conversion, reuse, and comprehensive utilization through molten procedures. Its “one-step method” and other efficient recovery processes have attracted more attention from scholars. It is expected that in the next few years, high efficiency combined use technology, reuse, and comprehensive utilization of resources will gradually become the mainstream of recycling technology, and the research on pretreatment and pyrometallurgy will be further intensified.

2.2.4. Analysis of Literature with High Citation Frequency

The citation frequency of a paper is an important index to evaluate the academic influence and quality of scientific research papers. It has been widely used to evaluate the academic level and achievement quality of a country, colleges and universities, scientific research institutions, and researchers. In this section, the citation frequency changes and distribution characteristics of research papers on spent rechargeable batteries recycling technology collected by Web of Science from 1999 to 2020 were analyzed in detail. The development trend, discipline influence, and academic influence were statistically analyzed. The statistical information of the top 10 literature with high citation frequency (a total of 11 literatures) in the statistical data of the research literature on recovery technology is shown in Table 2.

The top ten literatures with high citation frequency were all studies on the recycling of cathode materials of spent LIBs. Seven were hydrometallurgical technology studies, in which four
used inorganic acids to leach metal ions from cathode materials, ranking 4th,[69] 5th,[67] 8th,[34] and 10th,[68] respectively, and three organic acids to leach metal ions, ranking 1st, 3rd, and 8th, respectively (citric acid, malic acid, and ascorbic acid).[12a,69] Four studies were done on high-efficiency composite technology, all of which adopted pyrohydrometallurgical combined technology. Two studies were done on vacuum pyrohydrometallurgical combined technology ranked 7th[70] and 10th,[71] respectively. In addition, the literature ranked 8th and 10th (Tsinghua University) studied the technology of using ultrasonic treatment to improve the subsequent leaching efficiency, and the literature ranked 1st and 6th studied the resynthesis methods of electrode materials, which were respectively high-temperature solid-phase synthesis and sol–gel method. Among the 11 literature with high citation frequency, 2 literatures were published in Journal of Hazardous Materials, ranking 1st and 10th, respectively. 6 literatures were published in J. Power Source, ranking 1st, 4th, 5th, 6th, and 8th, respectively. Two articles were published in Waste Management, ranking the 3rd and 7th, respectively. No. 10 was published in the Chemosphere. Traditional hydrometallurgy has attracted much attention because it can effectively extract metal ions. With the attention to the environment, it is required to recycle with low-energy consumption, high efficiency, and pollution free as possible, so composite recycling technology has been developed.

3. Perspective of the Recycling of Rechargeable Battery

The comparison and summary of recycling processes for waste rechargeable batteries is shown in Table 3, which mainly includes pretreatment, recycling, and reuse processes.[73] The pretreatment process includes discharge, disassembly, and separation. The discharge technology mainly includes short discharge, liquid nitrogen low-temperature perforation. The separation technology mainly includes mechanical separation, acid/alkali dissolution, organic solvent dissolution, and heat treatment. The recycling process includes leaching/enrichment and separation and purification. Leaching/enrichment can be divided into pyrometallurgy recovery and hydrometallurgy recovery. Separation and purification refer to the transfer of metal components in active material of the positive electrode to solution by chemical solvent extraction and the leaching method. These recovered metals with high added value can be carried out through extraction, precipitation, adsorption, and

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**Table 2. Statistical information of literature with high citation frequency on the research of decommissioned power batteries recycling technology.**

| Number | Title | Year | Author | Institutions | Journal | Technology | Citations |
|--------|-------|------|--------|--------------|---------|------------|-----------|
| 1      | Recovery of cobalt and lithium from spent LIBs using organic citric acid as leachant | 2010 | Li Li, Wu Feng | Beijing Institute of Technology | Journal of Hazardous Materials | Hydrometallurgy | 247 |
| 2      | Development of a recycling process for LIBs | 2012 | Georgi-T.Georgi-Maschler, D-50 858 Cologne, Germany. | J. Power Source | Efficient composite technology | 247 |
| 3      | Environmental-friendly leaching reagent for cobalt and lithium recovery from spent LIBs | 2010 | Li Li, Wu Feng | Beijing Institute of Technology | Waste Management | Hydrometallurgy | 216 |
| 4      | Recovery of metal values from spent LIBs with chemical deposition and solvent extraction | 2005 | Nan Junmin, South China Normal University | J. Power Source | Hydrometallurgy | 209 |
| 5      | A laboratory-scale LIB recycling process | 2001 | Contestabile M, Scrosati B; Sapienza University of Rome | J. Power Source | Hydrometallurgy | 202 |
| 6      | Preparation of LiCoO2 from spent LIBs | 2002 | Churl KyoungLee; Korea Institute of Earth Sciences and Mineral Resources | J. Power Source | Efficient composite technology | 188 |
| 7      | Organic oxalate as leachant and precipitant for the recovery of valuable metals from spent LIBs | 2012 | Liang Sun, Keqiang Qiu; Central South University | Waste Management | Efficient composite technology | 174 |
| 8      | Ascorbic acid-assisted recovery of cobalt and lithium from spent LIBs | 2012 | Li Li, Wu Feng | Beijing Institute of Technology | J. Power Source | Hydrometallurgy | 161 |
| 9      | Hydrometallurgical separation of aluminum, cobalt, copper, and lithium from spent LIBs | 2009 | Daniel Alvarenga Ferreira, Marcelo Borges Mansur; Universidade Federal de Minas Gerais | J. Power Source | Hydrometallurgy | 161 |
| 10     | Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent LIBs | 2011 | Liang Sun, Keqiang Qiu; Central South University | Journal of Hazardous Materials | Efficient composite technology | 152 |
| 11     | A combined recovery process of metals in spent LIBs | 2009 | Li Jinhui; Tsinghua University | Chemosphere | Hydrometallurgy | 152 |
| Phase                  | Classification                   | Process                                                                 | Technology                        | Characteristics                                                                                                                                                                                                 |
|-----------------------|----------------------------------|-------------------------------------------------------------------------|------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Pretreatment          | Physical method                  | The electrode material is separated from other materials based on the difference of physical properties after the battery is crushed, mainly including magnetic separation, wind separation, screening, flotation, etc. | Mechanical treatment               | It is suitable for mass processing of batteries but has disadvantages such as introduction of impurities and loss of valuable materials.                                                                                                                                 |
|                       | Chemical method                  | Electrode materials are separated by chemical reaction process, and the efficient separation of materials is generally achieved by dissolving the fluid collector and destroying the binder. | Heat treatment                     | The binding force between the collector and the electrode active material is destroyed by heat treatment. Through ultrasonic and other auxiliary means, the separation process and separation effect can be more efficient.                                                                                                                                 |
|                       | Acid/alkali-soluble method       |                                                                          |                                    |                                                                                                                                                                                                                                                                         |
|                       | Organic solvent method           |                                                                          |                                    |                                                                                                                                                                                                                                                                         |
| Recycling process     | Pyrometallurgical technology     | A method of recovering valuable metal elements from positive electrode materials in the form of metals, alloys, oxides, etc., without using solution media, using methods such as high-temperature cracking, high-temperature reduction, and salt-dissolving roasting. Compared with the traditional high-temperature cracking method and high-temperature reduction method, the new composite technology molten salt roasting method can realize the full-component or selective recovery of the cathode material of the LIBs at a lower roasting temperature. | High-temperature pyrolysis method  | The structural instability of the positive electrode material in a high-temperature baking environment directly cracks the electrode material into compounds such as metals and alloys. This method has disadvantages such as high energy consumption, polluting gas emission, and loss of valuable metals. |
|                       | High-temperature reduction method |                                                                          |                                    | Using gas with reducing properties at high temperatures, active metals, coke, etc., as reducing agents, the reduction and recovery of valuable metals at lower temperatures can be achieved. Although the reaction temperature is lowered compared with the high-temperature cracking method, its energy consumption is still a problem to be solved.                                                                                     |
|                       | Molten salt roasting method       |                                                                          |                                    | By means of the chemical transformation reaction of cathode materials in a high-temperature molten salt environment, the high-priced insoluble electrode materials can be converted into low-priced soluble salts and oxides. The difference of reaction systems in this method mainly lies in the selection of different dissolved salts, acids, and alkaline. The selection of appropriate reaction reagents can effectively reduce the reaction temperature of the system and also achieve the effect of selective separation of valuable metals, which is one of the directions for the future short process and efficient recovery of electrode materials for LIBs. |
### Table 3. Continued.

| Phase Classification | Process | Technology | Characteristics |
|----------------------|---------|------------|-----------------|
| **Mechanochemical method** | The method of applying mechanical energy, usually mechanical ball milling, is used to cogrind the cathode material of the battery and the grinding aid to convert it into the material that is easy to leach and easy to be recovered later. This method is generally suitable for laboratory research, and industrial-scale recycling technology needs further research. |
| **Hydrometallurgical technology** | A process whereby valuable metals in electrode materials are transferred from solid phase to liquid phase through chemical/electrochemical, biological leaching, and other reactions, and then metal ions in the liquid phase are separated and enriched and finally recovered in the form of metals or other compounds. The separations and purification methods of organic metals mainly include ion exchange, extraction, precipitation, electrochemical deposition, and so on. Although the process of wet recovery is more complex, the recovery rate of valuable metals is higher, and it is the mainstream of industrial recovery technology. |
| **Acid/alkali leaching method** | In the process of traditional acid/alkali leaching recovery technology, acid leaching usually takes inorganic acid as leaching agent and hydrogen peroxide as reducing agent to reduce and dissolve high-priced insoluble compounds. Alkaline leaching generally takes ammonium chloride and other amino system solvents as leaching agent, ammonium sulfate as reducing agent and transition metal elements in cathode materials to form complex, so as to selectively leach valuable metals from the original stable compounds. In addition, green organic acid can also be used as leaching agent for leaching. The traditional acid/alkali leaching recovery technology, the leaching process will produce a lot of waste water and other by-products, which poses a potential threat to the environment. The acid leaching method using green organic acid as leaching agent can recover valuable metals in electrode materials green and efficiently, which is one of the ideal directions for the development of battery recycling technology in the future. |
| **Deep eutectic solution method** | Deep eutectic solvents are a class of compounds with an extremely high ability to dissolve metal oxides. As effective leaches and reducing agents, they can achieve high purity metal extraction without reducing agents and/or expensive solvent extractants, and without complex processes. However, in the leaching process, the leaching rate of valuable metals is relatively low, and it is difficult to realize selective separation of elements, which is still in the research and exploration stage. |
| **Electrochemical method** | Using electrochemical electrolysis to recycle cathode materials in suspension or molten salt can avoid the addition of leach and reducing agent in the leaching process, but its energy consumption and operation still need further technological improvement and technical breakthrough. |
| **Microbial metallurgy** | The valuable metals in the solid phase are dissolved through the natural metabolic process of microorganisms, such as oxidation, reduction, complexation and acidolysis. The useful components of the system are converted into soluble compounds and selectively dissolved and separated to obtain the solution containing valuable metals. The separation of target components and impurity components is realized, and the valuable metals are finally recovered. At present, the technology has just started, and there are still many problems to be solved, such as the breeding of efficient strains, the long culture cycle, and the control of leaching conditions. |
| Phase                | Classification | Process                                                                                                                                                                                                 | Technology                                      | Characteristics                                                                                                                                                                                                                                                                                                                                                     |
|---------------------|----------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| External recycling | Reuse of chemical grade product | With the products after recycling as raw materials, through chemical precipitation, high-temperature roasting, chemical extraction, ion exchange, electrochemical deposition, and other ways, we obtain a variety of valuable metal sulfates, chlorides, oxides, metal elemental chemical products. Although this method is simple to operate, the added value of chemical products obtained is low. Now more studies focus on internal recycling systems such as direct repair and regeneration of electrode materials and resynthesis. | Chemical precipitation method                   | With the different solubilities of valuable metal ions in the leaching solution in different precipitators, the valuable metal ions with different solubilities form insoluble compounds, and the insoluble valuable metals will be obtained by acidolysis again, and then chemical products can be obtained by means of reprecipitation and recrystallization. Chemical precipitation has high efficiency in purification and separation, and it is also the simplest and cost-effective separation and recovery method. However, its product purity is low, which requires multiple purification treatments.                                                                                                                                                                                                                                                                 |
| High-temperature roasting |                | Valuable metal oxides were obtained by directly calcining the leached product at high temperature. Although this method is simple to operate, it has the disadvantages of high-energy consumption and low product purity, so it is generally not applied to the recovery and treatment of products with high added value. |                                          |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |
| Chemical extraction method |                | The electrode material after leaching, extracting agent added to the leaching solution, using lithium, nickel, cobalt, manganese, and other metal ions in organic phases and water phase, and the solubility or partition coefficient of different specific metal ions from the water phase extraction to the organic phase, through reverse extraction, can get the valuable metal sulfate or chloride, after recrystallization can get high-purity chemical products. The chemical extraction method makes up for the disadvantage of the low purity of the product extracted by chemical precipitation. The two methods can also be used together to obtain the chemical products that meet the industrial needs. Although the chemical extraction method has the advantages of high product purity and selective extraction, the organic extraction agent is more expensive and flammable. |                                          |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |
| Ion exchange process |                | The different adsorption coefficients of ion exchange membranes on valuable metal ion complexes are used to realize metal separation and extraction. After the extraction of valuable metal elements, the chemical raw materials that meet the requirements are obtained by means of dissolution, precipitation, and recrystallization. The process is simple and easy to operate, but the process is long and the ion exchange membrane is expensive. |                                          |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |
| Electrochemical deposition method |                | Under the action of an external electric field, the current flows through the positive electrode material to leach the solution, and the valuable metal elements in the leachate undergo oxidation–reduction reaction to form a coating on the electrode, thereby obtaining high-purity metals and other chemical products. By controlling experimental conditions such as current density, metal films of different thicknesses can be obtained, but during the metal film deposition process, the purity of the leaching solution is relatively high. |                                          |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |
| Phase                        | Classification | Process                                                                                                                                         | Technology                                                                 | Characteristics                                                                                                                                                                                                 |
|------------------------------|----------------|------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Internal recycling           |                | As one of the main technical systems of internal recycling, the direct repair and regeneration method is to use the electrode materials obtained after pretreatment as raw materials, by adding lithium sources and other substances, using in situ roasting, electrochemistry, and other repair technologies to supplement the elements, and finally to repair and regenerate the electrode materials that have partially lost or lost electrochemical properties. | Solid-phase in situ lithium replenishment method                          | After adding lithium salt or other compounds, the pretreated electrode material is entered into the lithium-deficient cathode material through particle contact under the condition of high-temperature roasting, so the electrochemical performance of the cathode material can be recovered. This method needs to add excessive lithium source to achieve the purpose of repair, which is easy to cause the waste of lithium resources. |
| Direct restoration            |                |                                                                                                                                                    | Electrochemical lithium supplement method                                  | Lithium ion is embedded into the cathode material through electrochemical reaction. Usually, the electrode material and lithium metal are composed of a half battery. Lithium-rich dissolved salts or lithium metals are used as lithium replenishment agent to compensate and embed the lithium missing in the cathode material. Compared with solid-state high-temperature in situ remediation, electrochemical lithium remediation can better control the amount of lithium, but the process parameters need to be strictly controlled during electrochemical lithium remediation. |
| Regeneration                  |                |                                                                                                                                                    | Solid-phase synthesis method                                                | Taking the leaching solution as raw material, adding the corresponding missing metal elements to prepare the precursor, the prepared precursor and lithium source under the action of high-temperature, through atomic or ion diffusion, reactant interface contact, reaction, nucleation, crystal growth, so as to get a new regenerated cathode material. Although the high-temperature solid phase synthesis method has the advantages of simple synthesis process, the uniformity of product components is difficult to control, and in the synthesis process, the reaction atmosphere and experimental conditions need to be strictly controlled. |
| Resynthesis of electrode      |                | Electrode material resynthesis method is a method to generate new electrode materials by means of electrode material synthesis process after adding missing metal elements in electrode materials according to the chemical stoichiometric proportion of leaching solution, oxides, and other compounds as raw materials. | Sol–gel method                                                             | The recovered products are dispersed in the solvent, hydrolysis/repolymerization reaction takes place in the solution, and then the sol–gel is formed. Finally, after drying and heat treatment, the resynthesized electrode material is obtained. Sol–gel method can solve the problems of slow diffusion between reactants and poor homogeneity of components in high-temperature solid-phase synthesis method, but it has the disadvantages of a long time-consuming and long process. |
| materials                    |                |                                                                                                                                                   |                                                                           |                                                                                                                                                                                                               |
| Sol–gel method                |                | The recovered products are dispersed in the solvent, hydrolysis/repolymerization reaction takes place in the solution, and then the sol–gel is formed. Finally, after drying and heat treatment, the resynthesized electrode material is obtained. Sol–gel method can solve the problems of slow diffusion between reactants and poor homogeneity of components in high-temperature solid-phase synthesis method, but it has the disadvantages of a long time-consuming and long process. |                                                                           |                                                                                                                                                                                                               |
| Hydrothermal                  |                | In a special closed reaction vessel (autoclave), with water as the main medium, through heating to create a supercritical state (temperature: 100–1000 °C; the pressure is 1 MPa–1 GPa) and the new electrode material is prepared. This method can replace some high-temperature solid reaction to produce electrode materials with special morphology. |                                                                           |                                                                                                                                                                                                               |
electrolysis. The reuse process can be divided into external recycling and internal recycling. The former refers to the reuse of chemical products, while the latter refers to direct recovery and recycling and resynthesis of electrode materials. The reuse of chemical products mainly included chemical precipitation, high-temperature roasting, chemical extraction ion exchange, electrochemical deposition, and other technical means to produce nickel sulfate, cobalt sulfate, manganese sulfate, cobalt chloride, and manganese. Direct restoration and regeneration mainly included solid-phase-supplementing lithium method and electrochemical-supplementing lithium method to repair the crystal structure and electrochemical properties of the failed electrode materials. The resynthesis of electrode materials mainly adopted high-temperature solid-phase synthesis, sol–gel, hydrothermal synthesis, and electrodeposition regeneration to synthesize new precursors or electrode materials. Based on the above in-depth analysis of the recycling technology of waste rechargeable batteries, the recycling technology can be divided into traditional recycling technology, green recycling technology, and next-generation recycling technology.

### 3.1. Traditional Recycling Technology

Traditional prevention and control of rechargeable batteries waste pollution and resource utilization mainly include the following measures: 1) bring solid waste into the scope of resource management, 2) develop material recycling technology, 3) formulate solid waste management laws and regulations, and 4) reform production processes to reduce waste discharge. The task of recycling, disposal, and phased utilization of large-scale rechargeable batteries after centralized decommissioning was arduous. The traditional recycling process usually gives full consideration to efficiency and economy. From 1999 to about 2010, traditional recycling technologies, such as soil landfill and incineration, often ignored the secondary pollution caused by these processes and have problems such as high energy consumption and low economy. There were corresponding laws and regulations at the national level, yet perfect to regulate the recycling industry.

Overseas research on recycling of waste lead-acid batteries has been carried out for a long time and more experiences have been obtained, and the recycling efficiency has also been significantly improved. Although great progress has been made in the recycling and regeneration of lead-acid batteries, there are still some problems, such as low recovery rate and serious environmental pollution. With the production and consumption of more lead-acid batteries, too low recovery rate affects the reuse of lead resources. Therefore, the recycling of waste lead-acid batteries needs to start from two aspects of management and technology and gradually improve. The main factors affecting the hydrometallurgy treatment of spent Ni–MH batteries include acid type, acid concentration, leaching temperature, leaching time, liquid–solid ratio, stirring intensity, etc. Researchers generally determine the best treatment process under the influence of these factors through orthogonal experiments. Abundant achievements have been made in the treatment of waste Ni–MH batteries according to the current research reports. However, most of these treatment methods are still limited to the laboratory research stage, with few large-scale applications.

### 3.2. Green Recycling Technology

The green recycling of rechargeable batteries can be realized through step utilization or seeking the green recycling technology. Given the recycling and resource utilization strategy of decommissioned LIBs, the national ministries and commissions suggested that the principle of “making the best use of everything” should be followed and that the first step utilization should be followed by the comprehensive utilization of recycled resources. Retired LIBs still have 60–80% rated capacity after rediagnosis, sorting, and reorganization and can be used in scenarios with milder operating conditions, such as slow-speed EVs, power grid energy storage, base station standby, etc., to realize the step-by-step utilization of LIBs. Escalated utilization of batteries can give full play to their performance and improve their full life cycle value, which conforms to the “4R” principles of environmental protection, namely, Recycle, Reuse, Reduce, and Recover, and has potential economic value and good social benefits. With the continuous development and expansion of the new energy EV industry, the scale of retired LIBs is increasing. How to make good cascade utilization will be one of the key development directions in the future. However, due to the un unified technical standards, lack of industry supervision, and insufficient scale in the early development of the retired LIBs echelon recycling industry, the economy of echelon recycling is low. Optimistically, in the long run, these problems are expected to be solved as the production of LIBs becomes more standardized and the country's regulation of EVs and power batteries becomes more perfect.
In contrast to the traditional recycling technology, more environmentally friendly and more economical green recycling technologies have been developed since 2011, including organic acid extraction technology, nondestructive repair and regeneration technology, and short-range target metal extraction technology, which are also the mainstream laboratory recycling technologies at present. In addition to considering efficiency and economy at this stage, environment was also taken into consideration, forming a representative “3E” recycling concept. However, due to the need for fine control of recycling conditions, related technologies have not been widely used.

3.3. Next-Generation Recycling Technology

The current recycling model is passive, that is, R&D of resource utilization technologies for waste materials. To meet ecological sustainability, the next-generation recycling technology needs to achieve the full component recovery of key battery components and carbon-neutral recycling process as much as possible. Based on the above analysis of the application of rechargeable batteries recycling technology, the following opinions and suggestions are put forward for the next generation of recycling technology. It is crucial to transform passive recycling into active recycling mode in next-generation recycling. Technically, it is necessary to follow the “5M” principle (Figure 11). The first principle is that the source is more leading. For example, self-healing and self-repairing materials and easy-to-disassemble and easy-to-block battery systems can be developed for electrode materials and battery systems, respectively. In addition to adhering to the principle of more economical, more environmentally friendly, and more efficient, recycling technology should also emphasize on being more safe.

In terms of policies, it is necessary to implement the “3S” principles of strict control of standard layout, strict control of technology promotion, and strict control of responsibility management (Figure 11). Specifically, the promulgation of relevant industry standards can be further promoted to enable battery recycling companies to meet standard requirements, reduce corporate costs and risks, and encourage technological innovation. However, relevant legal issues must be properly dealt with to avoid technological monopoly, and the principle of fairness, rationality, and nondiscrimination must be implemented. Finally, the layout of the traceability system, management mechanism and data producer responsibility promotion system in the whole life cycle of battery technology to promote the complementary advantages between different segments of the industry, to determine the responsibility in the production of raw materials, production, equipment assembly, use, consumption, recycling and other aspects to enhance the policy effectiveness of participation and practice.

4. Conclusion

Based on bibliometrics, this work thoroughly analyzed the development status of rechargeable battery recycling strategies through content analysis involving the patents and scientific literature, thus giving a comprehensive quantitative description for the first time. Based on quantitative analysis, whole existing recycling strategies for rechargeable batteries were presented and discussed. If the background of the current rechargeable batteries recycling business model is not clear, it is necessary to establish a complete link related to rechargeable batteries recycling policies and regulations, technical research, market system, and environmental awareness, to jointly promote the construction of rechargeable batteries recycling model. According to the current situation of the industry, the number of patents and publications in the field of rechargeable batteries recycling will continue to increase and will enter a new period of development. The recycling technology should be further promoted with enterprises and scientific research institutions as the joint innovation subject, market oriented, the combination of industry, education and research, and the combination of policy and market guidance to promote the sustainable and healthy development of related technical fields. The main conclusions of this work are as follows. 1) Since 1999, the number of patent applications and issued papers on waste rechargeable battery recycling has been on the rise year by year, indicating that waste rechargeable battery recycling has received extensive attention at home and abroad and the development trend was largely the same. Compared with international research, there is a certain lag in China, but the research development trend is roughly the same as the international research. 2) The international recycling treatment of waste rechargeable batteries is mainly based on hydrometallurgical technology, including acid/alkali leaching method, deep eutectic solvent method, electrochemical method, followed by pyrometallurgical technology, including high-temperature pyrolysis method, high-temperature reduction method, and molten salt roasting method, and the research on biological metallurgical recycling technology is relatively few. 3) Within the scope of the top ten journals in the field of waste rechargeable battery recycling and treatment, most of the articles are published in high-level journals. The scientific research institutions with a large number of articles are mainly in countries with a large demand for rechargeable batteries.

In addition, the following four suggestions are put forward to achieve the goal of green recycling of rechargeable batteries. First, encourage the development of simple and efficient multi-component recycling technology to overcome the disadvantages
of various technical means. Second, from the perspective of economic cost accounting, we should reduce the recovery cost and increase the added value of products to ensure the economic benefits of rechargeable batteries recycling enterprises. Third, from the perspective of environment and safety, environment-benign electrode materials, binders, membranes, and electrolyte systems should be selected to reduce the environmental burden. Fourth, improve the battery recycling data tracking data platform to provide technical support for policy formulation and product development.

5. Experimental Section

The Derwent Innovations Index SM (DII) international patent database was used as the source of patent data in this work. As the most comprehensive patent citation database, DII cataloged more than 20 million essential invention patents from more than 40 patent offices worldwide, covering more than 100 countries. The retrieval methods in the DII database were as follows: 1) retrieval strategy, using advanced retrieval mode, \( [\text{T}] = \{\text{spent battery} \text{ OR waste battery} \text{ OR retired battery}\} \); 2) retrieval time, January 2021; 3) retrieval range, 1999–2020; and 4) the number of projects related to battery recycling was 3036 after the retrieval of data and manual denoising. As the lag period from patent application to publication was about 12 to 18 months on average, most of the patents filed in 2019 and 2020 were published; thus, the statistical results of patents in these two years were only for comparative reference. Based on the above data statistics, excel and other analysis software were used for patent data indexing, and through the patent information, the technical route was deeply interpreted and analyzed.

The literature data came from the Web of Science (WoS) core collection. As the world’s largest comprehensive academic information resource covering the largest number of disciplines, WoS contained the most influential and more than 8700 core academic journals in various research fields such as natural sciences, engineering technology, and biomedicine. It is an internationally recognized database reflecting the level of scientific research. Using “recycle batteries” as the search query, the retrieval time range was from 1999 to 2020, and the retrieval time was January 2021, and “article” and “review” literature were screened out. After manual denoising, 906 literature related to rechargeable batteries recycling were obtained. The retrieved papers on rechargeable batteries recycling were statistically analyzed from multidimensional perspectives, such as the overall international situation, the extent and concentration of countries’ participation in the research, the productive institutions and authors of the papers, and the classification of research fields.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

bibliometrics, patents, published literatures, rechargeable batteries, recycling

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