Electrochemical Energy Reviews (2024) 7:28
https://doi.org/10.1007/s41918-024-00220-1

REVIEW ARTICLE

Towards High Value-Added Recycling of Spent Lithium-Ion Batteries for Catalysis Application

Ruyu Shi[1] · Boran Wang[1] · Di Tang[1] · Xijun Wei[2] · Guangmin Zhou[1]

Received: 10 June 2023 / Revised: 31 December 2023 / Accepted: 20 March 2024

Abstract

With the proposal of the global carbon neutrality target, lithium-ion batteries (LIBs) are bound to set off the next wave of applications in portable electronic devices, electric vehicles, and energy-storage grids due to their unique merits. However, the growing LIB market poses a severe challenge for waste management during LIB recycling after end-of-life, which could cause serious environmental pollution and resource waste without proper treatment. Pyrometallurgical, hydrometallurgical, and direct recycling of spent LIBs have been developed, guided by the "waste to wealth" principle, and were applied to LIB remanufacturing. However, some spent LIB materials with low values or great direct regeneration difficulties may not be suitable for the above options, necessitating expanded application ranges of spent LIBs. Considering their unique compositions, using waste electrode materials directly or as precursors to prepare advanced catalysts has been proposed as another promising disposal technology for end-of-life LIBs. For example, transition metal elements in the cathode, like Ni, Co, Mn, and Fe, have been identified as catalytic active centers, and graphite anodes can serve as the catalyst loading matrix. This scheme has been adopted in various catalysis applications, and preliminary progress has been made. Therefore, this review summarizes and discusses the application of spent LIB recycling materials in catalysis and classified it into three aspects: environmental remediation, substance conversion, and battery-related catalysis. Moreover, the existing challenges and possible foci of future research on spent LIB recycling are also discussed. This review is anticipated to mark the start of close attention to the high-value-added applications of spent LIB products, enhancing economic efficiency and sustainable development.

Keywords Spent lithium-ion batteries · Recycling · Catalyst · Environmental remediation · Substance conversion · Battery-related catalysis

1 Introduction

The past two decades have witnessed the wide applications of lithium-ion batteries (LIBs) in portable electronic devices, energy-storage grids, and electric vehicles (EVs) due to their unique advantages, such as high energy density, superior cycling durability, and low self-discharge [1–3]. As shown in Fig. 1a, the global LIB shipment volume and market size are growing rapidly and are expected to reach 1 135.4 GWh and 99.4 billion dollars in 2025 [4], respectively. LIBs comprise the cathode, anode, electrolyte, and separator, and lithium ions migrate back and forth between the cathode and anode for energy storage and release [5, 6]. The cathode materials [e.g., $LiCoO_2$ (LCO), $LiMn_2O_4$, $LiFePO_4$ (LFP), and $LiNi_xCo_yMn_zO_2$] often undergo irreversible phase transitions after excessive charging/discharging cycles [7–9], and the interlayer structures of the anode materials (e.g., graphite) collapse [10], which are the main causes of battery capacity decay. Consequently, the average LIB lifespan is limited to 5 to 8 years due to the intrinsic structural barriers of electrode materials. With the accelerated updating and upgrading of electronic products, a lot of LIBs are discarded before reaching their maximum service life. Therefore, tens of millions

Xijun Wei
xijunwei1992@swust.edu.cn

Guangmin Zhou
guangminzhou@sz.tsinghua.edu.cn

[1] Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen 518055, Guangdong, China

[2] State Key Laboratory of Environment-Friendly Energy Materials, School of Materials and Chemistry, Southwest University of Science and Technology, Mianyang 621010, Sichuan, China

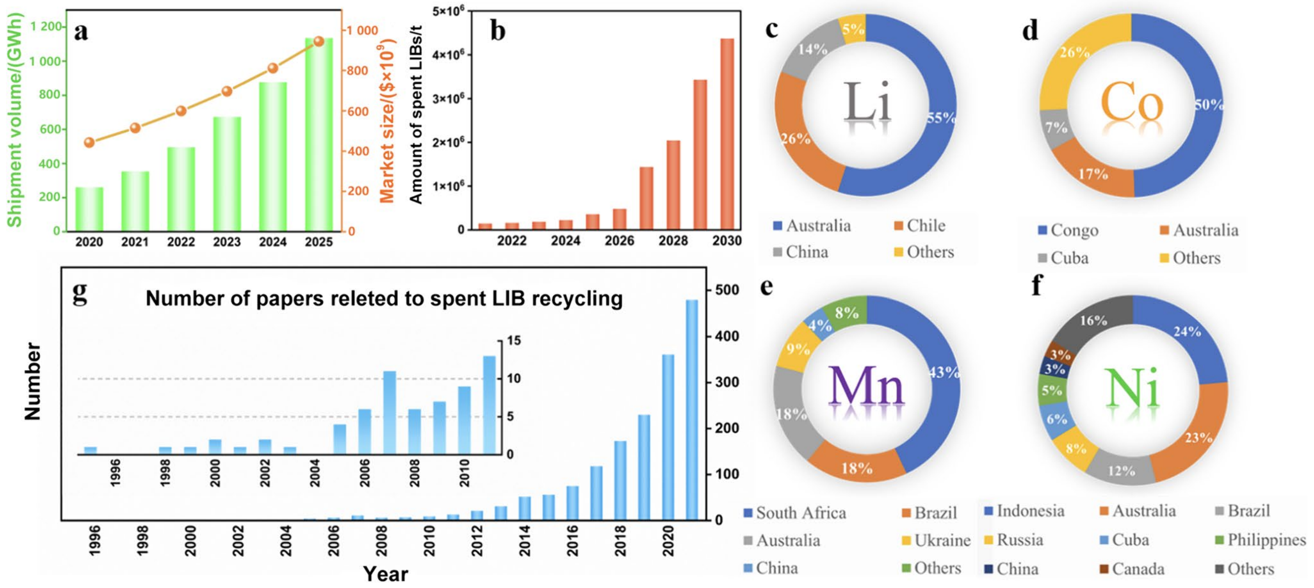

Fig. 1 **a** The global LIB shipment volume and market size from 2020 to 2025 [4], respectively. **b** Forecasts of the amounts of end-of-life LIBs generated in the near future [11]. **c–f** Geographical distribution of lithium, cobalt, manganese, and nickel in the earth's crust, respectively. **g** The number of papers related to spent LIB recycling (obtained from Web of Science)

of tons of spent LIBs are expected in the next 3 to 7 years, and a cumulative 12 million tons of end-of-life LIBs from EVs are expected by 2030 (Fig. 1b) [11]. The proper disposal of these spent LIBs poses a huge challenge but also presents some opportunities. To date, most spent LIBs end up in incineration furnaces or landfills like conventional waste due to the lack of relevant standards, sufficient economic efficiency, and efficient collection and recycling channels [12–14]. Their crude incineration emits CO_2 and toxic gases, exacerbating the greenhouse effect and atmospheric contamination, while their landfills leak hazardous fluorine, organic pollutants, and heavy metals, which seriously pollute soil and groundwater [15–18]. Therefore, effective recycling technologies are urgently needed for spent LIBs.

The necessary mineral resources for LIB productions, including lithium, cobalt, nickel, and manganese ores, have uneven geographical distributions and are mainly found in a few countries (Fig. 1c–f) [19, 20]. According to statistics, the global LIB market has already consumed 40% and 25% of all lithium and cobalt mining capacities, respectively [21]. Considering that the contents of many high-value metal elements, such as cobalt, lithium, copper, manganese, and nickel, in spent LIBs are higher than the corresponding natural ores, spent LIBs are regarded as mines for exploitation. Therefore, developing advanced technologies for recovering, recycling, or reusing these high-value components can mitigate resource constraints and eliminate environmental pollution while generating economic benefits [22]. However, the economic feasibility of spent LIB recycling poses a key challenge restricting its further large-scale development. Specifically, spent LIB recycling is a complex systematic project hindered by inadequate recycling facilities and technologies [23, 24]. At present, the recycling work is mainly driven by legislative obligations, with less consideration of economic returns. Thus, developing efficient, inexpensive, and environmentally friendly spent LIB recycling methods and scaling them up is crucial in future research.

Basic academic research has always been the cornerstone of industrial applications. Based on the "waste to wealth" concept, research on spent LIB recycling has increased exponentially from 2010 to 2021. According to the scientometric analysis based on the Web of Science core database, the number of relevant studies is shown in Fig. 1g. Besides recycling, spent LIBs can be utilized through remanufacturing and repurposing. Remanufacturing and repurposing are the most economically feasible disposal methods, which also extend the utilization of spent LIBs. Theoretically, the value of spent LIBs can be maximized through remanufacturing or repurposing, followed by recycling [25]. For example, spent LIBs from EVs can be repurposed for energy storage stations, electric tricycles, and communication base stations as they retain 80% of their initial energy or power density. Spent LIBs that cannot achieve echelon utilization can be recycled for raw materials. Researchers have proposed some simple and economical regeneration methods to repair the structure and composition of spent electrode materials [26–30]. Conventional pyrometallurgy, hydrometallurgy, bio-metallurgy, or their combinations for the sustainable development of LIBs have received more attention, and many reviews have summarized these spent

LIB recycling processes and closed-loop re-fabrication of electrode materials [31–34]. The direct regeneration method, i.e., using repaired spent LIB materials for new batteries, simplifies the recycling procedure and minimizes energy inputs, thus considered a more promising approach [35–38].

Despite the numerous spent LIB recycling strategies, many obstacles hinder their large-scale application or sustainable development. Owing to the mature technology, some pyrometallurgy and hydrometallurgy methods have achieved commercialization. However, as their profit model relies on valuable metals or their derivatives, such methods are only suitable for cathode materials with valuable metals (e.g., $LiCoO_2$ and $LiNi_xCo_yMn_zO_2$). In addition, the Co content in LIBs gradually decreases with the development of advanced LIB technologies. That is, “high-Ni and low-Co” LIBs are gradually replacing existing commercialized LIBs, thus reducing the profit margins of pyrometallurgy and hydrometallurgy methods that motivate their sustainable development, as Co products are their main source of profit. Meanwhile, previously reported direct regeneration methods are only suitable for regenerating one single component of spent LIB materials, while cathode materials of different components, even of the same type (e.g., $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ and $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$) are not able to be regenerated with the same process. In addition, the cost-effective regeneration strategies for the relatively low-value graphite, the main material for commercial LIB anode, are comparatively lacking. Therefore, it is necessary to explore new research directions for spent LIB recycling and expand the application of spent LIB recycling products.

The main components of LIB cathode, like Ni, Co, Mn, and Fe, are common in various catalysts. Meanwhile, the crystal structure of the degraded electrode materials is not completely destructed, which has potential catalytic activities, and the vacancies or other defects in spent LIB electrode materials may also promote the catalytic kinetics. Considering these intrinsic characteristics of spent LIB electrode materials, it is promising to convert spent LIBs into catalysts, and researchers have explored the application of spent LIB recycling products in catalysis, including pollutant degradation [39], photocatalysis [40, 41], electrocatalysis [42, 43], and high-value graphene [44, 45]. Although these alternative applications of recycled spent LIB materials present novel concepts for LIB recycling, few summaries or assessments have been conducted. Thus, a comprehensive review is crucial to guiding the benign development of this field.

This review focuses on the up-to-date research progress in catalysis-related applications of spent LIB recycling products, and the review outline is illustrated in Fig. 2. First, spent LIB pretreatments, including discharging, dismantling, and separation, are discussed and summarized in Sect. 2. Second, the feasibility of using spent LIB recycling materials as catalysts is discussed in Sect. 3. In addition, the specific research progress of spent LIB recycling materials in catalysis is summarized in Sects. 4, 5, and 6, including environmental remediation [degrading pollutants from wastewater and removing volatile organic compounds (VOCs)], substance conversion (biomass, synthesis gas, and electrocatalytic water decomposition), and battery-related catalysis [metal-air batteries, fuel cells, and lithium-sulfur batteries (LSBs)], respectively. As a reminder to the involved researchers, limitations on spent LIB recycling materials for catalytic applications are analyzed in Sect. 7. Moreover, the future challenges and opportunities for LIB recycling are considered from our perspective in Sect. 8, aiming to provide a reference for related research.

2 Before Converting Spent LIB Materials into Catalysts

Pretreatment is vital before recycling spent LIB components, including anode materials, cathode materials, current collector, diaphragm, etc. Effective pretreatments improve recovery efficiency and reduce energy consumption for subsequent steps [24, 46, 47]. According to previous experience, the pretreatment of spent LIBs mainly involves discharge, disassembly, and separation [47, 48].

The residual electricity in spent LIBs could trigger thermal runaway and cause irreparable disaster during recycling [49]. To ensure recycling safety, discharging spent LIBs is usually the first process of pretreatments, which includes physical and chemical methods. Physical discharge relies on external load circuits to consume the remaining electricity through heat release. However, this method is only suitable for discharging a few batteries and mainly applies to lead-acid batteries instead of LIBs. Chemical discharge is the most applied way to discharge spent LIBs by immersing batteries in aqueous solutions (e.g., NaCl and Na_2SO_4) to release the remnant energy. Relevant results showed that spent LIBs could be safely disassembled when their voltage was below 1.5 V [state of charge $(SOC) < 5\%$] [50]. However, high-efficiency discharge solutions usually severely corrode the battery shell, inducing safety accidents. Researchers have focused on mitigating or avoiding battery corrosion during discharging. For example, Fang et al. found that compared with other solutions, the zinc acetate solution can alleviate battery corrosion during discharging [49]. Wu et al. found that battery corrosion in electrolyte solutions can be solved using graphite electrodes instead of the battery tab for discharge [50].

At present, spent LIBs are often dismantled manually, lacking mature automatic disassembly methods [12]. Since some components of Li-ion cells react with O_2 and H_2O, a

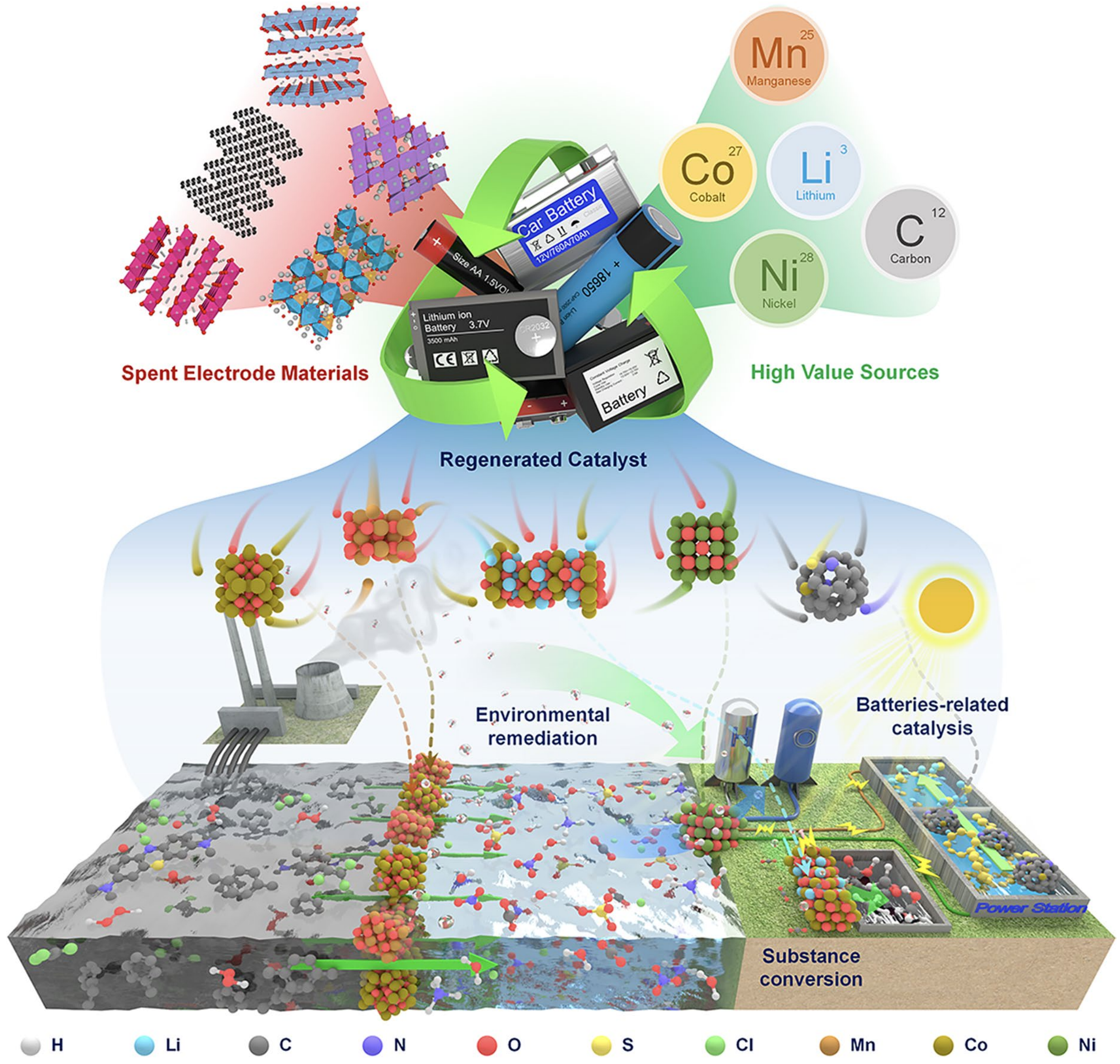

Fig. 2 The conceptual graph of spent LIB electrode materials applied in catalysis

glove box filled with highly pure Ar atmosphere containing H_2O and O_2 only in the lower parts per million range has to be utilized. Although discharging usually precedes battery disassembly, care must be taken to prevent internal circuit shorting during cell opening due to residual electricity. Therefore, an ideal cutting position has to be determined for each cell type through non-destructive techniques [e.g., X-ray analysis, X-ray computed tomography (CT), and neutron tomography] before opening spent Li-ion cells, and non-conductive tools are recommended [51]. Furthermore, it is reported that cryogenic treatment (e.g., liquid nitrogen) may reduce the risk during dismantling [52, 53].

After dismantling, effective separation of spent LIBs is essential for the subsequent precise recycling. Depending on the different physical properties (e.g., particle size, density, weight, and magnetism) of various components (e.g., cathode, anode, current collector, and diaphragm), various physical separation methods (e.g., crushing, sifting, and magnetic separation) can be utilized, and the electrolyte can be recovered if its content is sufficiently high [51, 54]. However, most studies neglect electrolyte recovery, which can be removed by heating, organic solvent washing, or supercritical CO_2 extraction [24, 25, 47].

Through appropriate discharging, dismantling, and separation, the different components of spent LIBs can be separated for subsequent precise recycling. For now, however, most of these steps are conducted manually or by human-robot co-working [12], which limits the recycling scale. Thus, innovative automatic pretreatment methods are needed for the industrialization application of spent LIB recycling.

In addition, some steps in the recycling process may involve the use of specific reagents, such as in hydrometallurgy, the use of acids and precipitants may introduce unwanted elements into the recovery products. Meanwhile, organic residues on the surface of spent LIB electrode materials, such as polyvinylidene fluoride (PVDF), are difficult to be removed completely. Some crude methods, such as calcination at high temperatures, can effectively remove these organic residues. However, this process destroys the original structure of the desired materials. Therefore, maintaining purity and avoiding new impurities are still important issues to consider. Some possible research directions are suggested as follows. First, acids with miscellaneous elements should be avoided when leaching elements, while acids containing only C, H, and O are recommended (e.g., citric acid). In terms of strategies not considering retaining the original structure of the raw materials for the desired products, the effect of high-temperature calcination pre-treatment is currently better than that of reagent pre-treatment in removing impurities. However, powerful decontamination reagents are still required, as the waste gas treatment of high-temperature calcination remains a problem, and the reagent reaction is relatively mild and easy to control. Finally, turning the impurities into treasures through clever design is also a research direction worth considering, which avoids the difficulty of removing such impurities but effectively utilizes these substances, thus eliminating the steps of subsequent treatment for them. For example, impurities like residual binder (e.g., PVDF) may serve as carbon sources to improve the electronic conductivity of the produced catalysts. Additionally, the accumulated transition metal elements on the graphite after excessive charging/discharging cycles may be exploited through appropriate designs to modify the product performance.

3 The Feasibility Analysis of Spent LIBs for Catalysts

Inappropriate disposal and recycling of the spent LIBs can lead to environmental degradation and severe resource waste as they contain heavy metals (e.g., cobalt, manganese, and nickel) and toxic organic electrolytes. Currently, the common idea is to regenerate LIB electrode materials from spent LIBs after disassembly and acid or high-temperature treatment for reutilization in producing new batteries [47, 55, 56]. However, traditional pyrometallurgy and hydrometallurgy methods consume extensive energy and inorganic/organic acids, making them expensive with complex follow-up procedures [57–59]. Meanwhile, regeneration does not apply to all commercial electrode materials due to their different market values. For instance, the economic benefits of recycling cheap $LiFePO_4$ for electrode material are low.

In addition to LIB remanufacturing, recycling and reusing spent LIBs for producing other advanced functional materials, such as catalysts, has attracted increasing attention. Spent LIBs with unique components (graphite anode and $LiCoO_2$, $LiMn_2O_4$, $LiFePO_4$, and $LiNi_xCo_yMn_zO_2$ cathodes) contain large amounts of valuable metals, such as lithium, iron, nickel, cobalt, manganese, copper, aluminum, and high value-added carbon materials. These residual components in the spent electrode materials could serve as core elements of functional catalysts, which has exhibited promising application aspects in catalysis. Appropriate recycling and reprocessing methods can be adopted to prepare corresponding metal compounds (e.g., oxides, sulfides, phosphides, and oxyhydroxides), multi-element alloys, carbon-based composites, and g-C_3N_4 materials, which are common catalysts in advanced oxidation, electrocatalysis, photocatalysis, and molecular catalysis [60–63].

The specific feasibility of converting spent LIB electrode materials into advanced catalysts for environmental remediation-related catalysis, battery-related catalysis, and substance conversion catalysis are as follows. (1) Water and air pollution, especially the organic pollutants harmful to human health, are serious issues faced by environmental remediation. Using the spent $LiFePO_4$ with rich Fe resources or other spent electrode materials to synthesize ferrite-type materials like Fenton or Fenton-like reagents for wastewater treatment has become a possible promising solution, which can catalyze the oxidation of organic compounds into small molecular substances. Replacing noble metal catalysts with transition metal-based catalysts for VOC oxidation is becoming a mainstream research direction in tackling air pollution. Using spent LIB electrode materials with rich transition metal resources to synthesize corresponding oxides (e.g., CoO_x and MnO_x) usually exhibits superior catalytic performance. (2) Substance conversion catalysis, including water splitting with oxygen evolution reaction/hydrogen evolution reaction, electrocatalytic carbon dioxide reduction, and biomass conversion, must overcome the high reaction energy barrier and thus consume large amounts of energy. Therefore, developing advanced catalysts is critical. Noble metal catalysts have shown excellent catalytic performance in substance conversion. However, the high prices and limited reserves of noble metals prevent their further large-scale application. Developing low-cost, non-precious metal-based catalysts to reduce the reaction energy barrier and enhance reaction efficiency is a prerequisite for further development in this field. Transition metal compounds have proven excellent catalytic properties comparable to noble metal catalysts by rational structural design and controlled synthesis process. Thus, developing advanced recycling technologies to transform spent LIB materials into efficient catalysts can effectively utilize a large number of LIB wastes and boost the development of advanced catalysts, demonstrating superior economic values and promising applications. (3) Different from those of traditional "rocking-chair" batteries, LSBs, metal-air batteries, and fuel cells are closely related to catalytic reactions, while carbon-based material and transition metal compounds are promising catalysts for these types of batteries. Spent LIB electrode materials with various transition metals and graphite closely related to the above catalyst materials of transition metal and carbon-based material compounds can be used to prepare battery-related catalysts by rational design and controllable synthesis. Meanwhile, as commercial cathode materials with unique crystal structures have potential catalytic activity, spent cathodes with volume changes, defect sites, and strains might promote electro-catalytic kinetics.

4 Environmental Remediation Related Catalysis

Environmental problems, especially water and air pollution, have become serious issues globally. Most organic pollutants (synthetics, plastics, pesticides, dyes, etc.) are difficult to be degraded naturally and can penetrate into crops through contaminated air, water, and soil, ultimately causing carcinogenic and teratogenic damage to humans and other organisms [64, 65]. Degrading the organic pollutants in sewage and air has aroused great attention. It is imperative to develop effective methods for removing these pollutants from the environment. Using appropriate catalysts to accelerate the degradation of these pollutants is considered as one of the most promising and practical way. In the meantime, spent LIB recycling is an important topic for environmental protection [66], while the abundant resources in spent LIBs, such as graphite and metal elements, are expected to serve as ideal catalyst raw materials. The "waste to wealth" concept has always been at the forefront of green chemistry research. Therefore, using spent LIBs as raw materials to prepare advanced catalysts for organic matter degradation in polluted water and air fulfills two purposes with one action. Existing studies have proved the feasibility and bright prospect of this approach, and the research progress is summarized and discussed, hoping to inspire follow-up research.

4.1 Application in Wastewater Treatment

Advanced oxidation is a classical chemical oxidation method, mainly including Fenton and Fenton-like methods, ultrasonic oxidation, ozone oxidation, photocatalytic oxidation, wet oxidation, wet catalytic oxidation, persulfate advanced oxidation, supercritical water oxidation, and other oxidation technologies, which can degrade organic compounds into small molecular substances through chemical reaction and further mineralize them into CO_2 and H_2O [67–69]. With high efficiency, strong operability, wide adaptability, and less secondary pollution, advanced oxidation is considered promising for wastewater treatment [70, 71]. In 1894, Fenton discovered that organics can be rapidly oxidized in a mixed solution of Fe^{2+} and H_2O_2 [72], i.e., the standard Fenton reagent, which has been widely studied and applied. The superior oxidizing property of this mixed system can be attributed to the presence of Fe^{2+}, which can promote the decomposition of H_2O_2 to produce HO· with strong oxidation capacity [73]. To further improve the organics removal effect based on the standard Fenton reagent, researchers developed various Fenton-like reagents with similar mechanisms by changing and coupling reaction conditions and improving reaction mechanisms, such as modified Fenton reagent, optical Fenton reagent, electric Fenton reagent, and ligand Fenton reagent [74].

Due to their intrinsic catalytic properties, some LIB materials could act directly as catalysts for specific reactions. In 2015, Goncalves et al. [75] proposed the degradation of methylene blue (MB) using spent LIB cathodes and the corresponding catalyst components containing $LiCoO_2$ (83% by weight), carbon (14.5% by weight) and others (Al, Al_2O_3 and Co_3O_4, 2.5% by weight) obtained by heat treatment to remove organic solvents. The activation energy (E_a) values) derived from the angular coefficient of the ln k vs. $1/T$ plot for MB degradation are 83 kJ mol^{-1} and 26 kJ mol^{-1} with pure H_2O_2 and coupled with spent LIB cathode (Fig. 3a), respectively, which is the main reason for the approximate 200 times faster speed in the absence of light when spent LIB cathode was used instead of pure H_2O_2 (Fig. 3b). Generating superoxide radicals ($O_2^{\cdot-}$) to promote the aromatic ring rupture and MB degradation is considered as the catalysis mechanism induced by the spent LIB cathode (Fig. 3c). This work promotes the application of end-of-life LIBs as catalysts for wastewater treatment. Guo and colleagues have realized the direct utilization of $LiFePO_4$ cathode materials from spent LIBs as photo-Fenton photocatalysts for the photodegradation of methyl orange (MO) with the help of low-molecular-weight organic acids (oxalic acid, $H_2C_2O_4$), constituting a unique approach for the application of low-value $LiFePO_4$ waste. In the UV-LFP-oxalate system with 20 mg L^{-1} LFP and 2 mmol L^{-1} oxalate, 50 mg L^{-1} MO was almost completely decomposed within 14 min, whereas only slight MO degradation was observed in the UV-LFP and UV-oxalate systems. The accelerated photodegradation of MO by oxalate can be attributed to the generation of the strong oxidizing agent ·OH and other oxidative free radicals through the catalysis of LFP, indicating that LFP could be an excellent photocatalyst for MO degradation by oxalate [76]. Another idea for reusing spent $LiFePO_4$ cathodes is proposed by Xu and colleagues [77], who synthesized iron hydroxyphosphate composites (FPOH) from waste $LiFePO_4$ through hydrothermal treatment. FPOH exhibited

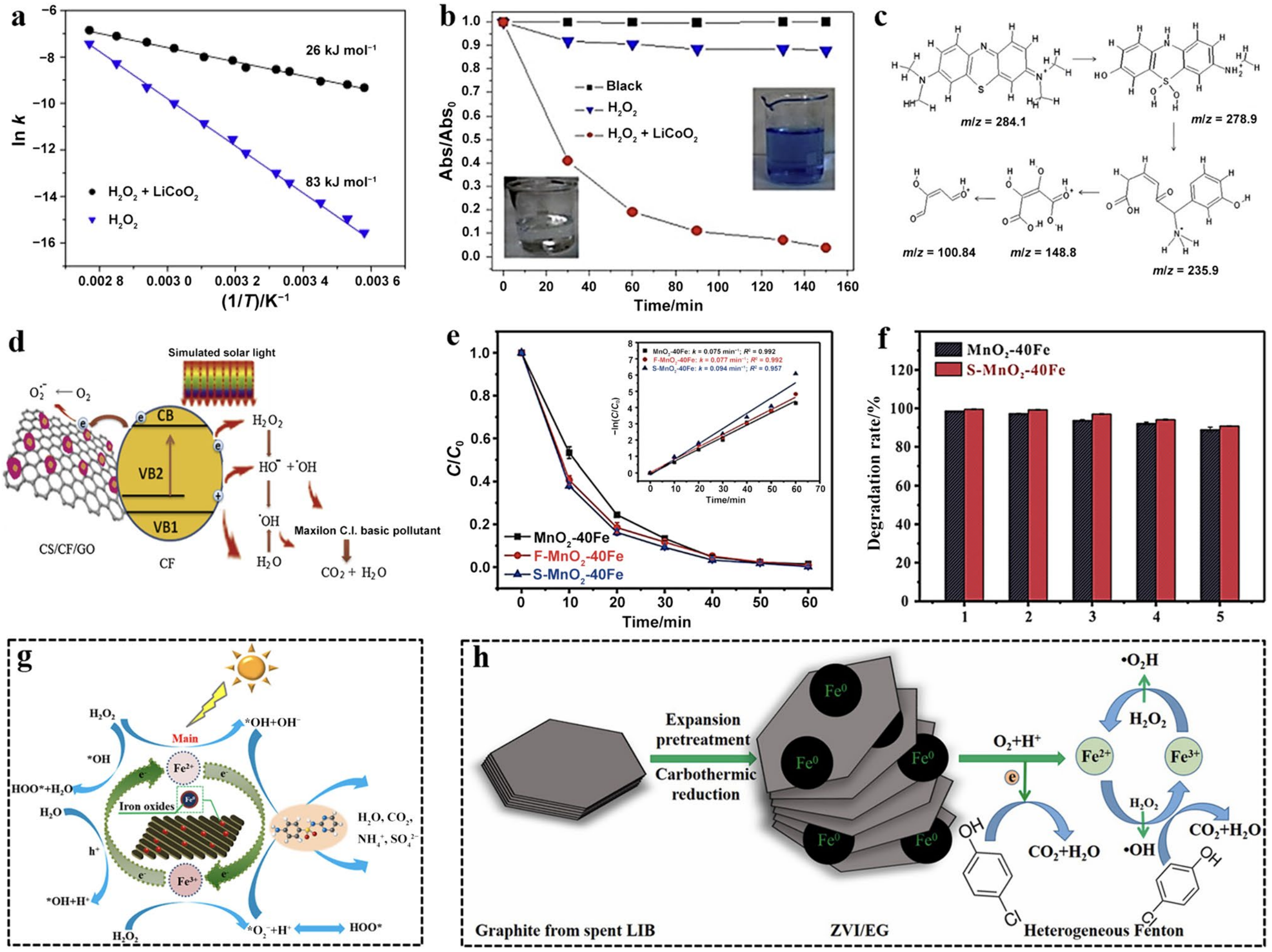

Fig. 3 **a** The ln *k* vs. 1/*T* for the degradation of 3 mg L^{-1} MB and 0.2 mol L^{-1} H_2O_2 with and without spent Li-BCT; **b** the visual aspect of MB degradation in the presence of spent Li-BCT; **c** the MB catalysis degradation mechanism. Reproduced with permission from Ref. [75]. Copyright © 2015, Elsevier Ltd. **d** Proposed mechanism of photodegradation at the solid-liquid interface with the solid state catalyst. Reproduced with permission from Ref. [80]. Copyright © 2016, Elsevier B.V. **e**, **f** First-order kinetics and degradation effect of MnO_2-40Fe, S-MnO_2-40Fe and F-MnO_2-40Fe; **g** multi-faceted photo-Fenton process mechanism of MnO_2-40Fe/H_2O_2/light system. Reproduced with permission from Ref. [83]. Copyright © 2021, Elsevier B.V. **h** Schematic of the zero-valent iron supported on expanded graphite and the mechanism of the degradation of 4-chlorophenol in water. Reproduced with permission from Ref. [86]. Copyright © 2021, Elsevier Ltd

exceptional Fenton-like catalytic performance in MB degradation while showing good adsorption performance for heavy metal lead. The catalytic ability of FPOH gradually decreased with the increase of organic dye concentration, where 10 μmol mol^{-1} MB could be completely removed in 24 h, and the MB removal rate in 20 μmol mol^{-1} reached 65% with 5 mL of H_2O_2 and 1 g L^{-1} of FPOH. In addition, FPOH can be reused with even better catalytic performance due to the more dispersed particles after recycling.

In addition to using spent LIB cathode materials for catalyzing pollutant degradation directly, the Li, Co, Ni, and Mn resources from spent LIB recycling can also serve as raw materials to synthesize ferrite, which has been used as catalysts in the heterogeneous photo-Fenton process to degrade wastewater pollutants [78, 79]. The oxidation reaction of $CoFe_2O_4$ with H_2O_2 produces radicals (·OH) at the solid-liquid interface with the solid-state catalysts (chitosan/$CoFe_2O_4$/graphene oxide: CS/CF/GO) for the degradation of Maxilon C.I. basic dye (Fig. 3d) [80]. Compared with homogeneous photo-Fenton reactions, the heterogeneous process can prevent sludge formation by iron hydroxide precipitation, expand the reaction pH range, and produce more hydroxyl radicals with strong chemical oxidation abilities. With these merits, Moura et al. [81] synthesized cobalt ferrite using the commercial reagents ($CoFe_2O_4$-R) and recycled cobalt from the spent LIB cathode materials ($CoFe_2O_4$-LIBs) via a co-precipitation method and applied them as the heterogeneous photo-Fenton catalysts to degrade MB. Structure analysis showed that $CoFe_2O_4$-LIBs and $CoFe_2O_4$-R had similar phase structures and microstructures. Moreover, they exhibited considerable discoloration efficiency of 87.7% for $CoFe_2O_4$-LIBs and 87.3% for $CoFe_2O_4$-R when used as catalysts for MB degradation, demonstrating the efficiency and capability of heterogeneous photo-Fenton catalysts obtained from spent LIBs for preventing environmental pollution and promoting recycling process sustainability. Another class of copper ferrite photocatalysts has also been synthesized ($CuFe_2O_4$) by Rocha et al. using copper recycled from spent LIBs for MB decolorization in a heterogeneous photo-Fenton process [82]. The $CuFe_2O_4$ exhibited a superior catalytic effect with a 96.1% decolorization efficiency after 45 min and could easily be separated from the water medium. In comparison, Fe_2O_3/Fe_3O_4 experiments were conducted under the same conditions, where the MB discoloration efficiency was only 77.5%. It should be noted that there is a considerable amount of Fe_2O_3 (45.7% of the total) in the synthesized $CuFe_2O_4$ from spent LIBs, and the combination of $CuFe_2O_4$ and Fe_2O_3 phases contributes to the photocatalysis because of their band gap energy. Loading Fe_2O_3 on $CuFe_2O_4$ enhanced its photocatalytic activity in the visible light range.

Besides ferrite-type materials, Chen et al. [83] prepared MnO_2/Fe(0) composite catalysts via a hydrothermal process by reclaiming Mn from the spent LIB cathode ($LiMn_2O_4$). They synthesized MnO_2/Fe(0) with various MnO_2:Fe(0) molar ratios using commercial reagents first, and the optimal catalytic activity for sulfadiazine degradation was observed under the ratio of 40:1 (MnO_2-40Fe). Then, the spent LIB cathode and fresh LIB cathode were used to prepare the MnO_2/Fe(0) under the optimal conditions, denoted as S-MnO_2-40Fe and F-MnO_2-40Fe, respectively. Further analysis and comparison found that among MnO_2-40Fe, S-MnO_2-40Fe, and F-MnO_2-40Fe, the S-MnO_2-40Fe delivers the highest sulfadiazine degradation efficiency (99.6%) with a rate constant of 0.094 min^{-1} (Fig. 3e, f). The enhanced catalytic activity of S-MnO_2-40Fe is ascribed to oxidation state changes, phase transition, and charge density of MnO_2 during inter-phasing Li ions with MnO_2-40Fe at the discharge stage. Compared to MnO_2-40Fe, the S-MnO_2-40Fe showed enhanced reactivity and stability for sulfadiazine degradation after being repeatedly used five times, which can be attributed to the retard of MnO_2 dissolution because of the binder used in spent cathode materials. As shown in Fig. 3g, a multi-faceted photo-Fenton process mechanism is proposed based on mass spectroscopic data with the final products of CO_2, H_2O, NH^{4+}, and $SO_4{}^{2-}$.

Compared with valuable spent cathode materials, the cheaper graphite anode gets less attention, and only a few works have reported graphite recovery from spent LIBs, such as attaching to the adsorbent, graphene synthesis, electrode materials, film materials, and sensors [84, 85]. Considering the high purity, good layer structure, and electrical conductivity of graphite in spent LIB anodes, recycling it for synthesizing advanced carbon materials is a promising option. Chen et al. [86] prepared zero-valent iron/carbon composites from wastes [Fe_2O_3 from mill scale and spent graphite (SG) from LIBs] and commercial Fe_2O_3 and graphite, respectively, through carbothermic reaction (denoted as ZVI/C-X and ZVI/C) and used them as catalysts for ibuprofen degradation by Fenton reaction from water. The results showed that ZVI/C-X and ZVI/C exhibited considerable catalytic performance, and ZVI/C-X exhibited low Fe-lixiviation and high stability, maintaining activity even after four consecutive cycles of use at a very low concentration of iron ions in solution after the reaction. ZVI/C-X particles have larger sizes than ZVI/C particles with the same ZVI content, which may facilitate their separation from the reaction media if needed. This work provides a new possibility for recovering graphite from spent LIBs to serve catalysis applications. This strategy was also adopted recently by Guan et al. [86] to prepare catalysts for the 4-chlorophenol (4-CP) removal from water by heterogeneous Fenton reactions, where the ferric chloride and graphite from spent LIBs were used as precursors to synthesize zero-valent iron supported with expanded graphite (ZVI/EG) (Fig. 3h). ZVI/EG delivers comparable activity to others previously reported while showing promising prospects, with the removal rate of ZVI/EG reaching 0.052 7 min^{-1}, higher than the 0.000 08 min^{-1} with Fe^{2+} ions and H_2O_2 reported by Jin et al., and the 0.016 min^{-1} with mesoporous carbon activated peroxydisulfate reported by Yang and colleagues [87, 88]. Moreover, the 4-CP removal percentage remained high even in the sixth cycle, demonstrating its considerable stability. However, these two efforts of recycling SG for catalysts did not utilize the structural or compositional advantages of SG over commercial graphite. The performance of catalysts prepared with recycled graphite resembles those synthesized directly from commercial raw materials. Examples in subsequent sections show the unique uses of SG for catalysis.

Advanced oxidization based on peroxymonosulfate (PMS) is also extensively utilized to remove organic pollutants from wastewater because of its high potential and mild pH requirement. Transition metal oxides (e.g., Fe, Co, Cu, and Mn-based materials) are recognized as appropriate catalysts to activate PMS and generate free radicals for contaminant degradation, which generate sulfate radical anion (SO_4^{2-}) with high redox potential (2.5–3.1 V) to degrade most contaminants in water into small molecules [89]. The spent LIB cathode materials cover a wide variety of transition metal species mentioned above, which can be used as low-cost raw materials to synthesize catalysts for PMS activation, thus arousing great interest. Based on this principle, Pi et al. [90] synthesized the $CoFeO_2$@carbon nitride

($CoFeO_2$@CN) composite catalyst using Co from spent LIBs. The $CoFeO_2$@CN composite exhibited good catalytic activity and recyclability for levofloxacin (LVF) degradation by the activation of PMS, where about 90% of the LVF was degraded in the optimal system, and the LVF degradation efficiency remained above 70% following five cycles. Thus, they have confirmed that SO_4^{2-} served the dominant active species in the catalyst system, capable of extending to the removal of refractory compounds. Zhao et al. [91] developed a facile calcination method at various temperatures by preparing a catalyst using cathode materials recovered from spent LIBs to activate PMS for degrading antibiotics in wastewater. The X-ray diffraction (XRD) patterns in Fig. 4a prove that the obtained series catalysts (CM-X) comprise 94% $LiCoO_2$ and 6% Co_3O_4, respectively. When used to degrade levofloxacin hydrochloride (LFX), CM-850 exhibited the optimal catalytic property with a removing efficiency of 94% under 400 mg L^{-1} catalyst dosage, 0.5 mmol L^{-1} PMS concentration, and an initial pH of 4.65. The mechanism exploration verified that the varied valence of Co^{2+}/Co^{3+} underpinned PMS activation, while SO_4^{2-} and non-radical $^1O_2$ were of critical significance in LFX degradation (Fig. 4b). Moreover, they also used the as-prepared catalysts to degrade five other types of antibiotic pollutants in wastewater, including terramycin, norfloxacin, ciprofloxacin, aureomycin, and tetracycline hydrochloride, yielding certain degradation effects (Fig. 4c). This work brings inspiration to the high value-added application of spent LIB recycling

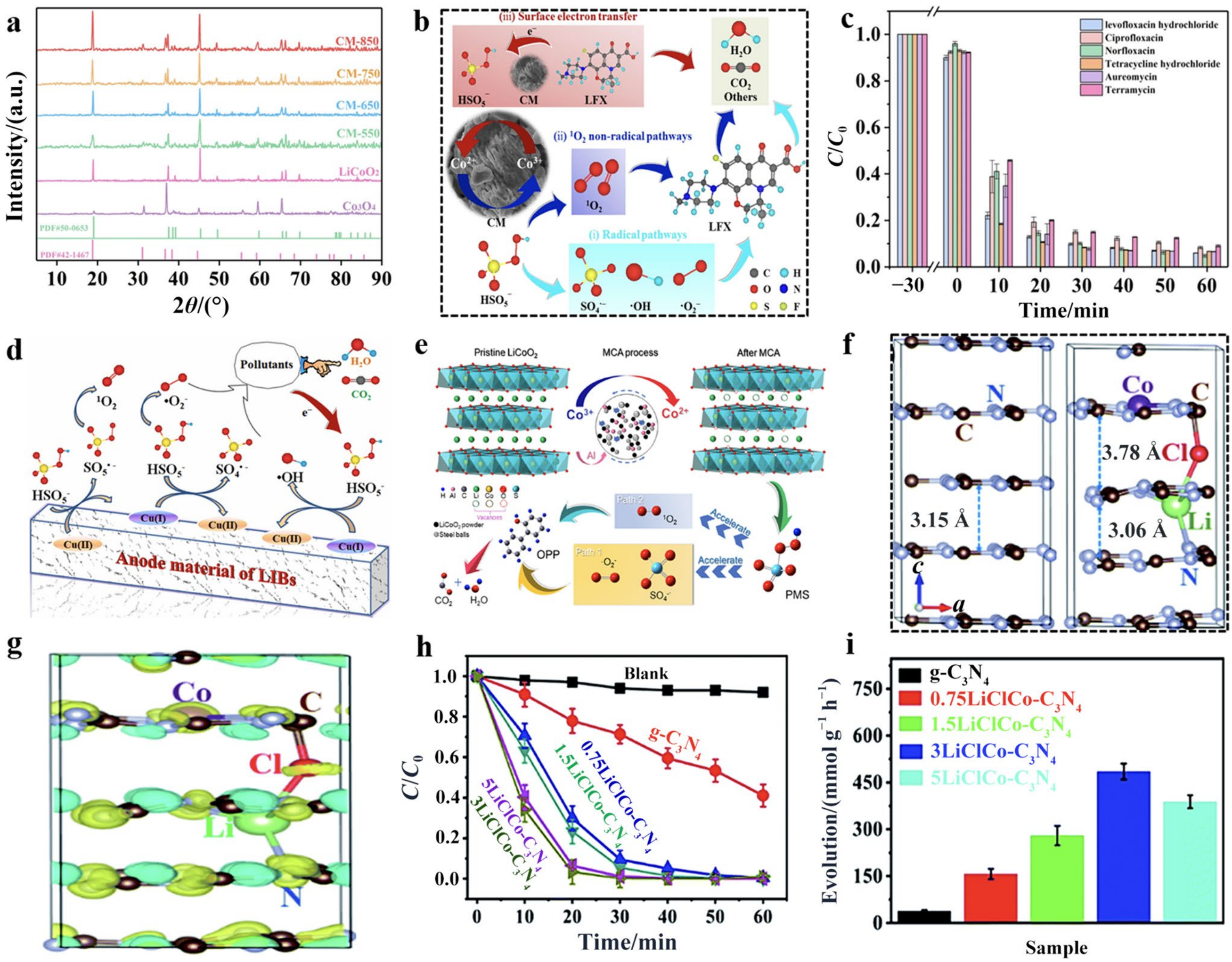

Fig. 4 **a** XRD patterns of pure Co_3O_4 and $LiCoO_2$, and CM catalysts; **b** the possible mechanism of PMS activation and LFX degradation in the CM/PMS system; **c** adaptability investigation of CM/PMS system on antibiotic pollutants degradation. Reproduced with permission from Ref. [91]. Copyright © 2020, Elsevier B.V. **d** The proposed reaction mechanism of PMS activation and RhB degradation in AM/PMS system. Reproduced with permission from Ref. [92]. Copyright © 2021, Elsevier B.V. **e** Schematic of MCA and degradation of OPP. Reproduced with permission from Ref. [93]. Copyright © 2021, American Chemical Society. **f**, **g** The optimal g-C_3N_4 and obtained stable LiClCo-C_3N_4 structure with charge difference distribution; **h**, **i** photocatalyst for RhB degradation and hydrogen evolution of 3LiClCo-C_3N_4, respectively. Reproduced with permission from Ref. [99]. Copyright © 2021, The Royal Society of Chemistry

materials. Except for preparing catalysts from spent cathode materials, Zhao and co-workers also synthesized a copper oxide and graphite carbon (CuO/C) catalyst from recycled spent LIB anode materials to activate PMS for degrading a wide range of organic contaminants (Fig. 4d), which exhibited excellent catalytic effects. In particular, the rhodamine B (RhB) degradation rate reached 100% in 13 min under the catalysis of CuO/C in the solution through the PMS activation. The admirable oxidation and degradation abilities of this catalyst/PMS system are generally attributed to the produced $SO_4^{\cdot-}$ and $^1O_2$, and the catalytic performance of this system was determined by the rate of Cu(II) reduction to Cu(I), where the SG base accelerated the redox cycle of Cu(II)/Cu(I), promptly activating PMS to produce reactive oxygen species (e.g., $HO^{\cdot}$, $SO_4^{\cdot-}$, and $^1O_2$) for degrading pollutants [92].

Although using spent LIB electrode materials as catalysts for advanced oxidation has attracted increasing attention, the catalytic efficiency and utilization rate remain limited. To achieve efficient utilization of the spent cathode and avoid wasting aluminum resources, Dang et al. [93] used aluminum foils in the cathode of spent $LiCoO_2$ batteries as reducing agents to convert part of Co^{3+} into Co^{2+} during the mechanochemical activation (MCA) process (Fig. 4e). The obtained Co^{2+} could serve as the active center to activate PMS efficiently and improve the catalytic effect. Furthermore, the degradation rate of *o*-phenylphenol could reach 100% within 30 min. To further simplify the catalyst manufacturing process and save costs, Wang et al. [94] investigated the effect of the lithiation extension on the catalytic activity, concluding that the catalytic activity increased as the lithiation depth extended. Based on this, they directly used the recycled active $LiMn_2O_4$ component from spent LIBs for PMS activation for decomposing RhB, where approximately 98.2% of RhB was degraded within 30 min, and the catalytic decomposition efficiency still reached 94.8% after 10 cycles.

Photocatalytic oxidation is another advanced oxidation method in wastewater treatment for recalcitrant and toxic organic compound degradation [95, 96], where advanced catalysts play a key role in improving energy utilization efficiency and promoting the thorough degradation of organic pollutants. The photocatalysts with heterostructures exhibit improved catalytic ability due to the regulated electronic structure and efficient photoinduced electron-hole pair separation process [97]. Spent LIB electrode materials with complex compositions can be used to prepare the hetero-structured photocatalysts. Many Co-based oxides have been synthesized from spent LIB cathodes to serve as efficient photocatalysts for decomposing textile dye and antibiotics in wastewater [91, 98]. Niu et al. [99] regulated the π-conjugated structure of g-C_3N_4 using the spent $LiCoO_2$ cathode, NH_4Cl, and melamine. The as-prepared LiClCo-C_3N_4 catalyst exhibited a unique crystal structure (Fig. 4f) composed of Co–N, Cl–C, Li–Cl, and Li–N bonds, which could boost the charge migration and separation between g-C_3N_4 layers (Fig. 4g). In order to achieve the optimal catalytic effect, they regulated the usage amount of spent $LiCoO_2$ (denoted as 5LiClCo-C_3N_4, 3LiClCo-C_3N_4, 1.5LiClCo-C_3N_4, and 0.75LiClCo-C_3N_4). When used as a photocatalyst for RhB degradation and hydrogen evolution, the as-prepared 3LiClCo-C_3N_4 showed superior catalytic performance with 12.6 and 15.3 times higher than those of pristine g-C_3N_4 (Fig. 4h, i). Furthermore, Zhang et al. [100] synthesized a graphene oxide-copper composite material from the spent anode graphite and copper foils in the LIB anode and studied their photocatalytic degradation performance on MB. Within the graphene oxide-copper composite, the graphene oxide acted as a carrier, which improved the catalytic activity and greatly increased the degradation rate of CuO on MB due to the large specific surface area and oxygen groups. Moreover, adding an electric field can significantly improve the catalytic degradation efficiency because more active free radicals can be produced. Furthermore, the graphite from the spent LIB anode can also be used to synthesize a series of graphene-based materials for catalytic ozonation of organic pollutants [101]. These works revealed a synergetic concept of recycling wastes as high-efficiency photocatalysts for environmental protection.

In general, end-of-life LIB electrode materials, including various commercial cathode and graphite anode, can be used directly or indirectly for advanced catalysts through calcination or hydrothermal regenerative processes and applied to pollutant degradation in wastewater through different catalytic reaction technologies. This unique strategy solves the huge challenges of spent LIB recycling and improves the water environment quality, thus warranting more attention in future research. Moreover, catalysts derived from spent LIBs are limited to metal oxides at present, and many other types of catalysts, such as alloys, single-atoms, and transition metal chalcogenides, can be developed from spent LIBs, leaving huge space for development and application.

4.2 Application in Volatile Organic Compounds Removal

VOCs are mainly derived from the incomplete combustion of fossil fuels and the escape of industrial products during manufacturing and application [102]. They are considered culprits of air pollution and the main harmful substances causing great damage to human health [103, 104]. Many techniques have been developed to control VOC emissions, such as adsorption, thermal combustion, photocatalytic degradation, plasma degradation, and catalytic oxidation [105]. Among them, catalytic oxidation is the most promising due to the harmless end products, high activity, and low price

[106]. Previous studies have shown that noble metals exhibit superior catalytic performance for VOC oxidation, but their widespread application is hindered by the high cost [107]. In contrast, transition metal compounds have become common high-performance catalysts for VOC removal due to their low cost and excellent low-temperature catalytic performance [108]. Therefore, it is feasible to utilize spent LIB cathodic material-derived transition metals to synthesis catalysts, which shows a promising application prospect.

As proof of concept, Guo et al. [109–112] synthesized a series of manganese-based metal oxide catalysts from spent LIB cathode materials using the impregnation, hydrothermal, co-precipitation, and combustion methods to catalyze VOC oxidation (Fig. 5a, b), which showed excellent catalytic activity due to the high-valence metal ion, high concentration of active oxygen species, and large specific surface area. Potassium permanganate and ozone have been used to thoroughly oxidize spent LIBs for selective and efficient manganese recovery, and transition metal-doped MnO_2 has been prepared for the catalytic oxidation of VOCs. The manganese recovery rate could reach about 100%, and the recovery rates of cobalt, nickel, and lithium were below 15%, 2%, and 1%, respectively. Introducing nickel, cobalt, and copper into manganese oxides significantly affected the catalytic performance compared to the substitute of lithium and aluminum. Consequently, Mn-based multi-oxide catalysts from spent LIBs exhibited better catalytic performance in toluene and formaldehyde removal than pure manganese oxides owing to their lower crystallinity, more defects, larger specific surface area, more oxygen vacancies, and better low-temperature redox abilities [109]. They further synthesized a series of manganese oxide catalysts modified by other elements (e.g., Fe, Bi, and Ce) and demonstrated that introducing appropriate amounts of these elements can enhance the catalytic performance of manganese oxides in VOC removal, which can be attributed to the lower crystallinity, larger specific surface area, small particle size, higher amount of active oxygen species and Mn^{4+} ions, and better low-temperature reducibility [111]. Besides MnO_x, Mn-based composite perovskite was also considered an effective candidate for VOC removal. Since ternary LIBs contain abundant metals such as manganese, cobalt, and nickel, spent ternary LIBs are considered promising sources of metal precursors for the preparation of Mn-based composite perovskite [112].

In addition to Mn-based oxides, cobalt-based oxides derived from spent LIBs also attracted attention in catalytic VOC oxidation [113]. Dai et al.[114] used an acid treatment method to improve the catalytic activity of spent $LiCoO_2$ for benzene oxidation (Fig. 5c). The as-prepared acid-modified $LiCoO_2$ catalyst showed O, Co, and Li vacancies, facilitating gaseous benzene adsorption and subsequent activation. The part of Co^{3+} leached out from the $[CoO_6]$ octahedra in $LiCoO_2$ by HNO_3 solution via the disproportion reaction with the deintercalation of interlayer Li^+ ions is the reason

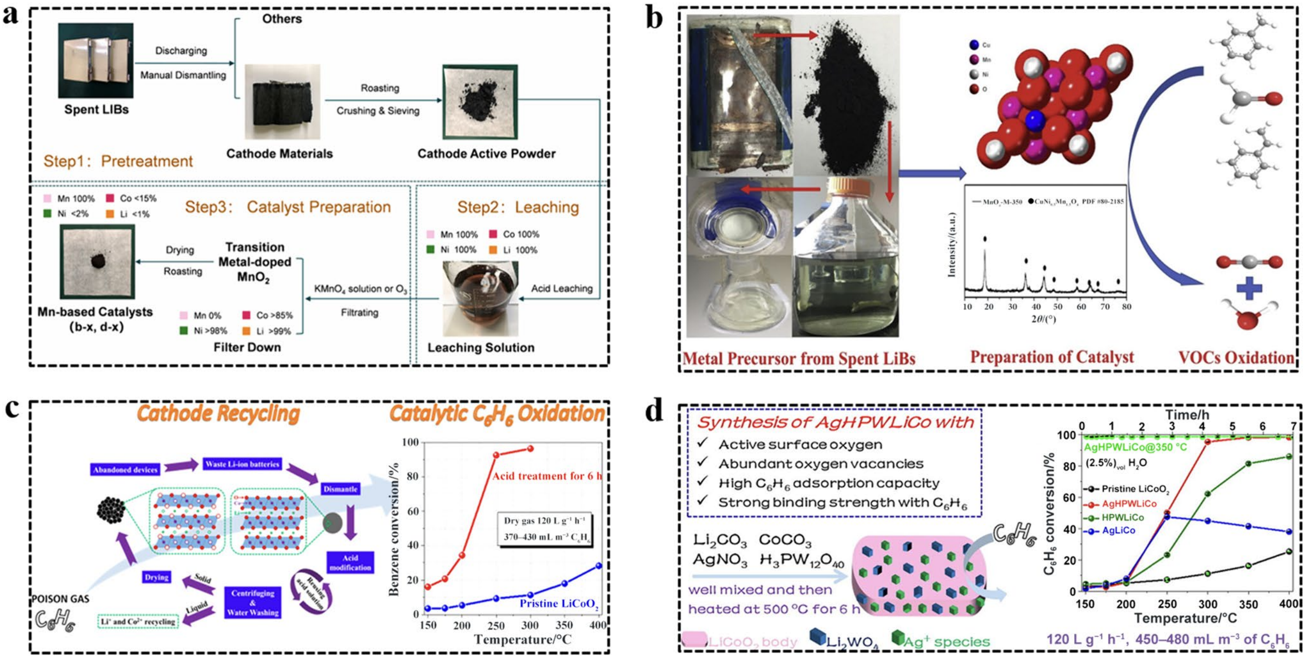

Fig. 5 **a**, **b** The research scheme of catalyst preparation from spent LIBs. Reproduced with permission from Refs. [109, 110]. Copyright © 2019 and 2020, Elsevier B.V. **c** Process of the LCO cathode recycling and the catalytic effect on the C_6H_6 oxidation. Reproduced with permission from Ref. [114]. Copyright © 2019, American Chemical Society. **d** Synthetic method of AgHPWLiCo and the catalytic effect on the C_6H_6 oxidation. Reproduced with permission from Ref. [115]. Copyright © 2019, Elsevier B.V

for vacancy formation. Liu et al. [115] (Fig. 5d) synthesized an $LiCoO_2$ catalyst modified by Ag and phosphotungstic acid with considerable performance on the catalytic oxidation of benzene vapor, indicating the potential of using cathode materials like $LiCoO_2$ as the raw materials to synthesize catalysts. Further, they proposed a simple and facile water treatment procedure to activate the recycled $LiCoO_2$ cathode materials from spent LIBs for promoting gaseous benzene oxidation. Water washing eliminated the chemically absorbed oxygen and surficial lattice oxygen, and the bulk lattice oxygen was thereby exposed and became more reachable to the gaseous benzene, which could oxidize benzene efficiently at high temperatures [116]. These strategies provided new concepts for deriving catalysts from spent LIB electrode materials for VOC degradation. However, these concepts are still in their infancy, with many opportunities and challenges.

In conclusion, the latest research progress in using spent LIBs to prepare advanced catalysts for environmental remediation (wastewater treatment and VOC removal) is summarized and discussed from the aspects of regeneration methods, catalytic performance, and catalytic reaction mechanism. Hopefully, this summary can be of reference significance to the subsequent research. Furthermore, the recycling of spent LIB materials for environmental remediation catalysts and the corresponding performance are shown in Table 1.

These examples show the feasibility and prospect of converting spent LIB materials into catalysts, which alleviates the environmental hazards caused by spent LIBs while realizing their high-value utilization. However, this approach is not mature, and it faces many challenges. Firstly, most studies on recycling spent LIB materials as catalysts are based on hydrometallurgy or pyrometallurgy, that is, extracting elements from spent LIB materials and synthesizing catalysts using their simple substances or compounds as raw materials, which does not have much competitive advantages over directly using commercially available raw materials. Typically, there is no performance difference between catalysts synthesized from recycled or non-recycled raw materials, and the advantages of using spent LIB materials in catalysis have not been fully exploited. Meanwhile, previous research mentioned that the crystal structure and defects may benefit the catalytic effect. More studies on the catalytic properties of spent LIB materials are needed. Furthermore, the application of the catalysts in these reports is relatively narrow, i.e., most reports studied the catalytic effects on particular pollutants, while there are various pollutants in actual sewage and exhaust gas, necessitating multifunctional catalysts. Therefore, researchers are recommended to develop and study the catalytic effect of catalysts on the degradation of various pollutants in follow-up studies. In addition, a comparison between catalysts synthesized from recycled and non-recycled materials is necessary, whereas the comparison between the newly developed and commercial catalysts also provides useful information. Hence, researchers are suggested to always add a commercial catalyst as a comparison when quantifying catalyst performance so that readers can better understand the performance of newly developed catalysts.

5 Spent LIB-Derived Catalysts for Substance Conversion

5.1 Oxygen/Hydrogen Evolution Reaction (OER/HER) Related Catalysis

Electrolytic water decomposition is the most promising method for hydrogen production as the hydrogen generated from electrolysis represents a non-CO_2 emission process and is often referred to as green hydrogen, which involves OER and HER [117, 118], respectively. Water electrolysis to produce hydrogen usually requires efficient catalysts. However, the application of noble-metal catalysts (e.g., platinum, ruthenium, and iridium) is limited due to their scarce storage and high cost [119]. Developing non-precious metal-based catalysts with low cost to reduce the reaction energy barrier and enhance the reaction efficiency is a prerequisite for further development in this field. Transition metal compounds have been proven to have excellent catalytic properties comparable to noble metal catalysts by rational structural design and controlled synthesis process. Therefore, the most common LIB cathode materials have potential catalytic activity for OER/HER due to the presence of transition metals such as cobalt, nickel, manganese, and iron, which can also be used as precursors to prepare various transition metal compounds to substitute noble-metals as electrocatalysts, which has aroused academic attention [120, 121]. According to the different types of cathode materials, this section summarized the latest progress of electrocatalyst preparation using $LiCoO_2$, $LiFePO_4$, and ternary $LiNi_{1-x-y}Co_xMn_yO_2$ (NCM) as raw materials, respectively, covering synthetic method, catalytic efficiency, and mechanism.

5.1.1 Spent Co-Based LIBs as Catalyst Sources

Among the various transition metals, cobalt-based materials exhibit better electrocatalytic performance than other non-noble metal materials [122]. $LiCoO_2$, as an important class of cobalt-based oxides, has been widely used in LIB cathode materials due to the high specific capacity, long cycle life, and high potential, which usually has two main types of spinels and layered crystal structures. As the cathode active material, LCO with layered structures allows smooth intercalation and extraction of lithium ions during

Table 1 Recycling of spent LIB materials for environmental remediation catalysts and corresponding performance

Spent material types	Regenerative methods	Catalyst	Application	Catalytic reaction mechanism	Performance	References
$LiCoO_2$	Heated to 200 °C for 5 h	Li-BCT (83% $LiCoO_2$, 14.5% C, 2.5% Al, Al_2O_3 and Co_3O_4)	Degradation of methylene blue	The generation of superoxide radicals	Reduced activation energy (26 kJ mol^{-1}) and increased velocity constant (203×10^{-4} min^{-1})	[75]
$LiFePO_4$	Direct utilization	LFP ($LiFePO_4$)	Degradation of methyl orange	The increased production of $^{\cdot}OH$ radicals and other active free radicals	Removal reaches 50 mg L^{-1} at pH = 4	[76]
	Hydrothermal treatment	FPOH (iron hydroxyphosphate)	Degradation of methylene blue	Fenton-like catalysis	Removal reached 65% in 20 μmol mol^{-1}	[77]
$LiCoO_2$	Co-precipitation method	$CoFe_2O_4$-LIBs ($CoFe_2O_4$ and Fe_2O_3)	Degradation of methylene blue	Heterogeneous photo-Fenton catalysis	Discoloration efficiency reached 87.7% at pH = 3	[81]
Current collector	Co-precipitation method	$CuFe_2O_4$-LiB ($CuFe_2O_4$ and Fe_2O_3)	Decolorization of methylene blue	Heterogeneous photo-Fenton catalysis	Decolorization efficiency reached 96.1% in 45 min	[82]
$LiMn_2O_4$	Hydrothermall process	MnO_2/Fe(0)	Decolorization of sulfadiazine	Multi-faceted photo-Fenton catalysis	Degradation efficiency reached 99.6% with 0.094 min^{-1}	[83]
Graphite	Carbothermic reactions	ZVI/C-X (Fe and C)	Removal of ibuprofen	Fenton catalysis	Removal reached 76.7%	[86]
Graphite	Carbothermic reduction	ZVI/EG (Fe and C)	Removal of 4-chlorophenol	Heterogeneous Fenton catalysis	Removal reached 97% in 1 h	[86]
$LiCoO_2$	Hydrothermal and carbonization	$CoFeO_2$@CN	Degradation of levofloxacin hydrochloride	PMS-based advanced oxidation	Removal > 90%	[90]
$LiCoO_2$	Calcination method	CM catalyst (94% $LiCoO_2$ and 6% Co_3O_4)	Eliminate antibiotics	PMS-based advanced oxidation	Removal reached 94% for levofloxacin hydrochloride	[91]
Graphite	Calcination process	CuO/C	Degradation of organic contaminants	PMS-based advanced oxidation	Removal reached 100% for rhodamine B	[92]
$LiCoO_2$	Mechanochemicalactivation	$LiCo_xO_y$ (Co: 2+)	Degradation of *o*-phenylphenol	PMS-based advanced oxidation	The degradation rate reached 100% within 30 min	[93]
$LiMn_2O_4$	Washing-recycling process	$LiMn_2O_4$	Degradation of rthophenylphenol	PMS-based advanced oxidation	Decomposition efficiency reached 94.8% after 10 cycles	[94]
$LiCoO_2$	One-pot sintering	LiClCo-C_3N_4	Degradation of rhodamine B	The generation of H^+, $^{\cdot}O_2^-$, and $^{\cdot}OH$	Degradation rates reached 15.3 times higher than pristine g-C_3N_4	[99]
$LiNi_xCo_yMn_zO_2$	Acid leaching and calcination	MnO_x-M-350 ($CuNi_{0.5}Mn_{1.5}O_4$)	VOCs oxidation	Mars-van-Krevelen mechanism	Over 90% conversion rates for toluene, ethylbenzene, and formaldehyde	[110]
$LiCoO_2$	Disproportion reaction	H-LiCo-6	Catalytic C_6H_6 oxidation	Mars-van-Krevelen mechanism	Benzene conversion > 90% at 350 °C	[114]
$LiCoO_2$	Thermal treatment	AgHPWLiCo	Catalytic C_6H_6 oxidation	Mars-van-Krevelen mechanism	Benzene conversion ~ 95% at 300 °C	[115]

charging and discharging. However, after excessive cycles of charge and discharge, the layered structure degrades to a spinel structure, leading to a decline in reversible capacity and representing a manifestation of material failure. Yet, the opposite is true in catalysis, where the spinel structure exhibits higher catalytic activity than others and deliver superior performance in OER and HER [123]. Therefore, spent LCO is considered a promising advanced electrocatalyst and has received more attention recently. Moreover, it is generally believed that the Co_3O_4 formed after delithiation is the actual active substance, and the high OER activity is also ascribed to the oxidation of cobalt ions and phase transition during the Li^+ extraction process [124]. After repeated charging/discharging cycles, the lattice volume undergoes obvious changes and generates more defects and strains of LCO, further promoting the electrochemical catalytic kinetics [125].

Specially, Chen et al. [126] reported a simple method to use the spent LCO cathode as OER catalysts. They discovered that with an increased number of cycles, a new phase of $Li_{1-x}CoO_2$ formed, and the LCO surface was corroded during the Li^+ insertion and extraction processes, contributing to a larger specific surface area of the catalysts. In addition, the OER catalytic activity of recycled LCO increased with the number of cell cycles. Recycled LCO after 500 cycles (2.8 V to 4.3 V at 0.5 C) showed improved OER catalytic activity with a reduced onset potential of 1.52 V and accelerated the current increase compared with the original LCO. It delivered a current density of 9.68 mA cm^{-2} at 1.65 V, approximately 3.8 times that of the pristine LCO. Meanwhile, it maintained the original catalytic activity after repeating the potential cycling test for 1 000 cycles. This study provides a simple and effective method to prepare catalysts from spent LIBs. High-temperature (HT)-LCO is also an important cathode material for LIBs with a hexagonal and layered structure [127]. Pegoretti et al. [128] extracted Co from the spent LCO cathode and prepared $Co(OH)_2$ as the precursor. After mixing with stoichiometric Li_2CO_3 and calcinated at 800 °C for 5 h, HT-LCO was obtained. The synthesized HT-LCO has considerable potential as a cathode material and exhibited promising OER catalytic effects. The as-prepared HT-LCO as an OER catalyst delivered a lower onset potential of 0.35 V than Ni and Pt plates (over 0.5 V). According to the Tafel plot and Nyquist diagrams, the activation energy of HT-LCO for OER was 28.0 kJ mol^{-1} with a transfer charge resistance of 1.55 Ω, which was regarded as excellent compared to other Co-based materials. These data indicate that HT-LCO is expected to be an excellent OER catalyst. However, the proposed recycling strategy ignored the recycling of lithium and has a large energy consumption, which still requires improvements. Huang et al. [129] developed an environmental-friendly and energy-efficient strategy to exfoliate strong Li–O interplanar bonding into ultra-thin CoOOH nanosheets from the spent LCO cathode with synergistic action of phase conversion and lattice expansion (Fig. 6a). Due to the sheet morphology, larger spacing, and exposed surface, the as-prepared ultra-thin CoOOH catalyst provided more electrocatalytic active sites for OER, delivering superior performance with a low Tafel slope of 53.8 mV dec^{-1} and a small overpotential of 305 mV at 10 mA cm^{-2} after long-term electrolysis for 65 h, superior to some advanced OER catalysts (e.g., fresh $LiCoO_2$ and commercial RuO_2) (Fig. 6b). In addition to LCO, a wide variety of layered cathode materials can be successfully converted into ultrathin nanosheets with this strategy, which can be further applied to OER catalysis. Besides, the Li in spent LCO was recycled as Li_3PO_4 for re-utilization. Further research revealed that the CoOOH catalyst with a modest content of oxygen vacancies and thinner nanosheets requires a lower Gibbs free energy difference during catalysis (Fig. 6c). Natarajan et al. [120] proposed a facile way to synthesize spinel microsphere $MnCo_2O_4$ catalysts from Co-rich (like LCO) and Mn-rich (like Li_xMnO_{x+1}) spent LIBs without using surfactant. The $MnCo_2O_4$ catalyst exhibited outstanding OER activity in an alkaline medium and maintained its catalytic ability after reusing in water electrolytes. Furthermore, it exhibited comparable activity to commercially available OER catalysts, such as RuO_2, Co_3O_4, and MnO_2. Although not requiring surfactant, some chemical reactants in this study, like acetic acid, were still required, and the calcination temperature of 650 °C consumed more energy, also regarded as an issue faced by traditional hydrometallurgy. In addition to using high-temperature sintering to synthesize catalysts from spent LIBs, other more straightforward strategies must be developed to reduce energy consumption. Caspar et al. [130] prepared an efficient Co-based catalyst (cobalt-aluminum borate) from spent cobalt-based LIBs to hydrolyze $NaBH_4$ for hydrogen production. This strategy does not require calcination, and the synthesis process could be achieved at relatively low temperatures. The average oxidation state of the prepared Co-based catalyst was 2.45, making it an inert catalyst at room temperature. However, when heated to 70 °C, the catalytic performance was improved, and the rapid hydrogen production could reach 49.3 L min^{-1} g^{-1} (Co), with each Co atom generating at least 1.2×10^7 hydrogen molecules. However, improving the long-term stability of this type of catalyst still needs further research, and using strong acid is also one of the most important issues hindering the practical application of such methods. Recently, hydrometallurgical methods tend to use less environment-polluting and more easily degradable organic acids as leaching agents. Arif et al. [131] utilized citric and lemon peel extracts to recover lithium cobalt oxide from waste LIBs, and the recovered cathode material was used as an OER catalyst. Liu et al. [132] used mild organic acids (glycine & ascorbic acid) to recover Co from spent

LIBs and synthesize cobalt nitride composite material (CoN-Gr-2) with the shape of 3D sea-urchin through hydrothermal method. The CoN-Gr-2 exhibited excellent performance as a bi-functional catalyst for OER and HER. When adopting CoN-Gr-2 as both anode and cathode materials for overall water splitting (in 1.0 mol L^{-1} KOH electrolyte), the assembled cell achieved a current density of 10 mA cm^{-2} at 1.61 V, close to that of Pt/C||RuO_2 benchmark (1.60 V). These methods help to further solve environmental pollution and generate good economic benefits. However, high temperature calcination with high energy consumption was inevitable in the pretreatment or subsequent resynthesis.

5.1.2 Spent LFP as Catalyst Sources

Pure LFP cathode materials possess poor active sites, weak intrinsic activity, and low conductivity, contributing to their inferior catalytic activity, which renders them unavailable for direct application. For this reason, the recovery of spent LFP-based LIBs as electrocatalysts is limited. Cui et al. [133] proposed a wetness impregnation method featuring facile and economic attributes to convert spent LFP into NiFe oxy/hydroxide catalyst with high catalytic activity for OER. Specifically, this catalyst was recycled from spent LFP cathode material, which was scraped in a $Ni(NO_3)_2 \cdot 6H_2O$ solution and evolved into defect-rich $NiFeO_xH_y$ nanosheets during OER (Fig. 6d). The evolved $NiFeO_xH_y$ showed better catalytic activity than the original LFP and RuO_2

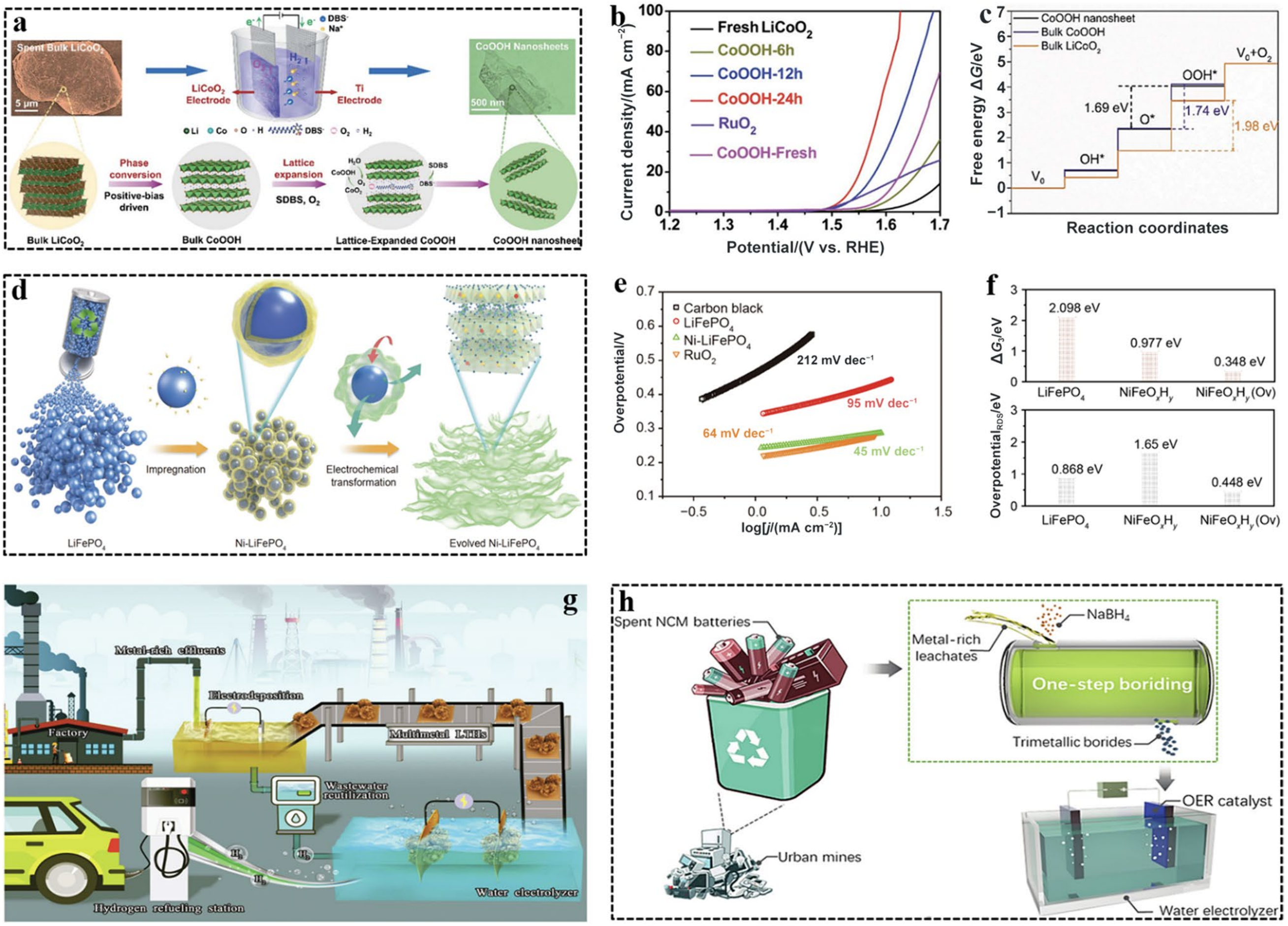

Fig. 6 **a** Schematic illustration of the in situ positive-bias driven exfoliation of $LiCoO_2$ into CoOOH; **b** polarization curves of various catalysts; **c** the Gibbs free energy profiles for the OER pathway. Reproduced with permission from Ref. [129]. Copyright © 2022, The Royal Society of Chemistry. **d** Schematic illustration of the proposed regeneration route for spent $LiFePO_4$; **e** the Tafel plots Ni-$LiFePO_4$, RuO_2, spent $LiFePO_4$ and carbon black; **f** the free energy change (ΔG_3) for the generation of *OOH in different models and the theoretical overpotentials for different models. Reproduced with permission from Ref. [133]. Copyright © 2021, Science China Press and Springer-Verlag GmbH Germany. **g** Schematic illustration of resource utilization of battery industrial wastewater. Reproduced with permission from Ref. [135]. Copyright © 2022, The Royal Society of Chemistry. **h** Schematic illustration on the synthesis of magnetic NCMB OER electrocatalysts from the spent LIB leachate via a facile boriding process. Reproduced with permission from Ref. [136]. Copyright © 2021, The Royal Society of Chemistry

benchmark catalyst, with a reduced overpotential of 285 mV at 10 mA cm^{-2} and a Tafel slope of 45 mV dec^{-1}. Its stability was well-maintained at 10 mA cm^{-2} for 14 h. The enhanced catalytic performance was attributed to the structural evolution and the introduction of the Ni promoter. The evolved nanosheets provided rich sites for reagents and products, and the numerous defects greatly boosted the OER kinetics. Furthermore, the addition of Ni optimized the free energy of *OOH intermediates, activating the Fe site effectively. The rich oxygen defects promoted oxygen desorption and synergistically improved the catalytic performance of LFP (Fig. 6e, f). This work corroborated the likelihood of converting spent LFP into high performance electrocatalyst. The spent LFP can be activated through appropriate modification, providing a new reference for designing advanced catalysts from a sustainable perspective of spent LIB recycling.

5.1.3 Spent NCM as Catalysts Sources

NCM, with low cost, high energy density, and good thermal stability, has gained increasing attraction as an alternative to $LiCoO_2$ cathode materials. A growing number of EVs, hybrid EVs, and energy storage equipment use NCM batteries as power sources, increasing the quantity of spent NCM batteries. Considering the high-value elements such as Ni and Co, efficient recycling of NCM has become the focus of academia and industry [134]. The industrial techniques of battery recovery include pyrometallurgy and hydrometallurgy. In the recycling process, hydrometallurgical recovery is more energy-saving, environmentally friendly, and efficient than pyrometallurgical recovery. Despite these advantages, industrial wastewater remains in this treatment. More environmentally friendly recycling and high-value utilization of NCM are urgently necessitated. Chen et al. [135] developed an economical and facile strategy to synergistically recover and reutilize metal ions (99.19% of Co, 99.15% of Mn, and 99.38% of Ni) from spent $LiNi_xCo_yMn_{1-x-y}O_2$ in industrial wastewater after hydrometallurgy (Fig. 6g). They extracted NiCoMn layered triple hydroxides (LTHs) with a hierarchical nanoflower structure from wastewater using electrodeposition. The electrodeposited electrode material was an efficient bifunctional catalyst for HER and OER. Additionally, it could be directly used to catalyze hydrogen production by electrolyzing the post-electrodeposition wastewater. As an HER catalyst, the optimal LTHs showed consistent catalytic activity with the Pt/C. In contrast, when catalyzing the OER, its catalytic performance was even better than the commercial RuO_2 due to the unique structure with considerable catalytic active sites and rich crystalline/amorphous phase boundaries. Encouraged by the excellent HER and OER catalysis performance, they further applied LTH||LTH couple to electrolyze post-electrodeposition wastewater for hydrogen production. This couple showed higher efficiency than the RuO_2||Pt/C couple, affording 10 mA cm^{-2} at voltages of 1.58 V vs. 1.62 V. Moreover, they also developed a one-pot boriding method to prepare magnetic mixed metallic borides as catalysts for efficient OER from the spent $LiNiCoMnO_2$ batteries (Fig. 6h) [136], attaining an overpotential of 372 mV at 500 mA cm^{-2} with superior cycling stability. It also outperformed the RuO_2 catalyst at high current densities, and the current density of the prepared catalyst at 1.53 V was 2.31 times higher than of RuO_2 catalyst. The excellent OER catalytic performance was attributed to the formation of core@shell structured tri-metal boride@(oxy)hydroxide nanostructures induced by boron etching. The current density remained stable for 12 h with a small loss (~ 5%) on the electrode.

Multi-metal oxide catalysts have excellent properties thanks to active site filling and electronic structure optimization. For example, Yang et al. [137] proposed an MCA method for selective recovery of $Ni_{0.5}Mn_{0.3}Co_{0.2}(OH)_2$ catalyst from spent $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ cathodes. After ball milling, the mixed spent $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ and $Na_2S{\cdot}9H_2O$, $Ni_{0.5}Mn_{0.3}Co_{0.2}(OH)_2$ nanoparticles were obtained. Deionized water was used to leach the Li species, and Li_2CO_3 can be precipitated using Na_2CO_3 as a precipitant. The production of $Ni_{0.5}Mn_{0.3}Co_{0.2}(OH)_2$ nanoparticles with increasing active sites exposed by MCA improved OER activity. It exhibited comparable OER activity to the commercial IrO_2. For instance, at a current density of 10 mA cm^{-2}, the required overpotentials of $Ni_{0.5}Mn_{0.3}Co_{0.2}(OH)_2$ nanoparticles and IrO_2 reached 0.28 V and 0.29 V, respectively. Furthermore, Lv et al. [138] developed an environmentally friendly and holistic method to promote spent LIB de-lithiation based on the direct current electric field (Fig. 7a). In the de-lithiation process, the lithium ions could be effectively recovered as Li_3PO_4 with simultaneous H_2 production. The de-lithiated electrode material ($Li_{1-\delta}Ni_{0.5}Co_{0.2}Mn_{0.3}O_2$) obtained a superior electrocatalytic performance. This improvement was ascribed to the high specific surface area of recycled cathode material, the abundant lattice oxygen and the increased oxidation state of Ni. Notably, the sample with the maximal de-lithiation degree exhibited relatively poor OER performance. The authors ascribed this phenomenon to the following two reasons: (i) the electrode materials partially peeled off from the conductive substrate due to the serious structural collapse after deep de-lithiation, leading to increased resistance; (ii) the electrophilic behavior of the de-lithiated materials were enhanced, favoring the adsorption of the hydroxyl group, yet over strong interaction hindered the electrocatalytic activity. This indicates that a relatively complete structure favors spent LIB materials as eligible catalysts.

Electrocatalytic oxygen production is a four-electron process with a higher reaction barrier than HER (two-electron process). This barrier serves as the rate-determining step in the overall water splitting. Accordingly, most research on the spent LIB electrode material-derived catalysts focuses on the catalytic performance of OER, leaving a research gap on the examples for HER, which is paramount in many fields. The commercial Pt/C catalyst exhibits excellent HER performance at a high cost. Thus, developing affordable and efficient HER catalysts is the premise of hydrogen production by water electrolysis. Waste LIB electrode materials with diverse valuable resources provide opportunities to prepare advanced HER catalysts, deserving more scholarly attention. Moreover, the in-depth mechanism of the OER and HER catalysts derived from spent LIBs remains inadequately explored. Furthermore, the research on the spent LFP recycling as catalysts is scarce. Additionally, its element cost is much lower than other LIBs, such as LCO and NCM, making pyrometallurgical or hydrometallurgical techniques to extract valuable elements unsuitable for spent LFP. The recycling path of spent LFP should be expanded, and converting it into catalysts to realize high-value applications is promising.

5.2 Syngas Preparation-Related Catalysis

Electrocatalytic CO_2 reduction syngas technology is effective in utilizing CO_2 resources and ideal for the sustainable synthesis of gas at room temperature, making it the current research hotspot based on the global targets for carbon neutrality [139, 140]. Materials recycled from spent LIBs can be used for hydrogen and oxygen preparation by water electrolysis and catalysis for the syngas preparation. For example, CO_2 can be converted into syngas (CO and H_2) using catalysts for CO_2 reduction reaction and HER [141]. Some studies proposed new routes for the utilization of the recovered products to catalyze the preparation of syngas. These ideas help solve the problem of spent LIB recycling and promote effective CO_2 use, presenting great significance to achieving the “carbon peak and carbon neutral” [142, 143].

Based on the above endeavors, Wang et al. [144] studied the influence of metal ratio in the Li-Co-Mn-Ni on electrochemical performance. They studied the difference among pristine NCMs with various metal ratios. On this basis, they found that the NCM622 (Ni:Co:Mn = 6:2:2) exhibited the best performance on catalyzing OER with the lowest Tafel slope of 82.3 mV dec^{-1} and the largest exchange current density of 0.215 mA cm^{-2}. The recycled NCM showed a consistent Tafel area and slope, indicating that the proved beneficial performance of Li, Ni, and Co to OER activity varied; in addition, the NCM with a relatively high cobalt ratio performed better despite the decreased activity due to elevated ratio of Mn. The influence of Li and Ni was not elucidated. Besides, thermal treatment temperature, time, and atmosphere were also discussed, and it was demonstrated that the NCM performance could be improved by proper heat treatment. The recycled material was applied as an anode catalyst in CO_2 electrolysis to produce CO, presenting a promising choice in industrial application with an excellent current efficiency of 97% under a high current density of 32 mA cm^{-2} at 2.25 V and an energy efficiency of 63%. Yu et al. [145] recycled Al foils and $LiCoO_2$ electrode materials from waste LIBs as catalysts to convert wood chip pyrolysis gas into hydrogen-rich gas. The introduction of the Al element promoted the in-situ atomic replacement reaction, fixed the volatile lithium elements, and transformed $LiCoO_2$ into $LiAlO_2$ with a higher melting point. The perfect Co nanoflower structure and stable CoO nanopore network were improved under the substitution reaction, forming an in-situ assembly catalytic structure with enhanced adsorption capacity, in line with the Li-CO_2 battery system with an 11.31 mmol g^{-1} hydrogen production rate and a 65.79% volume fraction, and the volume fraction of syngas ($H_2 + CO$) reached 91.82%. This marks a 90% improvement in the purity of the syngas prepared from Co-based catalytic materials for the first time. In addition, the in-situ assembled catalytic structure is eventually degraded to Co and $LiAlO_2$, allowing for ready recycling through the sorting method and avoiding the pollution of heavy metals from the waste catalysts (Fig. 7b). Spent LIB recycling mainly centers on cathode and anode materials. However, other components (such as electrolytes) are also valuable to be recycled and will greatly threaten the environment without appropriate treatment. In recycling spent LIB cathode or anode materials, the residual electrolytes typically decompose thermally under high-temperature conditions, producing toxic gases. For this reason, reasonable treatment and utilization of the gas emitted during recycling is promising. Jung et al. [146] investigated the thermal decomposition behavior of the widely used battery electrolyte $LiPF_6$ in ethyl carbonate and ethyl methyl carbonate (EC/EMC = 1/2 by volume). $LiPF_6$ pyrolysis was validated to yield different gaseous products (H_2, CO, CH_4, C_2H_4, and CO_2), which are difficult to separate. Based on the above analysis, finding an effective catalyst from the components of spent LIBs is significant for their reasonable recycling. In this context, the cathode material NCM 811 ($LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$) was found to be highly active for syngas formation and can be directly deployed as catalysts. This attribute mainly arises from the presence of Ni, Co, and Mn alloys (oxides), widely known as C–C, C–H, and C–O bond catalysts, respectively. At high temperatures, it shows the ability to convert the entire liquid compound into syngas (H_2 and CO). The synergistic action of the catalyst with CO_2 boosted CO generation. Given that the metallurgical process for battery operates at near 1 300 °C, the thermolysis of battery electrolyte can be incorporated to maximize the recovery of organic and metallic compounds in spent batteries. However, the catalytic mechanism of cathode materials, e.g., NCM being directly used as catalysts for syngas formation,

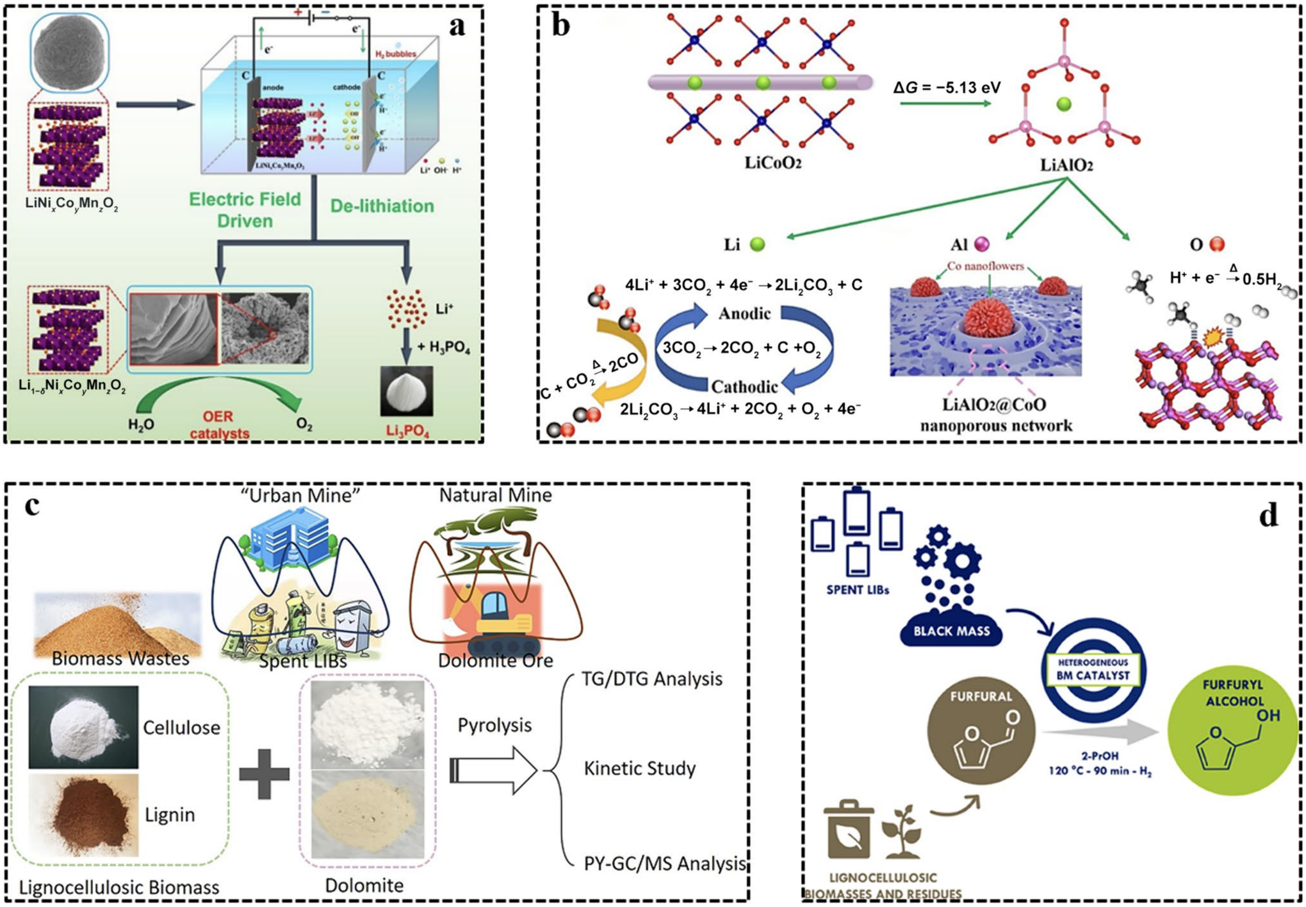

Fig. 7 **a** Schematic illustration of the electric field driven de-lithiation process for recycling the ternary $LiNi_xCo_yMn_zO_2$ electrode materials. Reproduced with permission from Ref. [138]. Copyright © 2020, Elsevier B.V. **b** Schematic illustration of the pyrolysis catalytic mechanism of sorption-enhanced in-situ-assembled porous structure. Reproduced with permission from Ref. [145]. Copyright © 2021, Elsevier B.V. **c** Schematic diagrams of recycling spent ternary LIBs for the dolomite modification and the application. Reproduced with permission from Ref. [149]. Copyright © 2021, Elsevier Ltd. **d** Schematic illustration of the catalyst from spent LIBs for biomass-derived aldehydes and ketones. Reproduced with permission from Ref. [151]. Copyright © 2022, American Chemical Society

needs more in-depth studies to guide the follow-up research and practical application.

5.3 Biomass Conversion-Related Catalysis

In the context of the ongoing consumption of fossil fuel sources and increasing energy demand, it is urgent to find new energy sources. Biomass is vital in the energy market due to its high efficiency in pyrolysis for the preparation of desired syngas and bio-oil, electricity generation, and heat processing, in which catalysts play an important role [147]. Some examples demonstrated that recycled metal oxides from spent LIBs could be applied in biomass pyrolysis and conversion. Chen et al. [148] utilized spent ternary LIB cathodes to study the pyrolysis characteristics of cellulose and lignin, which showed good thermostability even at high temperatures (700 °C). They added catalysts prepared from spent LIBs to the biomass through one-step (direct mixing) and two-step (impregnation-drying) methods. The latter included citric acid leaching of the spent cathode material, mixing of the solution with biomass, and the mixture drying. Consequently, it improved the catalytic effect on biomass pyrolysis, prominently reduced the content of cellulose-derived anhydrosugars, and increased the ketone levels by virtue of dehydration and decarboxylation. This method has a high potential for biomass pyrolysis or gasification in promoting the catalytic cracking (i.e., hydrogen transfer) of lignin-derived phenols (tar surrogates) to hydrocarbons and aliphatics (e.g., ketones). Besides, Wang et al. [149] used the leaching solution derived from the spent ternary LIBs for the modification of dolomite (Do), which can catalyze biomass pyrolysis (Fig. 7c). The modified Do exhibited excellent performance in reducing the activation energy and upgrading the volatile products. For the cellulose and lignin pyrolysis, the activation energy was noticeably decreased from 201 and 180 kJ mol^{-1} to 80 and 75 kJ mol^{-1}, respectively. It was ascribed to significant facilitation of the modified Do, which converts anhydrosugars into small-molecule components (e.g., ketones). In addition, changes occurred in the production of phenols (50% vs. 5.8%) and hydrocarbon (0.6% vs. 30.3%). This newly synthesized catalyst for biomass pyrolysis presents a good opportunity to valorize spent LIBs and biomass wastes. Different from the above examples, effective catalysts can also be prepared without the leaching process, which usually need the assistance of acids. Fuentes et al. [150] prepared a mixed oxide (Co/RZnO) by milling the recycled oxides ZnO (RZnO) from spent alkaline batteries and CoO (RCoO) from spent LIBs. The mixed oxides were used to investigate the chemical recycling of polyethylene terephthalate into bis(2-hydroxyethyl)ethylene terephthalate (BHET). The Co/RZnO showed excellent catalytic performance, which was attributed to the presence of weak and strong acid sites, its overall higher concentration of acid sites and the synergistic effect between Co_3O_4 and ZnO. The yields of BHET monomer were 80% (Co/RZnO), 50% (RZnO), and 10% (RCoO), respectively.

The mixture of anodic and cathodic materials from mechanical shredding of spent LIBs is called "black mass (BM)". The simple and crude use of hydrometallurgy and pyrometallurgy to recycle this material can cause a serious waste of resources since the carbon-based anode can be burned out due to high temperatures. A reasonable recycling design is necessary to avoid this waste and reuse the anode materials such as graphite rationally. Paone et al. [151] converted the BM into an efficient heterogeneous catalyst for the selective hydrogenation of biobased furfural and various biomass-derived aldehydes and ketones (Fig. 7d). The BM was filtered to homogenize the particle sizes and remove solid impurities (e.g., Al and Cu pieces), ball milled, calcined under atmospheric conditions at 600 °C for 6 h to erase traces of binder materials, and reduced at 500 °C for 6 h under H_2 flow to obtain the desired catalyst. Under the catalysis of the prepared catalyst, furfural could be fully converted into furfuryl alcohol, the sole reaction product, at 120 °C for 90 min in the presence of molecular hydrogen under batch conditions. These studies present new routes for recovering and utilizing spent LIBs as the catalysts for biomass decomposition and conversion. However, most examples are limited to hydrometallurgy or pyrometallurgy-like methods, weakening the advantages of the original structure of spent LIB materials.

In summary, we discussed the application of catalysts derived from spent LIBs in substance conversion, including electrolytic water splitting, syngas preparation, and biomass conversion, an interdisciplinary subject of energy and environment. Furthermore, the spent LIB material recycling as catalysts for substance conversion and the corresponding performance are shown in Table 2. Most proposed strategies are hydrometallurgy or pyrometallurgy-like types, i.e., extracting the useful elements (Ni, Co, and Mn) by acid leaching or conversion of the spent LIB cathode materials into metal oxides by calcinating and using the recycled compounds to synthesize desired catalysts. In principle, such strategies are more universally applicable and have the potential to reuse spent LIBs. Nevertheless, some intrinsically good properties of spent LIB materials for catalysis are lost in the rough recycling. In awareness of this deficiency, researchers have tried non-structurally destructive or low-structurally destructive strategies using spent LIBs materials as catalysts. These attempts present attractiveness and competitiveness and achieve comparable catalytic performance under certain circumstances. Despite these yields, such research is still lacking, highlighting the importance of exploring catalytic mechanisms on this basis.

6 Battery-Related Catalysis

With the ongoing advancement in new energy technologies, electrochemical energy storage devices with fast and convenient characteristics have attracted much attention. Different from traditional "rocking-chair" batteries, such as LIB and sodium-ion batteries, a variety of new-generation battery technologies, including LSBs, metal-air batteries, and fuel cells (power generation), are related to catalytic reactions [152–154]. Developing advanced catalysts is crucial for the performance improvement of these battery systems and has gained increasing attention and progress. For LSBs, the electrode catalysts mainly include metal-free catalysts (e.g., carbon-based materials and polymers) [155, 156], metal-based catalysts [157], and hetero-structure catalysts [158]. For metal-air batteries, in addition to precious metals and their oxides, carbon-based metal-free and transition metal-based materials are two kinds widely used as bifunctional catalysts for electrode reactions [159]. As for fuel cells, the commercial electrode catalyst is Pt/C, and non-noble metal catalysts are currently being developed for cost reduction and sustainability, targeting transition metals and their compounds [160]. In general, carbon-based materials and transition metal compounds are promising catalysts for the three kinds of battery technologies above.

The Li, Co, Ni, and Mn elements in spent LIB cathode materials, Al and Cu from current collectors, and graphite from anodes are related to the above catalyst materials with regard to transition metal compounds and carbon-based materials. Meanwhile, considering the unique crystal structures of existent commercial cathode materials, they merits potential catalytic activity, and the volume changes, defect sites, and strains in spent cathodes might promote the electro-catalytic kinetic process [161]. Additionally, the SG anode still preserves the fusion graphite structure after extended charging and discharging cycling and is qualified to prepare advanced carbon materials, such as graphene, to support catalysts [162]. To sum up, the application of recycled products from spent LIBs to battery-related catalysis technology, such as LSBs, fuel cells, and metal-air batteries, is worth exploring with profits and feasibility. In this chapter, we will summarize and discuss its classification in detail.

6.1 LSB Related Catalysis

LSBs have become a competitive candidate for next-generation energy storage devices due to their high theoretical energy density and eco-friendly features of sulfur. Based on the $S_8 + 16Li^+ + 16e^- \rightarrow 8Li_2S$ conversion reaction, LSBs exhibit a high theoretical capacity of 1 675 mAh g^{-1} and an energy density of 2 567 Wh kg^{-1}, which is five times higher than that of commercially available LIBs (387 Wh kg^{-1} for $LiCoO_2$/graphite batteries) [163]. Despite such a benefit, the fast capacity degradation stemming from the sluggish redox reaction kinetics and shuttle of fatal lithium polysulfides (LiPSs) hinder the implementation of LSBs [164]. Various rational design strategies have been proposed, involving inserting interlayers and introducing electrocatalytic sulfur host materials to solve or alleviate these issues. Manthiram's team [165] used interlayers to suppress the shuttle effect in 2012 by presenting a conductive multi-walled carbon nanotube paper between the cathode and the separator. The paper acted as a pseudo-upper current collector, improving the cyclability of LSBs, and its role is shown in Fig. 8a. On this basis, carbon-based, metal-based, and polymer-based interlayers have been extensively studied to ameliorate the electrochemical performance of LSBs.

However, the preparation methods of these functional materials are usually environmentally harmful, costly, and complicated with application barriers. Additionally, the abundant graphite and metal resources in spent LIBs present a good choice for the preparation of LSB catalysts and have received increasing attention. Based on the idea of turning waste into treasure, Xu et al. [166] prepared a functional interlayer to enhance polysulfide trapping and catalytic performance for LSBs by utilizing waste graphite recycled from spent LIB anode materials (Fig. 8b). The as-prepared SG showed more polar functional groups, defects, and porous structure compared to artificial graphite (AG) (Fig. 8c, d). Compared with AG, the SG possessed unique physical and chemical properties. For one thing, the porous structure and defects provided numerous adsorption sites to restrict solvable polysulfides. For another, the electrolyte decomposition introduced substantial polar functional groups on the graphite surface, facilitating polysulfide adsorption and electrolyte wetting. Transition metal elements can also anchor polysulfides, enhance electrical conductivity, and boost polysulfide conversion kinetics, such as Ni, Co, and Mn, which were introduced into SG owing to the cathode active material dissolution. Accordingly, applying the SG-modified separators for LSBs achieved superior cycling stability (low capacity-decay of 0.08% per cycle over 500 cycles at 1 C). In addition, it exhibited excellent rate performance with high reversible specific capacities of 1 317 and 813 mAh g^{-1} at 0.1 C and 2 C, higher than those of cells assembled with bare polypropylene and AG-modified separators (Fig. 8e).

Besides spent anode materials, cathode materials with unique layer structures are applicable to modify the LSB separators, as verified by Liu and co-workers [167]. In particular, the spent $LiCoO_2$ was coated on a commercial polypropylene separator (Fig. 8f), accelerating the redox reactions and suppressing the LiPS diffusion due to the spinel Co_3O_4. The assembled LSBs with spent $LiCoO_2$ delivered a high capacity retention of 74.68% at 1 600 mA g^{-1} over 200 cycles and superior rate capability. In addition to the modification of the functional separator to improve the performance of LSBs, developing advanced sulfur host materials, such as carbonaceous materials and metallic compounds with exceptional attributes, is a popular strategy. Yang et al. [168] employed SG without complex treatment from spent LIBs for the LSB cathode. The SG/S cathode exhibited an initial discharge capacity of 1 377 mAh g^{-1} at 0.2 C and outstanding cycling stability of capacity retention of 765 mAh g^{-1} after 500 cycles with a decay rate of 0.006% per cycle. The electrochemical performance is enhanced since the SG surface possesses abundant functional polar groups and metal elements during repeated charging/discharging, displaying excellent electronic conductivity and promoting the LiPS conversion kinetics efficiently. With the goals of efficient recovery of the ternary cathode (Li-NCM) and the effective performance improvement of LSBs, Liu and colleagues developed a natural sphagnum uptake of spent Li-NCM-based LIBs as host to load sulfur, as shown in Fig. 8g [169]. The obtained cathode exhibited high sulfur ratios (64% of the total electrode and 80% of the active material mass); additionally, it benefited from the chemical encapsulation and hierarchical structural stabilization, delivering a good specific capacity of 700 mAh g^{-1} after 300 cycles.

Considering the key scientific problems of the LiPS shuttle effect and the slow electrode reaction kinetics faced by LSBs, the functional modification of separators and the introduction of sulfur host materials from spent LIB cathodes and anodes were proposed to enhance the electrochemical performance. This field emerges with limited research, and the results obtained previously demonstrate its application prospects with in-depth development, such as combination with other LSBs for traditional catalysts and advanced treatment of spent cathode and anode materials to prepare graphite-supported metallic compound catalysts by direct mixing. These studies have been well-designed to convert spent LIB materials into catalysts, enabling reasonable use

Table 2 Recycling of spent LIB materials for substance conversion catalysts and corresponding performance

Spent material types	Regenerative methods	Catalyst	Application	Performance	References
$LiCoO_2$	Electrochemical activation	$Li_{1-x}CoO_2$	OER	Delivering a current density of 9.68 mA cm^{-2} at 1.65 V after 500 cycles	[126]
$LiCoO_2$	Leaching and calcination	HT-$LiCoO_2$	OER	Delivering a higher anodic peak potential of 0.35 V	[128]
$LiCoO_2$	In situ positive-bias driven exfoliation	CoOOH nanosheet	OER	Delivering a low Tafel slope of 53.8 mV dec^{-1} and a small overpotential of 305 mV at 10 mA cm^{-2} after 65 h long-term electrolysis	[129]
$LiFePO_4$	Electrochemical transformation	$NiFeO_xH_y$	OER	Delivering a low overpotential of 285 mV at 10 mA cm^{-2} and a small Tafel slope of 45 mV dec^{-1} well-maintained for 14 h	[133]
$LiNi_{1-x-y}Mn_xCo_yO_2$	Electrodeposition	NiCoMn layered triple hydroxides	OER and HER	Delivering a low voltage of 1.583 V to drive 10 mA cm^{-2} for the waste-water electrolysis	[135]
$LiNiCoMnO_2$	One-pot boriding method	NiCoMnBs	OER	Delivering an overpotential of 372 mV at 500 mA cm^{-2}, remained stable for 12 h running	[136]
$LiNi_{1-x-y}Mn_xCo_yO_2$	Acidic leaching, precipitation, hydrothermal and calcination	$MnCo_2O_4$	OER	Delivering an overpotential of 358 and 400 mV at 5 and 10 mA cm^{-2}	[120]
$LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$	Mechanochemical activation method	$Ni_{0.5}Mn_{0.3}Co_{0.2}(OH)_2$	OER	Delivering an overpotential of 280 mV at 10 mA cm^{-2}	[137]
$LiCoO_2$	Thermal treatment	$LiAlO_2$@CoO	Convert wood chip pyrolysis gas into hydrogen-rich gas	The yields and volume fractions of H_2 are 11.31 mmol g^{-1} and 65.79%, and the volume fraction of synthetic gas reaches 91.82%	[145]
$LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$	Thermal treatment	$LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$	Syngas formation using CO_2-assistance	Highly active for syngas formation $\leqslant$500 °C	[146]
$LiNi_{1-x-y}Mn_xCo_yO_2$	Acidic leaching and calcination	Ni, Co, Mn-modified dolomite	Biomass pyrolysis	Decreasing the apparent activation energy for cellulose and lignin pyrolysis, inhibiting the production of phenols and enhancing the production of hydrocarbons	[149]
$LiCoO_2$	Acidic leaching and calcination	Co/RZnO	Glycolysis of poly(ethylene terephthalate)	The catalytic efficiency reached 80%	[150]
$LiNi_{1-x-y}Mn_xCo_yO_2$	Calcination	Ni-Mn-Co oxides	Reductive upgrading of biomass derived furfural	Complete conversion of furfural into furfuryl alcohol is achieved after 90 min at 120 °C under 10^6 Pa H_2 in 2-propanol	[151]

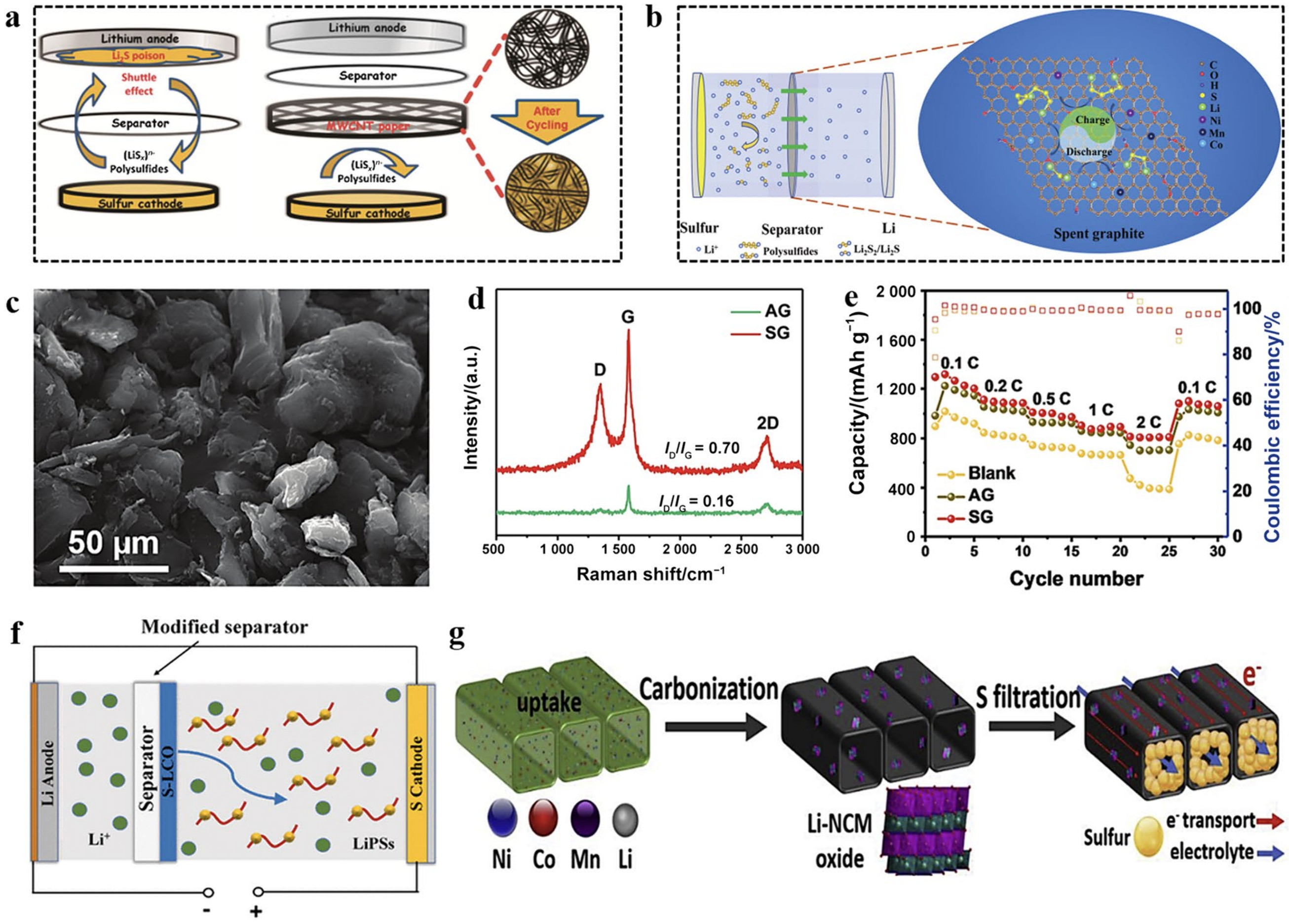

Fig. 8 **a** Schematic cell configuration of rechargeable LSBs without & with interlayer. Reproduced with permission from Ref. [165]. Copyright © 2012, The Royal Society of Chemistry. **b** Schematic of LSBs assembled with spent-graphite-modified separator; **c**, **d** SEM and Raman spectra of AG and SG; **e** the rate performance from 0.1 to 2 C of Li-S batteries with different separators. Reproduced with permission from Ref. [166]. Copyright © 2021, The Royal Society of Chemistry. **f** Schematic diagram of separator modification by spent $LiCoO_2$. Reproduced with permission from Ref. [167]. Copyright © 2022, American Chemical Society. **g** Schematic diagram of a naturally sphagnum uptake of spent Li-NCM based LIBs as host to load sulfur. Reproduced with permission from Ref. [169]. Copyright © 2019, Elsevier Ltd

of the inherent catalytic properties of those materials failed in LIBs. Pertinent research in this field is encouraged.

6.2 Fuel Cells Related Catalysis

Fuel cells as an energy conversion device with high power generation efficiency (theoretically 85%–90% without restricted by the Carnot cycle), environmentally friendly, high energy density and wide source of fuel, are considered the fourth-generation power generation technology to convert chemical energy into electrical energy, following hydro, thermal, and nuclear power. Typically, the fuel cell is composed of a cathode, anode, electrolyte, and external circuit. The oxygen reduction reaction (ORR) at the cathode with a high energy barrier determines the system kinetics. Developing advanced catalysts to reduce energy consumption is the cornerstone of the large-scale commercial application of fuel cells. Pt-based catalysts with excellent catalytic activity are popular in fuel cells [170, 171]. Cui et al. [172] tuned the catalytic activity of Pt catalysts by utilizing the widely tunable lattice constant of LIB electrode materials as the catalyst support (Fig. 9a). Modifying the electronic structure of the catalyst can improve activities. In contrast, either compressive or tensile, the lattice strain can alter the surface's electronic structure and upgrade the catalyst activities. During the charging/discharging process, LIB electrode materials undergo volume changes by depositing small Pt nanoparticles onto the $LiCoO_2$ or $Li_{0.5}CoO_2$ substrate surfaces and controlling the charging and discharging states of these substrates. Consequently, the ORR activities of Pt nanoparticles in an alkaline solution achieved approximately 90% improvement or more than 40% decrease under compressive and tensile strain, respectively. However, the scarcity and high cost of Pt resources limit its further large-scale application, presenting an urgent need to explore alternatives.

To improve the added value of end-of-life LIBs, Liivand et al. [173] used spent LIB graphite as a precursor material for preparing graphene oxide, followed by doping to obtain nitrogen-doped graphene. It was found that the nitrogen-doped graphene from the spent LIBs showed higher electrocatalytic activity toward ORR than commercial nitrogen-doped graphene. This is attributed to the increased content of active nitrogen species and the presence of carbon vacancies on the graphene surface. Specifically, the ORR onset potential for the nitrogen-doped graphene prepared by the spent LIBs and the commercial nitrogen-doped graphene were 0.867 V and 0.797 V, respectively. After injecting 3 mol L^{-1} methanol into the O_2-saturated 0.1 mol L^{-1} KOH solution, the current of the experimental sample exhibited a smaller decrease than the commercial one (16.5% vs. 33%), indicating that the spent LIB-derived nitrogen-doped graphene has a better catalytic long-term performance. Warczak et al. [174] proposed a hydrogen peroxide (H_2O_2) generation method using the leached LIB waste powder as the catalyst, showing higher electrocatalytic activity toward ORR than the unmodified glassy carbon. When assembled at a liquid-liquid interface, the waste powder increased the efficiency of H_2O_2 generation by about 20 times. The H_2O_2 has been applied as a fuel and oxidant in fuel cells [175, 176], suggesting that the as-prepared catalyst has the potential to be applied in fuel cells. Besides, Ruan and colleagues [177] prepared graphene nanosheets using SG anode from LIBs via a low-cost and green synthesis route (Fig. 9b). Especially, oxidation efficiency was improved by introducing a 5:1 mixture of H_2SO_4/H_3PO_4 and increasing $KMnO_4$ content in the graphite recycled from spent LIBs. Subsequently, high-quality graphene was obtained through a combined method of glucose reduction and annealing. The spent LIB-derived nitrogen-doped graphene exhibited favorable activity and stability for ORR, better than the Pt/C (Fig. 9c). The activity of nitrogen-doped graphene degraded about 7%, lower than that of the Pt/C catalyst (near 20%) in the durability test. Ruan's group [178] also prepared Fe-N-doped carbon catalyst for ORR by recycling graphite anode from spent LIB. The SG recycled from spent LIB anodes was used as a carbon carrier and doped with N and Fe via simple pyrolysis with polyaniline and iron salt. As a result, this catalyst displayed excellent ORR catalytic activity in 0.1 mol L^{-1} KOH solution with an onset potential of 0.91 V vs. RHE and a half-wave potential of 0.8 V vs. RHE. Under the same conditions, the durability and methanol resistance of the as-prepared Fe-N-doped carbon catalyst from spent LIBs exceeded those of a 20%Pt/C commercial catalyst. Furthermore, the prepared catalyst had better ORR catalytic stability than the commercial 20%Pt/C, and they maintained initial currents of 93% and 82% after running at a fixed potential of 0.38 V for 36 000 s.

The above findings indicated that the recycled products from spent LIB anodes can be transformed into catalysts for the ORR and applied to the catalysis for fuel cells. On the bright side, the affordable spent anode graphite is mainly used as ORR reaction catalysts for fuel cells in these samples, achieving the high-value recycling of the resource. Notably, recycling spent cathode as catalysts for ORR remains a research gap. To realize "closed-loop" recycling, more attention should be directed to the waste cathode materials, which contain high-value metals and can be used as precursors to prepare metal-based catalysts for fuel cells. For example, Ruan and colleagues proposed the employment of iron salt to synthesize the desired catalyst [178]. The spent LFP is rich in iron, presenting the potential of applying the iron in LFPs to replace the iron salt.

6.3 Metal-Air Battery-Related Catalysis

As an emerging battery technology with a high theoretical energy density and safety, metal-air batteries, consisting of metal anode and air cathode [179], have received more attention. The sluggish kinetics process of ORR and OER at the air cathode hinders its further large-scale application. Developing bifunctional catalysts with high activity for ORR/OER is crucial for improving the performance of metal-air batteries [180, 181]. The excellent performance of transition metal-based catalysts (e.g., Co, Ni, Fe, and Mn) for OER/ORR has been proved. LIB electrode materials with abundant transition metals can act as bifunctional electrocatalysts or catalyst precursors for metal-air batteries such as Zn-air and Li-air batteries. Manthiram et al. [182] have reported that $LiCoO_2$ synthesized at 400 °C with a spinel structure exhibited superior OER and ORR activity and relatively long service life, making it suitable to be a bifunctional electrocatalyst for rechargeable metal-air batteries. In 2020, Jin et al. [183] also used the LFP materials as Fe and P sources to react with ZIF-8 for the preparation of Fe-N-P co-doped carbon nanotube encapsulated Fe_2P nanoparticles (C-ZIF/LFP) for efficient ORR. The C-ZIF/LFP catalyst showed superior ORR activities under alkaline and acidic conditions. It achieved a high half-wave potential of 0.74 V in acidic electrolytes, a half-wave potential of 0.88 V in alkaline electrolytes, and a peak power density of 140 mW cm^{-2} in Zn-air batteries. The above analysis indicated that the $LiCoO_2$ and LFP electrode materials can act as precursors to prepare metal-air battery catalysts, delivering superior performance.

Spent LIB electrode materials with valuable transition metals have gained growing attention for catalysis in metal-air batteries. Our team has conducted studies based on the zinc-air battery catalysts and the reuse of spent LIBs [184,

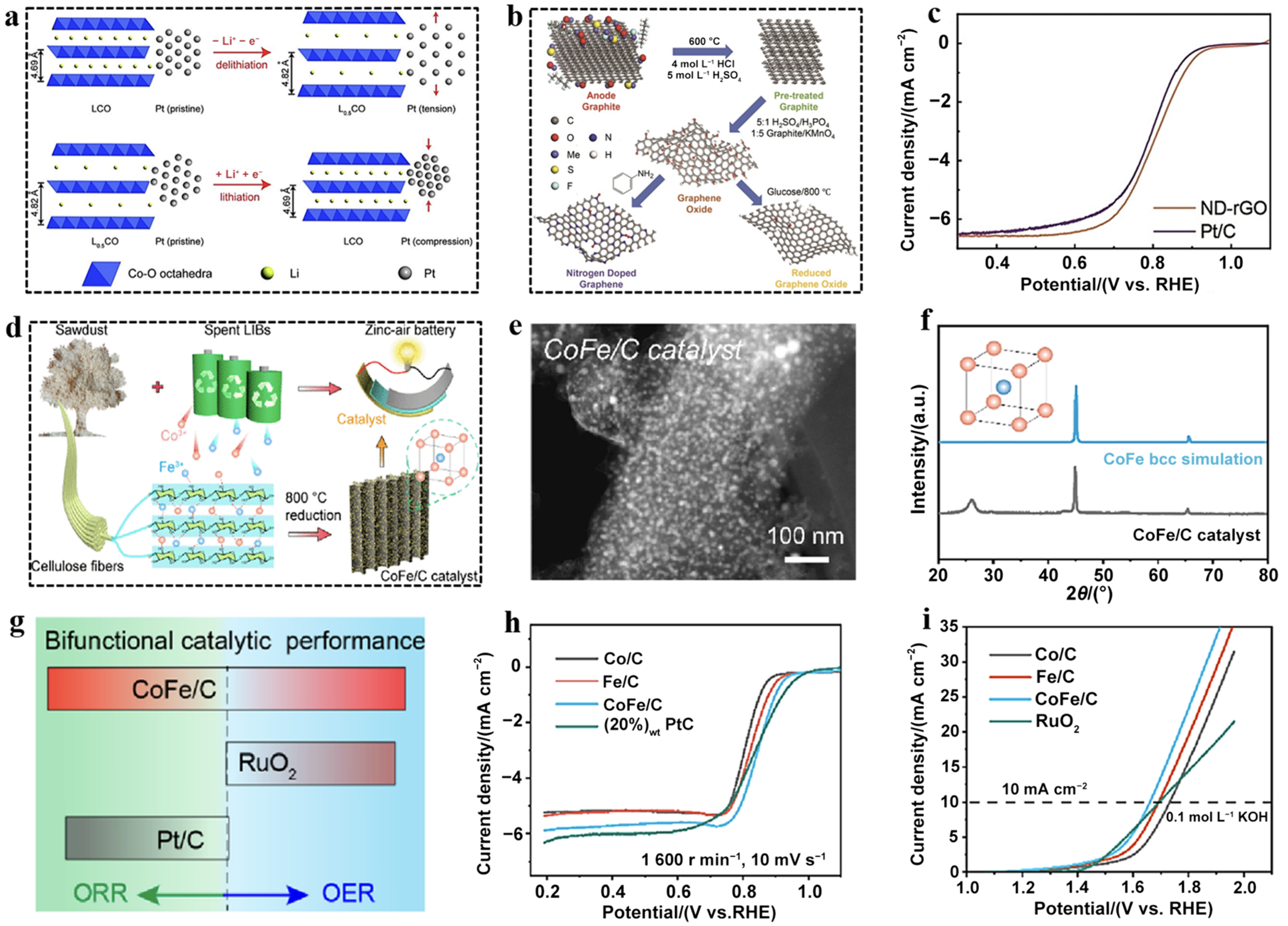

Fig. 9 **a** Schematic of the lattice constant change of LCO substrates and how the lattice strains are induced to Pt NPs. Reproduced with permission from Ref. [172]. Copyright © 2016, American Association for the Advancement of Science. **b** Preparation process of the nitrogen doped graphene and reduced oxide; **c** LSV of ND-rGO at 1 600 r min^{-1} and scanning rate of 0.005 V s^{-1} as compared to the Pt/C. Reproduced with permission from Ref. [177]. Copyright © 2021, Wiley-VCH GmbH. **d** Schematic diagram of design of the CoFe/C catalyst from spent LIBs; **e–g** STEM image, XRD spectrum and related crystal structure, and the bifunctional catalytic performance of the CoFe/C catalyst, respectively; **h**, **i** ORR curves and OER polarization curves of the related catalysts. Reproduced with permission from Ref. [184]. Copyright © 2022, American Chemical Society

185]. We reported a highly efficient bifunctional CoFe/C catalyst for zinc-air batteries using sawdust as the hosting agent to anchor recycled Co and Fe from spent LIB cathode materials, followed by simple carbonization (Fig. 9d) [184]. The as-prepared CoFe/C catalyst showed uniformly dispersed CoFe nanoparticles on the sawdust-based carbon after annealing (Fig. 9e) due to the hydroxyl groups in sawdust and electrostatic attraction between the Fe^{3+}/Co^{3+} cations. The electron redistribution in the CoFe/C catalyst induced by the Fe atoms were all isolated into single sites by the Co atoms (Fig. 9f), contributing to the consistent ORR/OER performance of CoFe/C with the commercial catalysts (Fig. 9g). When using as-prepared CoFe/C as the ORR and OER catalyst, it delivered a 0.85 V half-wave potential and a 1.66 V overpotential at 10 mA cm^{-2} (Fig. 9h, i). The assembled zinc-air batteries with CoFe/C catalyst on the cathode showed a noticeably large power density of 199.2 mW cm^{-2} and a long cycling stability of 350 h. Moreover, we developed a rapid thermal radiation process through rational design to prepare the NiMnCo-based catalyst from spent LiNi$_{1-x-y}$Mn$_x$Co$_y$O$_2$ cathodes for highly efficient zinc-air batteries (Fig. 10a) [185]. The as-prepared NiMnCo-activated carbon catalyst exhibited a unique core-shell structure involving spinel NiMnCoO$_4$ in the shell and face-centered cubic Ni in the core with the redistributed electronic structure of NiMnCoO$_4$ to decrease the energy barrier for OER/ORR. The corresponding characterizations and simulation structure are shown in Fig. 10b–d. The synergistic effect between the $NiMnCoO_4$ and the Ni core was proved by the density functional theory calculations. It was elucidated that the $NiMnCoO_4$ shell provided efficient active sites for the ORR/OER processes, and the Ni core redistributed the $NiMnCoO_4$ shell electronic structure (Fig. 10e, f). In the case of zinc-air battery cathode catalyst, the obtained NiMnCo-activated carbon catalyst delivered a low voltage gap of 0.72 V at the initial three cycles, high power density of 187.7 mW cm^{-2}, and superior cycling stability of 200 h at 10 mA cm^{-2} (Fig. 10g–i). These results provide an economical and environmentally friendly way for spent LIB disposal and metal-air battery development.

Wei et al. [186] also recycled Ni-Co-Mn oxides from spent LIBs to prepare bifunctional air electrodes, which realized 100% regeneration of Co, Ni, and Mn from the spent cathodes via a thermal treatment. Additionally, the Ni-Co-Mn oxides changed from an α-$NaFeO_2$-type structure to a spinel one after being heated at 600 °C. The regenerated Ni-Co-Mn oxides showed excellent bifunctional catalytic activities towards OER and ORR in aqueous KOH electrolyte; when applied to a zinc-air battery cathode, their calculated energy efficiency was approximately 75%. Yang et al.[187] reported an ultrafast method to regenerate $LiCoO_2$ from spent LIBs into the carbon layer/Co_3O_4 cathode for rechargeable zinc-air batteries. The spent $LiCoO_2$ was converted into nanosized Co_3O_4, and the cathode polymer binder (such as PVDF) was carbonized into a graphitized carbon layer after ultrafast high-temperature shock treatment. The graphitized carbon layer enhanced the electronic conductivity, and the nanosized Co_3O_4 boosted active sites. Consequently, the as-prepared carbon layer/Co_3O_4 catalyst delivered efficient catalytic activities, exhibiting a low OER overpotential of 245 mV at 10 mA cm^{-2}, and the ORR onset potential reached 0.90 V. Furthermore, the assembled aqueous zinc-air battery displayed high power density (131 mW cm^{-2}) and excellent cycling durability. The above representative reports have laid a foundation for the application of waste LIB electrode materials in advanced meal-air batteries. On this basis, a growing body of research, such as innovative synthesis methods, in-depth mechanism exploration, and excellent energy storage performance investigation, will arise.

To meet the increasing demand for energy supply and environmental quality requirements in the context of refined social civilization, various advanced energy storage technologies beyond LIBs are developed and await extensive application. This chapter concludes the latest progress of spent LIB electrode materials for battery-related catalysis, including LSBs, fuel cells, and metal-air batteries. They can solve the problems of environmental damage caused by improper disposal of end-of-life LIBs and improve the electrochemical performance of the waste effectively to improve its practical value. The recycling of spent LIB electrode materials for battery-related catalysis and the corresponding performance are summarized in Table 3.

Currently, calcination and acid leaching are the main methods for battery-related catalyst preparation. Considering high energy consumption and the risk of secondary pollution, greener regeneration methods have become the focus of future research. Some studies pioneeringly proposed the rational use of spent LIB materials by simplifying the recycling process and maximizing the structural and elemental advantages of failed materials. Despite these insightful contributions, many important issues remain. For example, the assumption that the defects in spent materials serve as sites promoting the catalytic performance lacks experimental evidence, deserving elaboration, and advanced characterizations.

Furthermore, impurities in spent LIB materials may affect the intrinsic or apparent performance of batteries. For example, the unknown impurity quantity interferes with the quantification of the active substance in the electrode, hindering the determination of the actual properties of the active materials. In addition, the impurities influence the electric conductivity of the catalysts or electrodes, impacting the charge transfer and the reaction kinetics. Moreover, the potential effect of the catalyst surface impurities on the gas diffusion layer can change the gas adsorption and desorption rates, leading to variations in the reaction rate. The unremoved impurities such as binder (e.g., PVDF) may improve the catalytic performance by retarding the dissolution of the prepared catalysts from spent LIBs [83]. The binder can also be rationally utilized to improve catalyst performance; for instance, Yang et al. [187] carbonized it to enhance product conductivity. These examples infer that the impurities can be turned into treasure, as discussed in Sect. 2. New impurities introduced during recycling also have a potential impact on battery performance. Zheng et al. [188] realized the importance of this aspect and studied the effects of sulfide introduced during hydrometallurgical recycling on the synthesized NCM622 cathode materials. The findings indicated that the sulfide impurity had a limited impact on the battery performance. Such studies can guide the screening of suitable reagents in recycling. However, research in this regard is lacking. Additionally, the number of impurities differs and cannot be controlled. This means that despite the consequence of impurities, it is difficult to ensure the batch uniformity of the products, hampering large-scale production. In this sense, the two directions of fine sorting and utilizing various spent LIB components need to be developed in tandem. The mechanism of impurities in catalysts affecting the performance of batteries remains

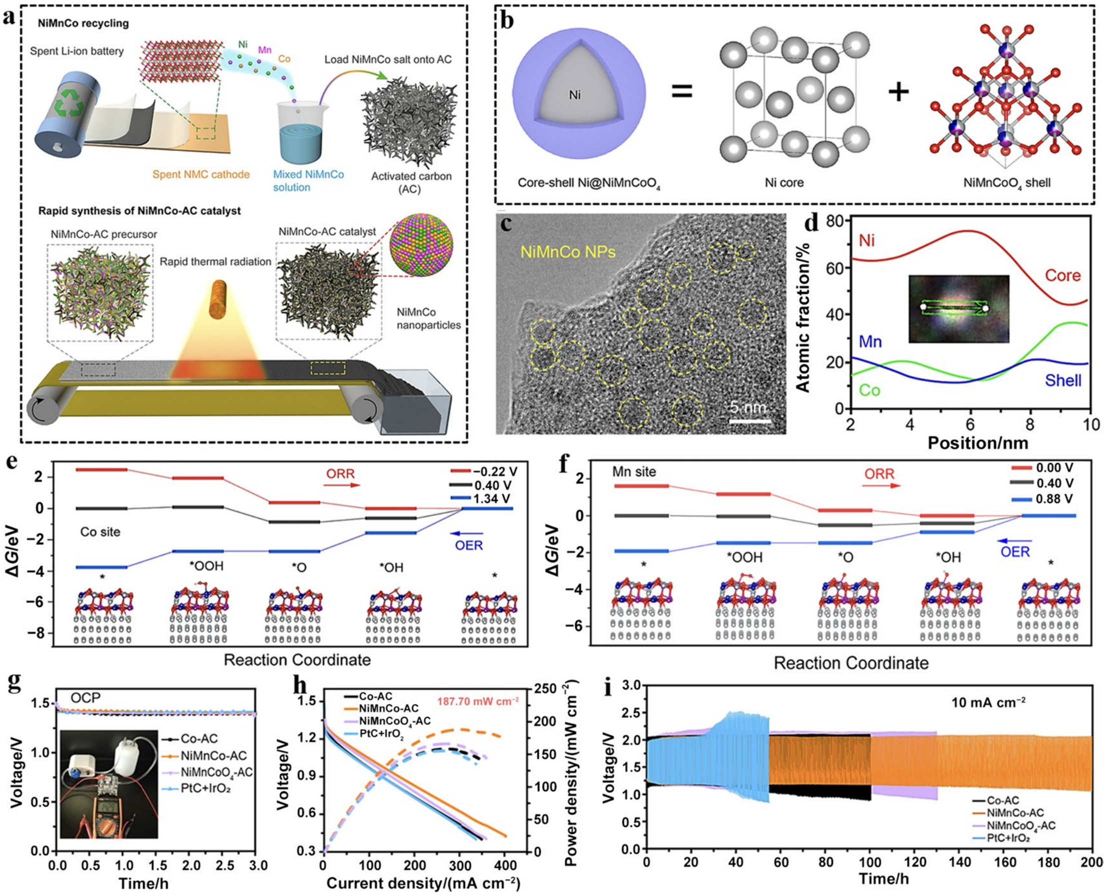

Fig. 10 **a** Schematic of the fabrication process of the NiMnCo-AC catalyst from spent LIBs; **b** identified core-shell structure of the NiMnCo nanoparticle; **c**, **d** HRTEM image and line-scan profiles of the NiMnCo-AC catalyst; **e**, **f** Gibbs free energy diagrams of ORR/OER on the Co sites and Mn sites of Ni(111)@NiMnCoO$_4$(110); the electrochemical performance of ZABs with various catalysts; **g** the open-circuit potential of primary ZABs at the initial 3 h; **h** the *i–v* polarization and power density curves of ZABs; **i** cycling performance of ZABs at the current density of 10 mA cm^{-2}. Reproduced with permission from Ref. [185]. Copyright © 2022, the Author(s)

underexplored, and its in-depth discussion will aid the effective utilization of components and the analysis of the catalyst failure mechanism.

Besides, the quantity of defects or the degree of degradation of spent LIB materials is not controllable for the decommissioned batteries. For this reason, the strategies relying on the original properties of spent LIB materials may exhibit limited versatility. By contrast, methods with structurally destructive properties, such as pyrometallurgy and hydrometallurgy, may ignore the different states of raw materials. The reason for this is that they primarily extract valuable elements into metallic compounds and use them for subsequent synthesis. In other words, for the battery material recovery in different states as catalysts, hydrometallurgy, and pyrometallurgy methods can ensure the product purity. For the strategies that rely on the initial structure of the spent LIB materials, the application of such methods needs strict classification of raw materials, putting forward requirements for the spent battery classification of the recycling industry chain upstream. In addition, developing the evaluation and detection technology of the spent battery status is also necessary to improve the recycling efficiency in the industrial chain downstream. It is recommended to distinguish between different types and states of spent batteries before the recovery processes. Generally, developing highly versatile strategies and effectively utilizing the advantage of failed materials still need significant endeavors.

7 Limitations on Recycling Spent LIB Materials for Catalytic Applications

The examples above suggest that recycling spent LIB materials for catalytic purposes is feasible and promising. However, this path has some limitations, reminding researchers to continually upgrade recycling technologies and expand their applications.

Table 3 Recycling of spent LIB materials for battery-related catalysts and corresponding performance

Spent material types	Regenerative methods	Catalyst	Application	Performance	References
Graphite	Without further treatment	Graphite	Li-S batteries (separator)	968 mAh g^{-1} with a low decay rate of 0.08% per cycle over 500 cycles at 1 C	[166]
$LiCoO_2$	Crystal-nondestructive process	S-LCO ($LiCoO_2+Co_3O_4$)	Li-S batteries	74.68% capacity retention over 200 cycles at 1 600 mA g^{-1}	[167]
Graphite	Without further treatment	Graphite	Li-S batteries (sulfur host)	765 mAh g^{-1} with a low decay rate of 0.006% per cycle over 500 cycles at 0.5 C	[168]
$LiNi_xCo_yMn_zO_2$	Carbonization	Li-NCM oxide	Li-S batteries	700 mAh g^{-1} after 300 cycles	[169]
Graphite	Thermal treatment	NG-Bat (nitrogen-doped graphene)	Fuel cells	ORR onset potential reached 0.867 V, and had a better catalytic long-term performance	[173]
Graphite	Acidic leaching and calcining	Graphene nanosheets	Fuel cells	Onset potential of 0.91 V, half-wave potential of 0.81 V and limiting current density of 6.6 mA cm^{-2}, with a better catalytic stability than Pt/C	[177]
Graphite	Pyrolysis with polyaniline and iron salt	Fe-N-doped carbon	Fuel cells	Onset potential of 0.91 V and half-wave potential of 0.8 V, and had a better ORR catalytic stability than commercial 20%Pt/C	[178]
$LiCoO_2$ and $LiFePO_4$	Acidic leaching and calcining	CoFe/C	Zinc-air batteries	Half-wave potential of 0.85 V, overpotential of 1.66 V at 10 mA cm^{-2}, and power density of 199.2 mW cm^{-2} with a long cycling stability of 350 h	[184]
$LiNi_{1-x-y}Mn_xCo_yO_2$	Acid leaching and radiative heating processes	NiMnCo-activated carbon (NiMnCo-AC)	Zinc-air batteries	High power density of 187.7 mW cm^{-2} and superior cycling stability of 200 h at 10 mA cm^{-2}	[185]
$LiNi_xCo_yMn_zO_2$	Alkali leaching and calcining	Ni-Co-Mn oxides	Zinc-air batteries	Energy efficiency reached 75%	[186]
$LiCoO_2$	High temperature shock process	Carbon layer/Co_3O_4 composite	Zinc-air batteries	ORR onset potential reached 0.90 V and high power density of 131 mW cm^{-2}	[187]

First, converting spent LIB materials into catalysts has limitations in element types. The possibility of such conversion is limited by the original material elements (mainly including Li, Ni, Co, Mn, Fe, Al, Cu, O, P, and C) and structure. Although it can be modified by introducing other elements (such as N), this path is less applicable than direct synthesis. The resynthesis after precise extraction of each element can solve this problem, but this approach sacrifices the structural advantages of the spent LIB materials that are conductive to catalysis. Second, given that the catalyst dosage is much smaller than that of electrode materials in production and application, converting all the spent materials into catalysts is impractical. It is more suitable for treating those materials that are difficult to handle with conventional recycling strategies such as hydrometallurgy or pyrometallurgy. In addition, the "LIB to LIB" path, namely the resynthesis or regeneration of spent LIB materials into new LIB materials, can be repeated to form a closed loop by adjusting the recycling process within a reasonable range. In terms of the path of "LIBs to catalysts", after the catalysts recycled from spent LIB materials are deactivated, advancing new technologies to restore their activity or recycle them for other purposes is necessary, and the recycling of such catalysts presents a scarcity of research.

8 Conclusion and Perspective

The rise of electronic and electric devices such as mobile phones and EVs has increased the demand for rechargeable batteries with high capacity, suitable size, and weight. Among different rechargeable batteries, LIBs stand out due to their high energy density, low self-discharge, and prolonged lifespan, displaying wide application in various fields. In the context of falling global crude oil reserves, people pay more attention to environmental issues. EVs will gradually replace gasoline vehicles, boosting the demand for LIBs. It has been expected that each batch of power batteries will usher in the renewal, leading to a mass of end-of-life LIBs for recycling. Researchers have been dedicated to developing battery recycling technologies in response to this situation. To satisfy growing global energy demand and accomplish sustainable energy development goals, using the recovered products of spent LIBs in preparing catalysts in academia and industry is promising.

With appropriate recycling approaches, the waste materials can be recovered as metals and compounds or reutilized as functional materials such as catalysts for various applications. Regarding the application of spent LIB recycling in catalysis, this paper reviewed it in three main aspects: pollutant degradation catalysis, substance conversion catalysis, and direct application of spent LIB recycled products in new battery-related catalysis. Some research studies demonstrated that the catalytic capacity of the recovered cathode or anode materials even exceeded that of the commercial catalysts after modification (Table 4). Due to chemical reactions and physical changes in the battery charging and discharging process, some recycled products without modification could also have good catalytic performance for specific catalytic reactions. The application of battery recycling in catalysis is insightful. In other words, it can be extended to other areas, and the recovered materials' properties in specific aspects may be better than the traditional ones with measurable treatments.

The high-value recovery of spent LIBs for catalyst application is promising, and more breakthroughs in this area will arise. Research on LIB recycling is still limited, leaving many crucial issues unsolved. The remaining challenges and perspectives are summarized as follows (Fig. 11):

I. Development of recycling strategies for the whole LIB components. Currently, the research of LIB recycling mainly involves the regeneration and utilization of cathode materials using pyrometallurgy, hydrometallurgy, and direct recovery, including $LiCoO_2$, $LiMn_2O_4$, LFP, and $LiNi_xCo_yMn_zO_2$. The recycling of other components of the batteries, such as anode, electrolyte, separator, and current collector, is neglected and should be highlighted due to their unique values. The high-purity Cu and Al foils used in current collectors can be recycled and used in metallurgy to prepare alloy materials or reused for current collector productions. For the SG anode with good carbon matrix structure and electrical conductivity, its recycling for advanced carbon material synthesis is promising. The electrolyte contains considerable organic solvents, inorganic salts, and additives, which can be recycled to synthesize chemicals and are environmentally friendly. In this sense, developing advanced technologies to recycle whole LIB components is a research direction and an important way to realize a circular economy.

II. National policy and industrial standard formulation. After decades of development, the LIB industry has mature laws and regulations to ensure its reasonable operation. In contrast, the spent LIB recycling market is at a disruption stage due to the lack of relevant management and industry standards. For this reason, a unified standard formulation is necessary to ensure the sustainable development of the battery recycling industry. The corresponding policies and regulations should include the following principles: governments should create incentives and subsidies to encourage sustainable recycling, and the industry should set purity standards for recycled products and strict requirements for the treatment of pollutants generated in this process.

III. Recycling process customization for various cathode materials. Cathode materials have different cost values due to the difference in their preparation technologies and composition. Typical recycling technologies, such as pyrometallurgy and hydrometallurgy, use high-temperature calcination and acid leaching. They are characterized by huge energy consumption and high environmental risk, making them unsuitable for all kinds of cathode materials. For example, the economic benefits of either pyrometallurgical or hydrometallurgical recovery are low since its processing spending is higher than the material value (even the affordable $LiFePO_4$ and $LiMn_2O_4$). Additionally, cathode materials contain high-value metals (e.g., nickel and cobalt), which can be economically feasible using typical processes. Developing direct recovery technology is novel to reduce cycling costs. Hence, cost control should be an important index of battery recycling technology, and this cost should be lower than the value of the recycled products to promote the development of this technology. Customizing different recycling processes for various cathodes is an important prerequisite for intelligent recycling in the future.

IV. Application expansion of recycled materials. The mainstream application of the spent LIBs after recovery is the repreparation of electrode materials. Despite the insightful catalysis usage reviewed in this article, the application of battery-recycled products is limited. With the development of battery recycling technology, recycled products should be applied to more fields, such as medical apparatus and instruments, intelligent manufacturing, ironmaking, and steelmaking.

V. Technology development beyond LIBs. LIBs occupy the major power battery and energy storage battery markets, and developing sustainable recycling methods is effective in solving the environmental risks and resource waste due to substantial end-of-life LIBs. In response to resource shortages and growing market demands, advanced energy storage technologies can be developed, including sodium/potassium-ion, lithium/sodium-sulfur, and metal-air batteries. These electrochemical energy storage devices can be employed in combination with LIBs or alone. Furthermore, they present diversified, safe, and green alternatives for energy supply and promote the sustainable development of new energy.

VI. Development of non-structurally destructive and green lithium extraction methods. In addition to Ni, Co, Mn, and Fe elements commonly used for catalysts, the Li element in spent LIBs is also valuable. Most works focused on recycling the spent LIBs as catalysts and neglected the recycling of Li. Many effective methods for Li extraction in academia and industry, such as hydrometallurgy, are available. Researchers used acidic solvent systems, such as inorganic acid (H_2SO_4, HCl, etc.) or organic acid ($H_2C_2O_4$, CH_3COOH, etc.), to convert Ni, Co, and Mn into precipitates, allowing for the retention of Li in the solution. Subsequently, Na_2CO_3 and Na_3PO_4 solutions were added to precipitate Li salt. The wetting process requires strong acid or alkaline leaching agents to dissolve the components completely, and the raw materials input is large, potentially causing environmental pollution due to the waste liquid after leaching. The deep eutectic solvent (DES) leaching method for spent LIB recycling has attracted wide attention. DES has green, highly efficient, and recyclable attributes. DES systems such as the betaine hydrochloride-ethylene glycol system and choline chloride-urea have been introduced to replace the traditional leaching process, featuring high environmental friendliness and lower costs. Despite these strengths, the hydrometallurgy process will destroy the original composition and structure of battery electrode materials, inactivating the catalyst activity. It is worth investigating how to extract lithium from the spent cathode materials without damaging its structure, so that it can then be converted into a catalyst.

VII. Catalyst service life extension. Long service life is an important indicator for the commercial application of catalysts. During usage, the efficiency of catalysts decreases, retarding the catalytic process. This leads to decreased economic benefits of the device's working process and even operation failure. Catalyst replacement is crucial to ensuring that the economic benefits of the components are higher than the rated value. This tedious process will be costly and time-consuming. If not handled properly, the heavy metal ions in the LIB-derived catalysts can also cause environmental pollution. The decline of catalyst life is attributed to many reasons, mainly including catalyst poisoning by poor service environment. The high-temperature thermal effect expands the particle size of the active components in the catalyst, reducing the specific surface area. Subsequently, the impurities in the reaction raw materials cover the active sites on the catalyst surface. The catalyst lifespan depends on the guaranteed service life under normal operating conditions proposed by the manufacturer, the actual operating conditions, and usage methods.

Table 4 Catalysts recycled from spent LIB materials that perform better than industrial ones and corresponding performance

Application	Spent material types	Catalyst	Industrial catalyst	Performance	References
OER	$LiCoO_2$	Ultra-thin CoOOH nanosheets	RuO_2	Recycled: mass activity of 99.2 A g^{-1} at 1.6 V, overpotential of 305 mV after 65 h at 10 mA cm^{-2}; RuO_2: 45.1 A g^{-1} at 1.6 V, overpotential of 367 mV after 23 h at 10 mA cm^{-2}	[129]
OER	$LiNi_xCo_yMn_zO_2$	NiCoMn layered triple hydroxides	RuO_2	Recycled: affords 10 mA cm^{-2} at an applied potential of 1.58 V; RuO_2: affords 10 mA cm^{-2} at an applied potential of 1.62 V	[135]
OER	$LiNi_xCo_yMn_zO_2$	NiCoMnBs	RuO_2	Recycled: affords 46.99 mA cm^{-2} at an applied potential of 1.53 V, overpotential of 318 mV at 100 mA cm^{-2}; RuO_2: affords 20.32 mA cm^{-2} at an applied potential of 1.53 V, overpotential of 501 mV at 100 mA cm^{-2}	[136]
OER	$LiNi_xCo_yMn_zO_2$	$Ni_{0.5}Mn_{0.3}Co_{0.2}(OH)_2$ nanoparticles	IrO_2	Recycled: overpotential of 0.28 mV at 10 mA cm^{-2}; IrO_2: overpotential of 0.29 mV at 10 mA cm^{-2}	[137]
ORR	Graphite	Nitrogen-doped graphene	Nitrogen-doped graphene	Recycled: onset potential of 0.867 V for ORR, current reduction of 16.5% after the injection of 3 mol L^{-1} methanol into the O_2-saturated, 0.1 mol L^{-1} KOH solution; Commercial nitrogen-doped graphene: onset potential of 0.797 V for ORR, current reduction of 33% after the injection of 3 mol L^{-1} methanol into the O_2-saturated, 0.1 mol L^{-1} KOH solution	[173]
ORR	Graphite	Graphene nanosheets	Pt/C	Recycled: onset potential of 0.91 V for ORR half-wave potential of 0.81 V, limiting current density of 6.6 mA cm^{-2}, activity degradation of 7% in the durability test; Pt/C: performance in onset potential, half-wave potential, limiting current density are worse than the recycled one (no specific data), activity degradation of 20% in the durability test	[177]
ORR	Graphite	Fe-N-doped carbon	20%Pt/C	Recycled: half-wave potential of 0.80 V, maintained an initial current of 93% after running at a fixed potential of 0.38 V for 36 000 s; 20%Pt/C: half-wave potential of 0.79 V, maintained an initial current of 82% after running at a fixed potential of 0.38 V for 36 000 s	[178]

In application, besides costs, the downtime-induced losses of catalysts due to catalyst replacement should also be considered. Some catalyst can be regenerated after being poisoned. However, after multiple regenerations, irreversible changes occur, leading to lower efficiency of the catalytic process than the specified indicators, indicating catalyst degradation. When designing and preparing catalysts from waste LIB electrode materials, specific application scenarios and possible influencing factors of the catalyst should be underscored, and targeted design should be carried

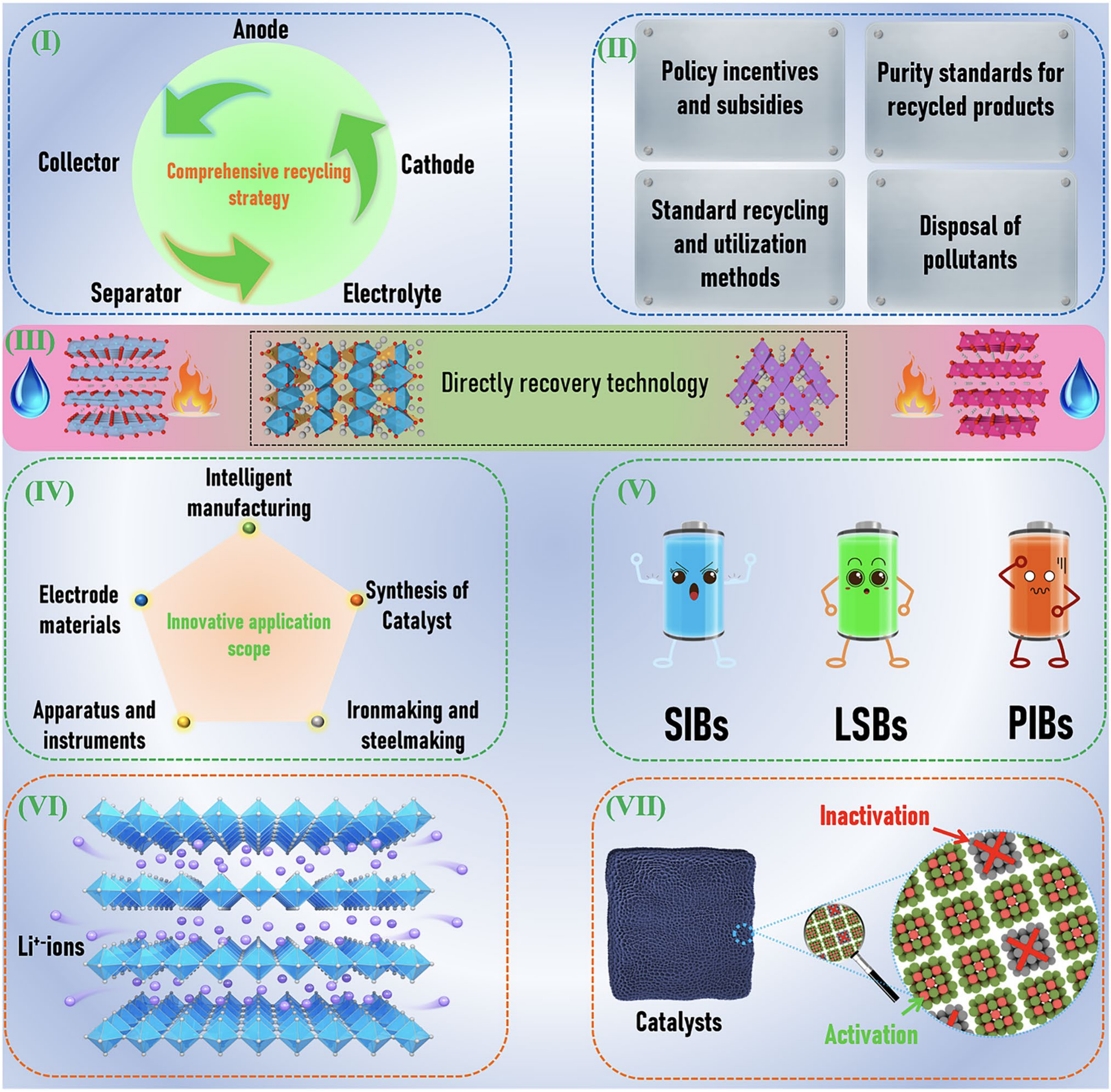

Fig. 11 Proposed perspectives for spent LIB recycling in the future

Fig. 12 Schematics of toward recycling-oriented configuration and closed-loop design rule in practical spent LIB recycling

out to maximize the service life of the synthesized catalyst.

VIII. Toward recycling-oriented configuration design. Considering resource scarcity and environmental pollution, spent LIB recycling has gained increasing attention. The model, specification, size, and composition of commercial LIBs have huge differences, hindering the development of large-scale recycling equipment as well as economical and sustainable battery recycling systems. In this sense, the upstream battery recycling industry should develop new types of batteries featuring easy recycling. Specifically, battery production should follow the closed-loop design rules to boost recycling performance and critically assess the possible recycling-oriented design of battery configuration, electrodes, and materials for recycling (Fig. 12).

Acknowledgements This work was supported by the National Natural Science Foundation of China (No. 52072205), Joint Funds of the National Natural Science Foundation of China (U21A20174), Shen-zhen Science and Technology Program (KQTD20210811090112002), Guangdong Innovative and Entrepreneurial Research Team Program (2021ZT09L197), Start-up Fund, and Interdisciplinary Research and Innovation Fund of Tsinghua Shenzhen International Graduate School, the Tsinghua Shenzhen International Graduate School-Shenzhen Pengrui Young Faculty Program of Shenzhen Pengrui Foundation (SZPR2023007), and the Natural Science Foundation of Sichuan Province (2023NSFSC1128) and Project of Southwest University of Science and Technology (20zx7142).

Author contributions R.S., B.W., and D.T. contributed equally to this review. Conceptualization: G.Z.; Formal analysis and investigation: R.S., B.W., and D.T.; Writing—original draft preparation: R.S., B.W., and D.T.; Writing—review and editing: X.W. and G.Z.; Funding acquisition: X.W. and G.Z.; Supervision: X.W. and G.Z.

Declarations

Conflict of interest The authors declare no conflict of interest.

Ethical approval All authors certify that they have no affiliations with or involvement in any organization or entity with any financial interest or non-financial interest in the subject matter or materials discussed in this manuscript.

References

1. Goodenough, J.B., Park, K.S.: The Li-ion rechargeable battery: a perspective. J. Am. Chem. Soc. **135**, 1167–1176 (2013). https://doi.org/10.1021/ja3091438
2. Choi, S.H., Kwon, T.W., Coskun, A., et al.: Highly elastic binders integrating polyrotaxanes for silicon microparticle anodes in lithium ion batteries. Science **357**, 279–283 (2017). https://doi.org/10.1126/science.aal4373
3. Ding, Y.L., Cano, Z.P., Yu, A.P., et al.: Automotive Li-ion batteries: current status and future perspectives. Electrochem. Energy Rev. **2**, 1–28 (2019). https://doi.org/10.1007/s41918-018-0022-z
4. Roy, J.J., Rarotra, S., Krikstolaityte, V., et al.: Green recycling methods to treat lithium-ion batteries e-waste: a circular approach to sustainability. Adv. Mater. **34**, 2103346 (2022). https://doi.org/10.1002/adma.202103346
5. Wu, Z.S., Ren, W.R., Wen, L.W., et al.: Graphene anchored with Co_3O_4 nanoparticles as anode of lithium ion batteries with enhanced reversible capacity and cyclic performance. ACS Nano **4**, 3187–3194 (2010). https://doi.org/10.1021/nn100740x
6. Ye, Y.S., Chou, L.Y., Liu, Y., et al.: Ultralight and fire-extinguishing current collectors for high-energy and high-safety lithium-ion batteries. Nat. Energy **5**, 786–793 (2020). https://doi.org/10.1038/s41560-020-00702-8
7. Shen, Y.F.: Recycling cathode materials of spent lithium-ion batteries for advanced catalysts production. J. Power Sources **528**, 231220 (2022). https://doi.org/10.1016/j.jpowsour.2022.231220
8. Pender, J.P., Jha, G., Youn, D.H., et al.: Electrode degradation in lithium-ion batteries. ACS Nano **14**, 1243–1295 (2020). https://doi.org/10.1021/acsnano.9b04365
9. Xiong, R., Pan, Y., Shen, W.X., et al.: Lithium-ion battery aging mechanisms and diagnosis method for automotive applications: recent advances and perspectives. Renew. Sust. Energ. Rev. **131**, 110048 (2020). https://doi.org/10.1016/j.rser.2020.110048
10. Keil, P., Schuster, S.F., Wilhelm, J., et al.: Calendar aging of lithium-ion batteries. J. Electrochem. Soc. **163**, A1872–A1880 (2016). https://doi.org/10.1149/2.0411609jes
11. Yu, W.H., Guo, Y., Xu, S.L., et al.: Comprehensive recycling of lithium-ion batteries: fundamentals, pretreatment, and perspectives. Energy Storage Mater. **54**, 172–220 (2023). https://doi.org/10.1016/j.ensm.2022.10.033
12. Harper, G., Sommerville, R., Kendrick, E., et al.: Recycling lithium-ion batteries from electric vehicles. Nature **575**, 75–86 (2019). https://doi.org/10.1038/s41586-019-1682-5
13. Ciez, R.E., Whitacre, J.F.: Examining different recycling processes for lithium-ion batteries. Nat. Sustain. **2**, 148–156 (2019). https://doi.org/10.1038/s41893-019-0222-5
14. Dunn, J.B., Gaines, L., Kelly, J.C., et al.: The significance of Li-ion batteries in electric vehicle life-cycle energy and emissions and recycling's role in its reduction. Energy Environ. Sci. **8**, 158–168 (2015). https://doi.org/10.1039/C4EE03029J
15. Wang, P., Guo, Y.G., Guan, J., et al.: Applications of spent lithium battery electrode materials in catalytic decontamination: a review. Catalysts **13**, 189 (2023). https://doi.org/10.3390/catal13010189
16. Zheng, M.T., Salim, H., Liu, T.F., et al.: Intelligence-assisted predesign for the sustainable recycling of lithium-ion batteries and beyond. Energy Environ. Sci. **14**, 5801–5815 (2021). https://doi.org/10.1039/D1EE01812D
17. Yang, Y., Okonkwo, E.G., Huang, G.Y., et al.: On the sustainability of lithium ion battery industry: a review and perspective. Energy Storage Mater. **36**, 186–212 (2021). https://doi.org/10.1016/j.ensm.2020.12.019
18. Bejigo, K.S., Natarajan, S., Bhunia, K., et al.: Recycling of value-added products from spent lithium-ion batteries for oxygen reduction and methanol oxidation reactions. J. Clean. Prod. **384**, 135520 (2023). https://doi.org/10.1016/j.jclepro.2022.135520
19. Ji, H.C., Wang, J.X., Ma, J., et al.: Fundamentals, status and challenges of direct recycling technologies for lithium ion batteries. Chem. Soc. Rev. **52**, 8194–8244 (2023). https://doi.org/10.1039/D3CS00254C
20. Wang, X.T., Gu, Z.Y., Ang, E.H.X., et al.: Prospects for managing end-of-life lithium-ion batteries: present and future. Interdiscip. Mater. **1**, 417–433 (2022). https://doi.org/10.1002/idm2.12041
21. Niu, B., Xiao, J.F., Xu, Z.M.: Recycling spent $LiCoO_2$ battery as a high-efficient lithium-doped graphitic carbon nitride/Co_3O_4 composite photocatalyst and its synergistic photocatalytic mechanism. Energy Environ. Mater. **6**, e12312 (2023). https://doi.org/10.1002/eem2.12312
22. Or, T., Gourley, S.W.D., Kaliyappan, K., et al.: Recycling of mixed cathode lithium-ion batteries for electric vehicles: current status and future outlook. Carbon Energy **2**, 6–43 (2020). https://doi.org/10.1002/cey2.29
23. Zhang, X.X., Li, L., Fan, E., et al.: Toward sustainable and systematic recycling of spent rechargeable batteries. Chem. Soc. Rev. **47**, 7239–7302 (2018). https://doi.org/10.1039/C8CS00297E
24. Fan, E.S., Li, L., Wang, Z.P., et al.: Sustainable recycling technology for Li-ion batteries and beyond: challenges and future prospects. Chem. Rev. **120**, 7020–7063 (2020). https://doi.org/10.1021/acs.chemrev.9b00535
25. Chen, M.Y., Ma, X.T., Chen, B., et al.: Recycling end-of-life electric vehicle lithium-ion batteries. Joule **3**, 2622–2646 (2019). https://doi.org/10.1016/j.joule.2019.09.014
26. Wang, J.X., Liang, Z., Zhao, Y., et al.: Direct conversion of degraded $LiCoO_2$ cathode materials into high-performance $LiCoO_2$: a closed-loop green recycling strategy for spent lithium-ion batteries. Energy Storage Mater. **45**, 768–776 (2022). https://doi.org/10.1016/j.ensm.2021.12.013
27. Wang, J.X., Ma, J., Jia, K., et al.: Efficient extraction of lithium from anode for direct regeneration of cathode materials of spent Li-ion batteries. ACS Energy Lett. **7**, 2816–2824 (2022). https://doi.org/10.1021/acsenergylett.2c01539
28. Zhao, Y., Kang, Y.Q., Fan, M.C., et al.: Precise separation of spent lithium-ion cells in water without discharging for recycling. Energy Storage Mater. **45**, 1092–1099 (2022). https://doi.org/10.1016/j.ensm.2021.11.005
29. Lin, J., Fan, E.S., Zhang, X.D., et al.: Sustainable upcycling of spent lithium-ion batteries cathode materials: stabilization by in situ Li/Mn disorder. Adv. Energy Mater. **12**, 2201174 (2022). https://doi.org/10.1002/aenm.202201174
30. Fan, M., Meng, Q.H., Chang, X., et al.: In situ electrochemical regeneration of degraded $LiFePO_4$ electrode with functionalized

prelithiation separator. Adv. Energy Mater. **12**, 2103630 (2022). https://doi.org/10.1002/aenm.202103630

31. Jin, Y.C., Zhang, T., Zhang, M.D.: Advances in intelligent regeneration of cathode materials for sustainable lithium-ion batteries. Adv. Energy Mater. **12**, 2201526 (2022). https://doi.org/10.1002/aenm.202201526
32. Mao, J.F., Ye, C., Zhang, S.L., et al.: Toward practical lithium-ion battery recycling: adding value, tackling circularity and recycling-oriented design. Energy Environ. Sci. **15**, 2732–2752 (2022). https://doi.org/10.1039/D2EE00162D
33. Wu, J.W., Zheng, M.T., Liu, T.F., et al.: Direct recovery: a sustainable recycling technology for spent lithium-ion battery. Energy Storage Mater. **54**, 120–134 (2023). https://doi.org/10.1016/j.ensm.2022.09.029
34. Murdock, B.E., Toghill, K.E., Tapia-Ruiz, N.: A perspective on the sustainability of cathode materials used in lithium-ion batteries. Adv. Energy Mater. **11**, 2102028 (2021). https://doi.org/10.1002/aenm.202102028
35. Ma, J., Wang, J.X., Jia, K., et al.: Adaptable eutectic salt for the direct recycling of highly degraded layer cathodes. J. Am. Chem. Soc. **144**, 20306–20314 (2022). https://doi.org/10.1021/jacs.2c07860
36. Jia, K., Ma, J., Wang, J.X., et al.: Long-life regenerated $LiFePO_4$ from spent cathode by elevating the d-band center of Fe. Adv. Mater. **35**, 2208034 (2023). https://doi.org/10.1002/adma.202370034
37. Tran, M.K., Rodrigues, M.T.F., Kato, K., et al.: Deep eutectic solvents for cathode recycling of Li-ion batteries. Nat. Energy **4**, 339–345 (2019). https://doi.org/10.1038/s41560-019-0368-4
38. Xu, P.P., Dai, Q., Gao, H.P., et al.: Efficient direct recycling of lithium-ion battery cathodes by targeted healing. Joule **4**, 2609–2626 (2020). https://doi.org/10.1016/j.joule.2020.10.008
39. Cao, Z.Q., Zheng, X.H., Cao, H.B., et al.: Efficient reuse of anode scrap from lithium-ion batteries as cathode for pollutant degradation in electro-Fenton process: role of different recovery processes. Chem. Eng. J. **337**, 256–264 (2018). https://doi.org/10.1016/j.cej.2017.12.104
40. Santana, I.L., Moreira, T.F.M., Lelis, M.F.F., et al.: Photocatalytic properties of Co_3O_4/$LiCoO_2$ recycled from spent lithium-ion batteries using citric acid as leaching agent. Mater. Chem. Phys. **190**, 38–44 (2017). https://doi.org/10.1016/j.matchemphys.2017.01.003
41. Garole, D.J., Hossain, R., Garole, V.J., et al.: Recycle, recover and repurpose strategy of spent Li-ion batteries and catalysts: current status and future opportunities. Chemsuschem **13**, 3079–3100 (2020). https://doi.org/10.1002/cssc.201903213
42. Zheng, X.R., Zhao, X., Lu, J.D., et al.: Regeneration of spent cathodes of Li-ion batteries into multifunctional electrodes for overall water splitting and rechargeable Zn-air batteries by ultrafast carbothermal shock. Sci. China Mater. **65**, 2393–2400 (2022). https://doi.org/10.1007/s40843-021-1984-8
43. Bian, H.D., Wu, W.B., Zhu, Y.Y., et al.: Waste to treasure: regeneration of porous Co-based catalysts from spent $LiCoO_2$ cathode materials for an efficient oxygen evolution reaction. ACS Sustain. Chem. Eng. **11**, 670–678 (2023). https://doi.org/10.1021/acssuschemeng.2c05534
44. Dąbrowska, A., Urbańska, W., Warczak, M., et al.: Battery powder as a source of novel graphene nanocarbons. Phys. Status Solidi B **259**, 2100588 (2022). https://doi.org/10.1002/pssb.202100588
45. Jena, K.K., Mayyas, A.T., Mohanty, B., et al.: Recycling of electrode materials from spent lithium-ion batteries to develop graphene nanosheets and graphene-molybdenum disulfide nanohybrid: environmental benefits, analysis of supercapacitor performance, and influence of density functional theory calculations. Energy Fuels **36**, 2159–2170 (2022). https://doi.org/10.1021/acs.energyfuels.1c03789
46. Wu, X.X., Ji, G.J., Wang, J.X., et al.: Toward sustainable all solid-state Li-metal batteries: perspectives on battery technology and recycling processes. Adv. Mater. **35**, 2301540 (2023). https://doi.org/10.1002/adma.202301540
47. Fan, M., Chang, X., Meng, Q.H., et al.: Progress in the sustainable recycling of spent lithium-ion batteries. SusMat **1**, 241–254 (2021). https://doi.org/10.1002/sus2.16
48. Wu, X.X., Ma, J., Wang, J.X., et al.: Progress, key issues, and future prospects for Li-ion battery recycling. Glob Chall **6**, 2200067 (2022). https://doi.org/10.1002/gch2.202200067
49. Fang, Z., Duan, Q.L., Peng, Q.K., et al.: Comparative study of chemical discharge strategy to pretreat spent lithium-ion batteries for safe, efficient, and environmentally friendly recycling. J. Cleaner Prod. **359**, 132116 (2022). https://doi.org/10.1016/j.jclepro.2022.132116
50. Wu, L.X., Zhang, F.S., He, K., et al.: Avoiding thermal runaway during spent lithium-ion battery recycling: a comprehensive assessment and a new approach for battery discharge. J. Cleaner Prod. **380**, 135045 (2022). https://doi.org/10.1016/j.jclepro.2022.135045
51. Waldmann, T., Iturrondobeitia, A., Kasper, M., et al.: Review—post-mortem analysis of aged lithium-ion batteries: disassembly methodology and physico-chemical analysis techniques. J. Electrochem. Soc. **163**, A2149–A2164 (2016). https://doi.org/10.1149/2.1211609jes
52. Dorella, G., Mansur, M.B.: A study of the separation of cobalt from spent Li-ion battery residues. J. Power Sources **170**, 210–215 (2007). https://doi.org/10.1016/j.jpowsour.2007.04.025
53. Chen, L., Tang, X.C., Zhang, Y., et al.: Process for the recovery of cobalt oxalate from spent lithium-ion batteries. Hydrometallurgy **108**, 80–86 (2011). https://doi.org/10.1016/j.hydromet.2011.02.010
54. Grützke, M., Kraft, V., Hoffmann, B., et al.: Aging investigations of a lithium-ion battery electrolyte from a field-tested hybrid electric vehicle. J. Power Sources **273**, 83–88 (2015). https://doi.org/10.1016/j.jpowsour.2014.09.064
55. Lv, W.G., Wang, Z.H., Cao, H.B., et al.: A critical review and analysis on the recycling of spent lithium-ion batteries. ACS Sustain. Chem. Eng. **6**, 1504–1521 (2018). https://doi.org/10.1021/acssuschemeng.7b03811
56. Makuza, B., Tian, Q.H., Guo, X.Y., et al.: Pyrometallurgical options for recycling spent lithium-ion batteries: a comprehensive review. J. Power Sources **491**, 229622 (2021). https://doi.org/10.1016/j.jpowsour.2021.229622
57. Piątek, J., Afyon, S., Budnyak, T.M., et al.: Sustainable Li-ion batteries: chemistry and recycling. Adv. Energy Mater. **11**, 2003456 (2020). https://doi.org/10.1002/aenm.202003456
58. Du, K.D., Ang, E.H.X., Wu, X.L., et al.: Progresses in sustainable recycling technology of spent lithium-ion batteries. Energy Environ. Mater. **5**, 1012–1036 (2022). https://doi.org/10.1002/eem2.12271
59. Qu, G.R., Wei, Y.G., Liu, C.P., et al.: Efficient separation and recovery of lithium through volatilization in the recycling process of spent lithium-ion batteries. Waste Manag. **150**, 66–74 (2022). https://doi.org/10.1016/j.wasman.2022.06.039
60. Zhang, A., Liang, Y.X., Zhang, H., et al.: Doping regulation in transition metal compounds for electrocatalysis. Chem. Soc. Rev. **50**, 9817–9844 (2021). https://doi.org/10.1039/D1CS00330E
61. Xin, Y., Li, S.H., Qian, Y.Y., et al.: High-entropy alloys as a platform for catalysis: progress, challenges, and opportunities. ACS Catal. **10**, 11280–11306 (2020). https://doi.org/10.1021/acscatal.0c03617

62. Boutin, E., Merakeb, L., Ma, B., et al.: Molecular catalysis of CO_2 reduction: recent advances and perspectives in electrochemical and light-driven processes with selected Fe, Ni and Co aza macrocyclic and polypyridine complexes. Chem. Soc. Rev. **49**, 5772–5809 (2020). https://doi.org/10.1039/D0CS00218F
63. Yang, Y., Yang, Y., Pei, Z., et al.: Recent progress of carbon-supported single-atom catalysts for energy conversion and storage. Matter **3**, 1442–1476 (2020). https://doi.org/10.1016/j.matt.2020.07.032
64. Zeng, H., Deng, L., Zhang, H., et al.: Development of oxygen vacancies enriched CoAl hydroxide@hydroxysulfide hollow flowers for peroxymonosulfate activation: a highly efficient singlet oxygen-dominated oxidation process for sulfamethoxazole degradation. J. Hazard. Mater. **400**, 123297 (2020). https://doi.org/10.1016/j.jhazmat.2020.123297
65. Windsor, F.M., Durance, I., Horton, A.A., et al.: A catchment-scale perspective of plastic pollution. Glob. Change Biol. **25**, 1207–1221 (2019). https://doi.org/10.1111/gcb.14572
66. Miao, Y.P., Liu, L.L., Zhang, Y.P., et al.: An overview of global power lithium-ion batteries and associated critical metal recycling. J. Hazard. Mater. **425**, 127900 (2022). https://doi.org/10.1016/j.jhazmat.2021.127900
67. Hodges, B.C., Cates, E.L., Kim, J.H.: Challenges and prospects of advanced oxidation water treatment processes using catalytic nanomaterials. Nat. Nanotechnol. **13**, 642–650 (2018). https://doi.org/10.1038/s41565-018-0216-x
68. Moreira, F.C., Boaventura, R.A.R., Brillas, E., et al.: Electrochemical advanced oxidation processes: a review on their application to synthetic and real wastewaters. Appl. Catal. B-Environ. **202**, 217–261 (2017). https://doi.org/10.1016/j.apcatb.2016.08.037
69. Martinez-Huitle, C.A., Rodrigo, M.A., Sires, I., et al.: Single and coupled electrochemical processes and reactors for the abatement of organic water pollutants: a critical review. Chem. Rev. **115**, 13362–13407 (2015). https://doi.org/10.1021/acs.chemrev.5b00361
70. Dong, G.H., Chen, B., Liu, B., et al.: Advanced oxidation processes in microreactors for water and wastewater treatment: development, challenges, and opportunities. Water Res. **211**, 118047 (2022). https://doi.org/10.1016/j.watres.2022.118047
71. Wang, S.Z., Wang, J.L.: Electron beam technology coupled to fenton oxidation for advanced treatment of dyeing wastewater: from laboratory to full application. ACS EST Water **2**, 852–862 (2022). https://doi.org/10.1021/acsestwater.2c00040
72. Fenton, H.J.H.: Oxidation of tartaric acid in presence of iron. J. Chem. Soc. **65**, 899–910 (1894). https://doi.org/10.1039/CT8946500899
73. He, J., Yang, X.F., Men, B., et al.: Interfacial mechanisms of heterogeneous Fenton reactions catalyzed by iron-based materials: a review. J. Environ. Sci. **39**, 97–109 (2016). https://doi.org/10.1016/j.jes.2015.12.003
74. Yang, Z.C., Qian, J.S., Yu, A.Q., et al.: Singlet oxygen mediated iron-based Fenton-like catalysis under nanoconfinement. Proc. Natl. Acad. Sci. **116**, 6659–6664 (2019). https://doi.org/10.1073/pnas.1819382116
75. Goncalves, M.C., Garcia, E.M., Taroco, H.A., et al.: Chemical recycling of cell phone Li-ion batteries: application in environmental remediation. Waste Manage. **40**, 144–150 (2015). https://doi.org/10.1016/j.wasman.2015.02.014
76. Guo, J., Zhang, J., Chen, C., et al.: Rapid photodegradation of methyl orange by oxalic acid assisted with cathode material of lithium ion batteries $LiFePO_4$. J. Taiwan Inst. Chem. Eng. **62**, 187–191 (2016). https://doi.org/10.1016/j.wasman.2015.02.014
77. Xu, L., Chen, C., Huo, J.B., et al.: Iron hydroxyphosphate composites derived from waste lithium-ion batteries for lead adsorption and fenton-like catalytic degradation of methylene blue. Environ. Technol. Innov. **16**, 100504 (2019). https://doi.org/10.1016/j.eti.2019.100504
78. Yang, L., Xi, G., Lou, T.J., et al.: Preparation and magnetic performance of $Co_{0.8}Fe_{2.2}O_4$ by a sol-gel method using cathode materials of spent Li-ion batteries. Ceram. Int. **42**, 1897–1902 (2016). https://doi.org/10.1016/j.ceramint.2015.09.159
79. Yao, L., Xi, Y.B., Xi, G., et al.: Synthesis of cobalt ferrite with enhanced magnetostriction properties by the sol-gel-hydrothermal route using spent Li-ion battery. J. Alloys Compd. **680**, 73–79 (2016). https://doi.org/10.1016/j.jallcom.2016.04.092
80. Al-Kahtani, A.A., Abou Taleb, M.F.: Photocatalytic degradation of Maxilon C.I. basic dye using CS/$CoFe_2O_4$/GONCs as a heterogeneous photo-Fenton catalyst prepared by gamma irradiation. J. Hazard. Mater. **309**, 10–19 (2016). https://doi.org/10.1016/j.jhazmat.2016.01.071
81. Moura, M.N., Barrada, R.V., Almeida, J.R., et al.: Synthesis, characterization and photocatalytic properties of nanostructured $CoFe_2O_4$ recycled from spent Li-ion batteries. Chemosphere **182**, 339–347 (2017). https://doi.org/10.1016/j.chemosphere.2017.05.036
82. Rocha, A.K.S., Magnago, L.B., Santos, J.J., et al.: Copper ferrite synthesis from spent Li-ion batteries for multifunctional application as catalyst in photo Fenton process and as electrochemical pseudocapacitor. Mater. Res. Bull. **113**, 231–240 (2019). https://doi.org/10.1016/j.materresbull.2019.02.007
83. Chen, X., Deng, F., Liu, X., et al.: Hydrothermal synthesis of MnO_2/Fe(0) composites from Li-ion battery cathodes for destructing sulfadiazine by photo-Fenton process. Sci. Total. Environ. **774**, 145776 (2021). https://doi.org/10.1016/j.scitotenv.2021.145776
84. Yi, C.X., Zhou, L.J., Wu, X.Q., et al.: Technology for recycling and regenerating graphite from spent lithium-ion batteries. Chin. J. Chem. Eng. **39**, 37–50 (2021). https://doi.org/10.1016/j.cjche.2021.09.014
85. Niu, B., Xiao, J.F., Xu, Z.M.: Advances and challenges in anode graphite recycling from spent lithium-ion batteries. J. Hazard. Mater. **439**, 129678 (2022). https://doi.org/10.1016/j.jhazmat.2022.129678
86. Guan, J., Li, Z.X., Chen, S., et al.: Zero-valent iron supported on expanded graphite from spent lithium-ion battery anodes and ferric chloride for the degradation of 4-chlorophenol in water. Chemosphere **290**, 133381 (2022). https://doi.org/10.1016/j.chemosphere.2021.133381
87. Jin, Q., Chen, Q., Shen, J.M., et al.: Development of Fe(II) system based on *N,N′*-dipicolinamide for the oxidative removal of 4-chlorophenol. J. Hazard. Mater. **354**, 206–214 (2018). https://doi.org/10.1016/j.jhazmat.2018.04.058
88. Yang, J., He, X.Q., Dai, J., et al.: Electron-transfer-dominated non-radical activation of peroxydisulfate for efficient removal of chlorophenol contaminants by one-pot synthesized nitrogen and sulfur codoped mesoporous carbon. Environ. Res. **194**, 110496 (2021). https://doi.org/10.1016/j.envres.2020.110496
89. Cai, C., Zhang, H., Zhong, X., et al.: Ultrasound enhanced heterogeneous activation of peroxymonosulfate by a bimetallic Fe-Co/SBA-15 catalyst for the degradation of orange II in water. J. Hazard. Mater. **283**, 70–79 (2015). https://doi.org/10.1016/j.jhazmat.2014.08.053
90. Pi, Y.Q., Gao, H.Q., Cao, Y.D., et al.: Cobalt ferrite supported on carbon nitride matrix prepared using waste battery materials as a peroxymonosulfate activator for the degradation of levofloxacin hydrochloride. Chem. Eng. J. **379**, 122377 (2020). https://doi.org/10.1016/j.cej.2019.122377

91. Zhao, Y.L., Yuan, X.Z., Jiang, L.B., et al.: Reutilization of cathode material from spent batteries as a heterogeneous catalyst to remove antibiotics in wastewater via peroxymonosulfate activation. Chem. Eng. J. **400**, 125903 (2020). https://doi.org/10.1016/j.cej.2020.125903
92. Zhao, Y.L., Wang, H., Li, X.D., et al.: Recovery of CuO/C catalyst from spent anode material in battery to activate peroxymonosulfate for refractory organic contaminants degradation. J. Hazard. Mater. **420**, 126552 (2021). https://doi.org/10.1016/j.jhazmat.2021.126552
93. Dang, S., Zhou, P.S., Shi, P.H., et al.: In situ aluminothermic reduction induced by mechanochemical activation enhances the ability of the spent $LiCoO_2$ cathode to activate peroxymonosulfate. ACS Sustain. Chem. Eng. **9**, 15375–15385 (2021). https://doi.org/10.1021/acssuschemeng.1c03583
94. Wang, X., Zhang, X.F., Dai, L., et al.: Recycling the cathode scrap of spent lithium-ion batteries as an easily recoverable peroxymonosulfate catalyst with enhanced catalytic performance. ACS Sustain. Chem. Eng. **8**, 11337–11347 (2020). https://doi.org/10.1021/acssuschemeng.0c03389
95. Zhang, X.Y., Wang, X., Chai, J., et al.: Construction of novel symmetric double Z-scheme $BiFeO_3/CuBi_2O_4/BaTiO_3$ photocatalyst with enhanced solar-light-driven photocatalytic performance for degradation of norfloxacin. Appl. Catal. B-Environ. **272**, 119017 (2020). https://doi.org/10.1016/j.apcatb.2020.119017
96. Lee, C.G., Javed, H., Zhang, D., et al.: Porous electrospun fibers embedding TiO_2 for adsorption and photocatalytic degradation of water pollutants. Environ. Sci. Technol. **52**, 4285–4293 (2018). https://doi.org/10.1021/acs.est.7b06508
97. An, J.H., Song, X.T., Wan, W.B., et al.: Kinetics of the photoelectron-transfer process characterized by real-time single-molecule fluorescence imaging on individual photocatalyst particles. ACS Catal. **11**, 6872–6882 (2021). https://doi.org/10.1021/acscatal.1c00983
98. Nascimento, M.A., Cruz, J.C., Rodrigues, G.D., et al.: Synthesis of polymetallic nanoparticles from spent lithium-ion batteries and application in the removal of reactive blue 4 dye. J. Clean. Prod. **202**, 264–272 (2018). https://doi.org/10.1016/j.jclepro.2018.08.118
99. Niu, B., Xiao, J.F., Xu, Z.M.: Utilizing spent Li-ion batteries to regulate the π-conjugated structure of g-C_3N_4: a win-win approach for waste recycling and highly active photocatalyst construction. J. Mater. Chem. A **9**, 472–481 (2021). https://doi.org/10.1039/D0TA10881B
100. Zhang, W.X., Liu, Z.P., Xu, C.J., et al.: Preparing graphene oxide-copper composite material from spent lithium ion batteries and catalytic performance analysis. Res. Chem. Intermed. **44**, 5075–5089 (2018). https://doi.org/10.1007/s11164-018-3410-4
101. Xu, Z.M., Wang, J., Sun, S.H., et al.: Simple route for graphite recycling from waste lithium-ion batteries to environmental functional materials. ACS Sustain. Chem. Eng. **10**, 13435–13443 (2022). https://doi.org/10.1021/acssuschemeng.2c04090
102. Xie, L.H., Liu, X.M., He, T., et al.: Metal-organic frameworks for the capture of trace aromatic volatile organic compounds. Chem **4**, 1911–1927 (2018). https://doi.org/10.1016/j.chempr.2018.05.017
103. Hakim, M., Broza, Y.Y., Barash, O., et al.: Volatile organic compounds of lung cancer and possible biochemical pathways. Chem. Rev. **112**, 5949–5966 (2012). https://doi.org/10.1021/cr300174a
104. Mellouki, A., Wallington, T.J., Chen, J.: Atmospheric chemistry of oxygenated volatile organic compounds: impacts on air quality and climate. Chem. Rev. **115**, 3984–4014 (2015). https://doi.org/10.1021/cr500549n
105. Kamal, M.S., Razzak, S.A., Hossain, M.M.: Catalytic oxidation of volatile organic compounds (VOCs): a review. Atmos. Environ. **140**, 117–134 (2016). https://doi.org/10.1016/j.atmosenv.2016.05.031
106. Huang, H.B., Xu, Y., Feng, Q.Y., et al.: Low temperature catalytic oxidation of volatile organic compounds: a review. Catal. Sci. Technol. **5**, 2649–2669 (2015). https://doi.org/10.1039/C4CY01733A
107. Liotta, L.F.: Catalytic oxidation of volatile organic compounds on supported noble metals. Appl. Catal. B-Environ. **100**, 403–412 (2010). https://doi.org/10.1016/j.apcatb.2010.08.023
108. Liao, Y.T., Jia, L., Chen, R.J., et al.: Charcoal-supported catalyst with enhanced thermal-stability for the catalytic combustion of volatile organic compounds. Appl. Catal. A-Gen **522**, 32–39 (2016). https://doi.org/10.1016/j.apcata.2016.04.028
109. Min, X., Guo, M.M., Liu, L.Z., et al.: Synthesis of MnO_2 derived from spent lithium-ion batteries via advanced oxidation and its application in VOCs oxidation. J. Hazard. Mater. **406**, 124743 (2021). https://doi.org/10.1016/j.jhazmat.2020.124743
110. Guo, M.M., Li, K., Liu, L.Z., et al.: Manganese-based multioxide derived from spent ternary lithium-ions batteries as high-efficient catalyst for VOCs oxidation. J. Hazard. Mater. **380**, 120905 (2019). https://doi.org/10.1016/j.jhazmat.2019.120905
111. Guo, M.M., Li, K., Zhang, H.B., et al.: Promotional removal of oxygenated VOC over manganese-based multi oxides from spent lithium-ions manganate batteries: Modification with Fe, Bi and Ce dopants. Sci. Total. Environ. **740**, 139951 (2020). https://doi.org/10.1016/j.scitotenv.2020.139951
112. Guo, M.M., Li, K., Liu, L.Z., et al.: Resource utilization of spent ternary lithium-ions batteries: synthesis of highly active manganese-based perovskite catalyst for toluene oxidation. J. Taiwan Inst. Chem. Eng. **102**, 268–275 (2019). https://doi.org/10.1016/j.jtice.2019.06.012
113. Guo, M.M., Li, K., Liu, L.Z., et al.: Insight into a sustainable application of spent lithium-ion cobaltate batteries: preparation of a cobalt-based oxide catalyst and its catalytic performance in toluene oxidation. Ind. Eng. Chem. Res. **59**, 194–204 (2019). https://doi.org/10.1021/acs.iecr.9b05298
114. Dai, T.C., Zhou, H., Liu, Y., et al.: Synergy of lithium, cobalt, and oxygen vacancies in lithium cobalt oxide for airborne benzene oxidation: a concept of reusing electronic wastes for air pollutant removal. ACS Sustain. Chem. Eng. **7**, 5072–5081 (2019). https://doi.org/10.1021/acssuschemeng.8b05894
115. Liu, Y., Gao, W., Zhan, J.J., et al.: One-pot synthesis of Ag-$H_3PW_{12}O_{40}$-$LiCoO_2$ composites for thermal oxidation of airborne benzene. Chem. Eng. J. **375**, 121956 (2019). https://doi.org/10.1016/j.cej.2019.121956
116. Liu, Y., Gao, W., Zhou, H., et al.: Highly reactive bulk lattice oxygen exposed by simple water treatment of $LiCoO_2$ for catalytic oxidation of airborne benzene. Mol. Catal. **492**, 111003 (2020). https://doi.org/10.1016/j.mcat.2020.111003
117. Suen, N.T., Hung, S.F., Quan, Q., et al.: Electrocatalysis for the oxygen evolution reaction: recent development and future perspectives. Chem. Soc. Rev. **46**, 337–365 (2017). https://doi.org/10.1039/C6CS00328A
118. Zhu, J., Hu, L.S., Zhao, P.X., et al.: Recent advances in electrocatalytic hydrogen evolution using nanoparticles. Chem. Rev. **120**, 851–918 (2020). https://doi.org/10.1021/acs.chemrev.9b00248
119. Mahmood, N., Yao, Y.D., Zhang, J.W., et al.: Electrocatalysts for hydrogen evolution in alkaline electrolytes: mechanisms, challenges, and prospective solutions. Adv. Sci. **5**, 1700464 (2018). https://doi.org/10.1002/advs.201700464
120. Natarajan, S., Anantharaj, S., Tayade, R.J., et al.: Recovered spinel $MnCo_2O_4$ from spent lithium-ion batteries for enhanced

electrocatalytic oxygen evolution in alkaline medium. Dalton Trans. **46**, 14382–14392 (2017). https://doi.org/10.1039/C7DT02613G
121. Natarajan, S., Krishnamoorthy, K., Sathyaseelan, A., et al.: A new route for the recycling of spent lithium-ion batteries towards advanced energy storage, conversion, and harvesting systems. Nano Energy **101**, 107595 (2022). https://doi.org/10.1016/j.nanoen.2022.107595
122. Wang, J.H., Cui, W., Liu, Q., et al.: Recent progress in cobalt-based heterogeneous catalysts for electrochemical water splitting. Adv. Mater. **28**, 215–230 (2016). https://doi.org/10.1002/adma.201502696
123. Lee, S.W., Carlton, C., Risch, M., et al.: The nature of lithium battery materials under oxygen evolution reaction conditions. J. Am. Chem. Soc. **134**, 16959–16962 (2012). https://doi.org/10.1021/ja307814j
124. Lu, Z., Wang, H.T., Kong, D.S., et al.: Electrochemical tuning of layered lithium transition metal oxides for improvement of oxygen evolution reaction. Nat. Commun. **5**, 4345 (2014). https://doi.org/10.1038/ncomms5345
125. Gardner, G., Al-Sharab, J., Danilovic, N., et al.: Structural basis for differing electrocatalytic water oxidation by the cubic, layered and spinel forms of lithium cobalt oxides. Energy Environ. Sci. **9**, 184–192 (2016). https://doi.org/10.1039/C5EE02195B
126. Chen, N., Qi, J., Du, X., et al.: Recycled $LiCoO_2$ in spent lithium-ion battery as an oxygen evolution electrocatalyst. RSC Adv. **6**, 103541–103545 (2016). https://doi.org/10.1039/C6RA23483F
127. Antolini, E.: $LiCoO_2$: formation, structure, lithium and oxygen nonstoichiometry, electrochemical behaviour and transport properties. Solid State Ionics **170**, 159–171 (2004). https://doi.org/10.1016/j.ssi.2004.04.003
128. Pegoretti, V.C.B., Dixini, P.V.M., Magnago, L., et al.: High-temperature (HT) $LiCoO_2$ recycled from spent lithium ion batteries as catalyst for oxygen evolution reaction. Mater. Res. Bull. **110**, 97–101 (2019). https://doi.org/10.1016/j.materresbull.2018.10.022
129. Huang, C., Lv, H., Yang, Z.H., et al.: Exfoliating spent cathode materials with robust interlayer interactions into atomic-thin nanosheets for boosting the oxygen evolution reaction. J. Mater. Chem. A **10**, 3359–3372 (2022). https://doi.org/10.1039/D1TA08046F
130. de Bruin-Dickason, C., Budnyk, S., Piątek, J., et al.: Valorisation of used lithium-ion batteries into nanostructured catalysts for green hydrogen from boranes. Mater Adv **1**, 2279–2285 (2020). https://doi.org/10.1039/D0MA00372G
131. Arif, A., Xu, M., Rashid, J., et al.: Efficient recovery of lithium cobaltate from spent lithium-ion batteries for oxygen evolution reaction. Nanomaterials **11**, 3343 (2021). https://doi.org/10.3390/nano11123343
132. Liu, T.T., Cai, S., Zhao, G.F., et al.: Recycling valuable cobalt from spent lithium ion batteries for controllably designing a novel sea-urchin-like cobalt nitride-graphene hybrid catalyst: towards efficient overall water splitting. J. Energy Chem. **62**, 440–450 (2021). https://doi.org/10.1016/j.jechem.2021.03.052
133. Cui, B.H., Liu, C., Zhang, J.F., et al.: Waste to wealth: defect-rich Ni-incorporated spent $LiFePO_4$ for efficient oxygen evolution reaction. Sci. China Mater. **64**, 2710–2718 (2021). https://doi.org/10.1007/s40843-021-1682-0
134. Fan, X.M., Hu, G.R., Zhang, B., et al.: Crack-free single-crystalline Ni-rich layered NCM cathode enable superior cycling performance of lithium-ion batteries. Nano Energy **70**, 104450 (2020). https://doi.org/10.1016/j.nanoen.2020.104450
135. Chen, Z.J., Wei, W.F., Zou, W.S., et al.: Integrating electrodeposition with electrolysis for closed-loop resource utilization of battery industrial wastewater. Green Chem. **24**, 3208–3217 (2022). https://doi.org/10.1039/D1GC04891K
136. Chen, Z.J., Zou, W.S., Zheng, R.J., et al.: Synergistic recycling and conversion of spent Li-ion battery leachate into highly efficient oxygen evolution catalysts. Green Chem. **23**, 6538–6547 (2021). https://doi.org/10.1039/D1GC01578H
137. Yang, Y.X., Yang, H.L., Cao, H.B., et al.: Direct preparation of efficient catalyst for oxygen evolution reaction and high-purity Li_2CO_3 from spent $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ batteries. J. Clean. Prod. **236**, 117576 (2019). https://doi.org/10.1016/j.jclepro.2019.07.051
138. Lv, H., Huang, H.J., Huang, C., et al.: Electric field driven de-lithiation: a strategy towards comprehensive and efficient recycling of electrode materials from spent lithium ion batteries. Appl. Catal. B-Environ. **283**, 119634 (2021). https://doi.org/10.1016/j.apcatb.2020.119634
139. Li, L., Li, X.D., Sun, Y.F., et al.: Rational design of electrocatalytic carbon dioxide reduction for a zero-carbon network. Chem. Soc. Rev. **51**, 1234–1252 (2022). https://doi.org/10.1039/D1CS00893E
140. Huang, X.F., Zhang, Y.B.: Reticular materials for electrochemical reduction of CO_2. Coord. Chem. Rev. **427**, 213564 (2021). https://doi.org/10.1016/j.ccr.2020.213564
141. Daryanavard, M., Hadadzadeh, H., Farrokhpour, H., et al.: Utilization of CO_2 as a carbon source for production of CO and syngas using a ruthenium(II) electrocatalyst. J. CO2 Util. **26**, 612–622 (2018). https://doi.org/10.1016/j.jcou.2018.06.018
142. Hua, Y., Zhang, B.W., Hao, W.B., et al.: Boosting CO desorption on dual active site electrocatalysts for CO_2 reduction to produce tunable syngas. Cell Rep. Phys. Sci. **3**, 100703 (2022). https://doi.org/10.1016/j.xcrp.2021.100703
143. Deng, L.P., Xu, Z.J., Wang, M.M., et al.: CO_2 treatment enables non-hazardous, reliable, and efficacious recovery of spent $Li(Ni_{0.5}Co_{0.2}Mn_{0.3})O_2$ cathodes. Green Chem. **24**, 779–789 (2022). https://doi.org/10.1039/D1GC02628C
144. Wang, L.Y., Gu, Y.Y., Wei, J.C., et al.: Li-Ni-Co-Mn oxides powders recycled from spent lithium-ion batteries for OER electrodes in CO_2 reduction. Environ. Technol. Innovation **18**, 100732 (2020). https://doi.org/10.1016/j.eti.2020.100732
145. Yu, J.D., Zou, S.J., Xu, G.Y., et al.: In-situ enhanced catalytic reforming behavior of cobalt-based materials with inherent zero-valent aluminum in spent lithium ion batteries. Appl. Catal. B-Environ. **303**, 120920 (2022). https://doi.org/10.1016/j.apcatb.2021.120920
146. Jung, S.Y., Kwon, D., Park, S., et al.: Valorization of a spent lithium-ion battery electrolyte through syngas formation using CO_2-assisted catalytic thermolysis over a battery cathode material. J. CO2 Util. **50**, 101591 (2021). https://doi.org/10.1016/j.jcou.2021.101591
147. Zhou, Y.J.J., Kerkhoven, E.J., Nielsen, J.: Barriers and opportunities in bio-based production of hydrocarbons. Nat. Energy **3**, 925–935 (2018). https://doi.org/10.1038/s41560-018-0197-x
148. Chen, L., Wang, P., Shen, Y.F., et al.: Spent lithium-ion battery materials recycling for catalytic pyrolysis or gasification of biomass. Bioresour. Technol. **323**, 124584 (2021). https://doi.org/10.1016/j.biortech.2020.124584
149. Wang, P., Chen, L., Shen, Y.F.: Recycling spent ternary lithium-ion batteries for modification of dolomite used in catalytic biomass pyrolysis: a preliminary study by thermogravimetric and pyrolysis-gas chromatography/mass spectrometry analysis.

Bioresour. Technol. **337**, 125476 (2021). https://doi.org/10.1016/j.biortech.2021.125476

150. Fuentes, C.A., Gallegos, M.V., García, J.R., et al.: Catalytic glycolysis of poly(ethylene terephthalate) using zinc and cobalt oxides recycled from spent batteries. Waste Biomass Valori. **11**, 4991–5001 (2019). https://doi.org/10.1007/s12649-019-00807-6
151. Paone, E., Miceli, M., Malara, A., et al.: Direct reuse of spent lithium-ion batteries as an efficient heterogeneous catalyst for the reductive upgrading of biomass-derived furfural. ACS Sustain. Chem. Eng. **10**, 2275–2281 (2022). https://doi.org/10.1021/acssuschemeng.1c08008
152. Han, Z.Y., Gao, R.H., Jia, Y.Y., et al.: Catalytic effect in Li-S batteries: from band theory to practical application. Mater. Today **57**, 84–120 (2022). https://doi.org/10.1016/j.mattod.2022.05.017
153. Xu, Y., Deng, P., Chen, G., et al.: 2D nitrogen-doped carbon nanotubes/graphene hybrid as bifunctional oxygen electrocatalyst for long-life rechargeable Zn-air batteries. Adv. Funct. Mater. **30**, 1906081 (2019). https://doi.org/10.1002/adfm.201906081
154. Primbs, M., Sun, Y.Y., Roy, A., et al.: Establishing reactivity descriptors for platinum group metal (PGM)-free Fe-N-C catalysts for PEM fuel cells. Energy Environ. Sci. **13**, 2480–2500 (2020). https://doi.org/10.1039/D0EE01013H
155. Zhu, J.W., Cao, J.Q., Cai, G.L., et al.: Non-trivial contribution of carbon hybridization in carbon-based substrates to electrocatalytic activities in Li-S batteries. Angew. Chem. Int. Ed. **62**, e202214351 (2023). https://doi.org/10.1002/anie.202214351
156. Liao, K.X., Chen, S.T., Wei, H.H., et al.: Micropores of pure nanographite spheres for long cycle life and high-rate lithium-sulfur batteries. J. Mater. Chem. A **6**, 23062–23070 (2018). https://doi.org/10.1039/C8TA08361D
157. Ng, S.F., Lau, M.Y.L., Ong, W.J.: Lithium-sulfur battery cathode design: tailoring metal-based nanostructures for robust polysulfide adsorption and catalytic conversion. Adv. Mater. **33**, 2008654 (2021). https://doi.org/10.1002/adma.202008654
158. Zhang, H., Ono, L., Tong, G.Q., et al.: Long-life lithium-sulfur batteries with high areal capacity based on coaxial CNTs@TiN-TiO_2 sponge. Nat. Commun. **12**, 4738 (2021). https://doi.org/10.1038/s41467-021-24976-y
159. Liu, Q.F., Pan, Z.F., Wang, E., et al.: Aqueous metal-air batteries: fundamentals and applications. Energy Storage Mater. **27**, 478–505 (2020). https://doi.org/10.1016/j.ensm.2019.12.011
160. Jose, V., Elouarzaki, K., Fisher, A.C., et al.: Design and integration of molecular-type catalysts in fuel-cell technology. Small Methods **2**, 1800059 (2018). https://doi.org/10.1002/smtd.201800059
161. Natarajan, S., Aravindan, V.: Recycling strategies for spent Li-ion battery mixed cathodes. ACS Energy Lett. **3**, 2101–2103 (2018). https://doi.org/10.1021/acsenergylett.8b01233
162. Liu, J.J., Shi, H., Hu, X.Y., et al.: Critical strategies for recycling process of graphite from spent lithium-ion batteries: a review. Sci. Total. Environ. **816**, 151621 (2022). https://doi.org/10.1016/j.scitotenv.2021.151621
163. Chen, Y., Wang, T.Y., Tian, H.J., et al.: Advances in lithium-sulfur batteries: from academic research to commercial viability. Adv. Mater. **33**, 2003666 (2021). https://doi.org/10.1002/adma.202003666
164. Feng, S., Fu, Z.H., Chen, X., et al.: A review on theoretical models for lithium-sulfur battery cathodes. InfoMat **4**, 12304 (2022). https://doi.org/10.1002/inf2.12304
165. Su, Y.S., Manthiram, A.: A new approach to improve cycle performance of rechargeable lithium-sulfur batteries by inserting a free-standing MWCNT interlayer. Chem. Commun. **48**, 8817–8819 (2012). https://doi.org/10.1039/C2CC33945E
166. Xu, Q., Wang, Y., Shi, X.Y., et al.: The direct application of spent graphite as a functional interlayer with enhanced polysulfide trapping and catalytic performance for Li-S batteries. Green Chem. **23**, 942–950 (2021). https://doi.org/10.1039/D0GC04033A
167. Liu, R.D., Yin, W.Z., Chen, Y.X., et al.: Facile utilization of spent $LiCoO_2$ in separator decoration of lithium-sulfur batteries. Ind. Eng. Chem. Res. **59**, 17911–17917 (2020). https://doi.org/10.1021/acs.iecr.0c03490
168. Yang, X.Y., Li, R., Yang, J.X., et al.: A novel route to constructing high-efficiency lithium sulfur batteries with spent graphite as the sulfur host. Carbon **199**, 215–223 (2022). https://doi.org/10.1016/j.carbon.2022.06.067
169. Liu, Y.Y., Ge, Z., Sun, Z.H., et al.: A high-performance energy storage system from sphagnum uptake waste LIBs with negative greenhouse-gas emission. Nano Energy **67**, 104216 (2020). https://doi.org/10.1016/j.nanoen.2019.104216
170. Liu, M.L., Zhao, Z.P., Duan, X.F., et al.: Nanoscale structure design for high-performance Pt-based ORR catalysts. Adv. Mater. **31**, 1802234 (2019). https://doi.org/10.1002/adma.201802234
171. Hu, Y.M., Zhu, M.Z., Luo, X., et al.: Coplanar Pt/C nanomeshes with ultrastable oxygen reduction performance in fuel cells. Angew. Chem. Int. Ed. **60**, 6533–6538 (2021). https://doi.org/10.1002/anie.202014857
172. Wang, H.T., Xu, S.C., Tsai, C., et al.: Direct and continuous strain control of catalysts with tunable battery electrode materials. Science **354**, 1031–1036 (2016). https://doi.org/10.1126/science.aaf7680
173. Liivand, K., Kazemi, M., Walke, P., et al.: Spent Li-ion battery graphite turned into valuable and active catalyst for electrochemical oxygen reduction. Chemsuschem **14**, 1103–1111 (2021). https://doi.org/10.1002/cssc.202002742
174. Warczak, M., Osial, M., Urbanska, W., et al.: Hydrogen peroxide generation catalyzed by battery waste material. Electrochem. Commun. **136**, 107239 (2022). https://doi.org/10.1016/j.elecom.2022.107239
175. Yamada, Y., Yoshida, S., Honda, T., et al.: Protonated iron-phthalocyanine complex used for cathode material of a hydrogen peroxide fuel cell operated under acidic conditions. Energy Environ. Sci. **4**, 2822–2825 (2011). https://doi.org/10.1039/C1EE01587G
176. Miglbauer, E., Wójcik, P.J., Głowacki, E.D.: Single-compartment hydrogen peroxide fuel cells with poly(3,4-ethylenedioxythiophene) cathodes. Chem. Commun. **54**, 11873–11876 (2018). https://doi.org/10.1039/C8CC06802J
177. Ruan, D.S., Zhang, Z.H., Wu, X.F., et al.: Synthesizing high-quality graphene from spent anode graphite and further functionalization applying in ORR electrocatalyst. ChemistrySelect **6**, 90–95 (2021). https://doi.org/10.1002/slct.202004230
178. Ruan, D.S., Zou, K., Du, K., et al.: Recycling of graphite anode from spent lithium-ion batteries for preparing Fe-N-doped carbon orr catalyst. ChemCatChem **13**, 2025–2033 (2021). https://doi.org/10.1002/cctc.202001867
179. Nie, Y., Li, L., Wei, Z.D.: Recent advancements in Pt and Pt-free catalysts for oxygen reduction reaction. Chem. Soc. Rev. **44**, 2168–2201 (2015). https://doi.org/10.1039/C4CS00484A
180. Huang, Y.Y., Wang, Y.Q., Tang, C., et al.: Atomic modulation and structure design of carbons for bifunctional electrocatalysis in metal-air batteries. Adv. Mater. **31**, 1803800 (2019). https://doi.org/10.1002/adma.201803800
181. Chen, Z., Yu, A., Higgins, D., et al.: Highly active and durable core-corona structured bifunctional catalyst for rechargeable metal-air battery application. Nano Lett. **12**, 1946–1952 (2012). https://doi.org/10.1021/nl2044327
182. Maiyalagan, T., Jarvis, K.A., Therese, S., et al.: Spinel-type lithium cobalt oxide as a bifunctional electrocatalyst for the oxygen

evolution and oxygen reduction reactions. Nat. Commun. **5**, 3949 (2014). https://doi.org/10.1038/ncomms4949

183. Jin, H.H., Zhou, H., Ji, P.X., et al.: ZIF-8/LiFePO$_4$ derived Fe-N-P co-doped carbon nanotube encapsulated Fe$_2$P nanoparticles for efficient oxygen reduction and Zn-air batteries. Nano Res. **13**, 818–823 (2020). https://doi.org/10.1007/s12274-020-2702-3
184. Jiao, M.L., Zhang, Q., Ye, C.L., et al.: Isolating contiguous Fe atoms by forming a Co-Fe intermetallic catalyst from spent lithium-ion batteries to regulate activity for zinc-air batteries. ACS Nano **16**, 13223–13231 (2022). https://doi.org/10.1021/acsnano.2c06826
185. Jiao, M.L., Zhang, Q., Ye, C.L., et al.: Recycling spent LiNi$_{1-x-y}$Mn$_x$Co$_y$O$_2$ cathodes to bifunctional NiMnCo catalysts for zinc-air batteries. Proc. Natl. Acad. Sci. **119**, 2202202119 (2022). https://doi.org/10.1073/pnas.2202202119
186. Wei, J., Zhao, S., Ji, L., et al.: Reuse of Ni-Co-Mn oxides from spent Li-ion batteries to prepare bifunctional air electrodes. Resour. Conserv. Recy. **129**, 135–142 (2018). https://doi.org/10.1016/j.resconrec.2017.10.021
187. Yang, C.G., Jin, Z.W., Zhang, X., et al.: Ultrafast regenerating spent LiCoO$_2$ lithium-ion batteries into bifunctional electrodes for rechargeable Zn-air batteries. ChemElectroChem **9**, 202101494 (2022). https://doi.org/10.1002/celc.202101494
188. Zheng, Y., Liu, Y., Hou, J., et al.: Unraveling the nature of sulfide ions in hydrometallurgical recycling of NCM622 cathode material. Energy Storage Mater. **65**, 103128 (2024). https://doi.org/10.1016/j.ensm.2023.103128

Ruyu Shi is currently a Ph.D. candidate at Tsinghua Shenzhen International Graduate School, Tsinghua University, supervised by Prof. Guangmin Zhou. His research interests mainly focus on the recycling and upcycling of key components of spent lithium-ion batteries.

Boran Wang is a postdoctoral fellow at Tsinghua Shenzhen International Graduate School, Tsinghua University. His research interests are the recycling of spent lithium-ion battery electrode materials and their high-value reuse in catalysis and environmental protection.

Di Tang is currently a master student under the supervision of Prof. Guangmin Zhou at Tsinghua Shenzhen International Graduate School, Tsinghua University. His research interests are direct recycling of cathode materials of spent lithium-ion batteries and sustainable upcycling of mixed spent lithium-ion batteries.

Xijun Wei earned his Ph.D. degree in Chemical Engineering and Technology from Chongqing University, Chongqing, China in 2020. He is currently working as distinguished associate professor at the Southwest University of Science and Technology, and conducting research in Guangmin Zhou's group as a visiting scholar. His research interests center on alkali metal-ion secondary batteries and recycling of spent lithium-ion batteries.

Guangmin Zhou is an associate professor at Tsinghua Shenzhen International Graduate School, Tsinghua University. He received his Ph.D. degree from Institute of Metal Research, Chinese Academy of Sciences in 2014, and then worked as a postdoc at UT Austin and Stanford University from 2014 to 2019. His research mainly focuses on the development of advanced energy-storage materials and devices and battery recycling.