Progress in the sustainable recycling of spent lithium-ion batteries

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Abstract
Lithium-ion batteries (LIBs) are booming in multiple fields due to a rapid development in the last decade. However, limited by operational lifespans, a growing number of spent LIBs reaching the end of their lives are consequently faced with serious accumulation and descended to hazardous waste. Without proper disposal, the spent LIBs will inevitably cause negative influence on the ecology and undermine the sustainable manufacture of LIBs. The initial research of recycling strategies mainly focused on the optimization of metallurgical processes. Recently, the sustainability of the recycling process has attracted much more attention and become an important factor. Here, we summarize the recent progress of the spent LIBs recycling from a sustainable perspective, especially discussing the green innovations in recycling strategies for spent LIBs. Through this article, we expect to reveal the challenges and developing tendency of the recycling strategies and provide a guideline for future researches on processing spent LIBs and beyond, like the recycling of the solid-state lithium metal batteries.

KEYWORDS
applications, recycling strategies, regeneration, solvents, spent lithium-ion batteries

1 | INTRODUCTION

The market for lithium-ion batteries (LIBs) has witnessed a skyrocketing worldwide demand since 1991. The LIBs have exceptional advantages over traditional secondary power sources in terms of energy density, power density, as well as lifespan. Thanks to the sweeping cost reductions in last decade, the LIBs dominated the battery market rapidly, first in consumer electronics, and followed by electric vehicles (EVs) and grids.1–3 Especially in the field of EVs, the stock keeps rapidly rising both in China and the world from 2010 to 2019 (Figure 1A). Booming of the markets brings
remarkable opportunities while it also induces side potential problems. The first thing is the hasty depletion of the various metal resources, for example, lithium and transition metals including cobalt, nickel, and manganese.\textsuperscript{4–6} According to data in Figure 1B, the metal demand for the batteries of the EVs in 2019 was about 19 kilotonnes (kt) for cobalt, 17 kt for lithium, 22 kt for manganese and that will be expanded to ten times in 2030. Once the natural resources run out, the steady supplies which underpin the development of the LIBs will be cut off from the source. However, the LIBs retired from the applications contain a much higher content of the valuable metals than natural ores, which are seen as urban mines. Realizing the recycling and reuse of the metals means a circular economy of LIBs, solving the dilemma of resource exhaustion. On the other hand, the limited lifetime of the LIBs leads to a situation that large numbers of batteries are facing retirements and accumulations. One of the side-effects of the spent LIBs is the potential pollution to the ecosystem.\textsuperscript{7} It will be incompatible with the preached greem mode of EVs if the spent LIBs are disposed improperly. As LIBs contain many toxic substances, for example, heavy metals like Co, Ni, and Mn, as well as organic electrolytes. They will inevitably contaminate the soil, underground water, and atmosphere once leaked, threatening the circumstance where we live.\textsuperscript{8,9} As shown in Figure 1C, recycling of spent battery could reduce a great deal of negative impacts on environment, even in traditional pyrometallurgical or hydrometallurgical way. Apart from the above-mentioned issues, the safety concerns of the spent LIBs cannot be neglected. There still remains some electric power in the spent LIBs, and the packs are usually sealed solidly to prevent looseness of contact, so it is easy to catch on fire or explosions during violent disposing.\textsuperscript{10} Concerning the current situation, the reasonable treatment of the spent LIBs is of vital importance and urgency.\textsuperscript{11} Maximizing the
TABLE 1 Recycling status of the typical companies and representative research groups around the world

| Company/Group                  | TRL level | Country       | Technology                        | Capacity (tonnes) |
|--------------------------------|-----------|---------------|-----------------------------------|------------------|
| Akkuser Oy                     | Commercial| Finland       | Mechanical                        | 1000             |
| Umicore                        | Commercial| Belgium       | Pyro-/Hydrometallurgical          | 7000             |
| Li-cycle                       | Pilot     | Canada        | Mechanical                        | 7500             |
| Retrieve Technologies           | Pilot     | United States | Mechanical                        | 4500             |
| Accurec                        | Commercial| Germany       | Mechanical and thermal Treatment  | 2500             |
| Valdi                          | Commercial| France        | Mechanical and hydrometallurgical | 20 000           |
| GEM High-Tech                  | Commercial| China         | Mechanical and hydrometallurgical | 10 000           |
| Brunp                          | Commercial| China         | Mechanical and hydrometallurgical | 25 000-30 000    |
| JX Nippon Mining and Metals    | Commercial| Japan         | Pyro-/Hydrometallurgical          | 5000             |
| Shengming Xu’s group           | /         | China         | Thermal treatment and Hydrometallurgical | Bench scale |
| Renjie Chen’s group            | /         | China         | Hydrometallurgical                | Bench scale     |
| Vanchiappan Aravindan’s group  | /         | India         | Graphite Recovery                 | Bench scale     |
| Zheng Chen’s group             | /         | United States | Direct regeneration               | Bench scale     |
| Ganguli Ajayan’s group         | /         | United States | Deep eutectic solvent leaching    | Bench scale     |

Economic value and minimizing the side-effect of spent LIBs can assure the stable and healthy development of LIBs.

Actually, the researchers have recognized the significance of the recycling and smelt the commercial opportunities in this field. Recycling of spent LIBs has attracted great attention from both academic and industrial fields, and technologies developed during the past 5 years are transforming quickly from an infancy stage to a diversified situation. Nowadays, learning from the experiences in e-waste recycling, the companies have implemented some traditional metallurgical processes and achieved effective recycling. Representative recycling routes of some commercial entities are summarized in Table 1. As far as we can see, pyro-/hydrometallurgical routes tend to be the first choice of the companies due to their simplicity, feasibility in large scale operation, and developed recycling devices for dealing with waste water, gas, and suspended solids.

While these routes are effective in recycling the spent LIBs, some problems are accompanied with the process. Generally, pyrometallurgical processes need high energy consumption and produce toxic and highly corrosive gases like HF, and for hydrometallurgy, more complicated processes are integrated and secondary pollutions are unavoidable. The intrinsic defects of the traditional recycling ways determine that they are not satisfactory in terms of sustainability, thus, promoting the further exploration. Therefore, numerous laboratory studies dealing with these problems have emerged recently. A few research groups with representative studies have also been listed in Table 1, from which we can intuitively see the diversity of recycling strategies at bench scale. Some studies report on the exploring of alternative systems for greener leaching and new strategies with less steps to cut costs. And some researchers pay attention to the recycling of components other than the cathode, trying to maximize the utilization of the spent LIBs. These tactics with great innovation and novelty are expected to be preferable for recycling the spent LIBs. At the same time, the changes in the recycling ways also point out that the recycling of spent LIBs must advance in a more sustainable direction.

Here, in this article, we especially concentrate on the progress in sustainable strategies for the recycling of spent LIBs. The sustainable revolutions of existing recycling technologies can be summarized as the following three aspects, involving exploration of alternatives to conventional acid/alkali, diversification of innovative tactics, and development of multifarious applications. We outline the new ideas and identify their implementary feasibility for the recycling of spent LIBs. Furthermore, we investigate the challenges and developing tendency of the recycling strategies with an aim to provide a guideline for future researches on processing spent LIBs and beyond.

2 EXPLORATION OF ALTERNATIVES TO CONVENTIONAL ACID/ALKALI

Hydrometallurgical process is common for industry as well as academia, and has been proved effective in extracting and refining metals. Numerous studies drawing experiences from other e-waste recovery have been conducted in the spent LIBs recycling. The conventional hydrometallurgical methods can be divided.
into inorganic/organic acid leaching, alkaline leaching, and bioleaching. The bioleaching is ecofriendly but has been far from meeting the requirements of practical application. Acid/alkaline leaching is facile and efficient while the secondary pollution accompanying the process cannot be neglected, requiring much cost invested to deal with the pollution. In this case, some researchers shift perspectives to novel solvents. Deep eutectic solvent (DES) is one type of new solvents with low toxicity and great ability of dissolving metal oxides. Compared with ionic liquids (ILs), DESs are obtained more cheaply and easily. DESs are a bunch of compounds that are eutectic mixtures associating with each other via hydrogen bonds interactions. In consequence, the melting point of a DES is bound to be lower than that of each individual component.

The first study of DESs for cathode recycling was reported by Ajayan and coworkers. They chose nontoxic and biodegradable precursors (choline chloride and ethylene glycol) to prepare the DES, ensuring the proposed process more ecofriendly than existing alternatives. The cathode waste inserted into the DES was heated and stirred, followed by a filtering process to separate the aluminum foil, conductive carbon, and binder from the leachate. Figure 2A obviously displayed that the color of the DES changed from clear to darker hues of blue with time increasing. More than 90% of the Co and Li was leached from LiCoO$_2$ while lower efficiencies for cobalt occurred when leaching in LiNi$_x$Co$_y$Mn$_z$O$_2$ ($x + y + z = 1$, NCM). FTIR spectra in Figure 2B confirmed the precipitate was CoCO$_3$ and the calcination promoted the conversion...
of the precipitate into Co$_3$O$_4$. This work also applied the electrodeposition to the recycling of the DES and proved the recyclability of DES (Figure 2C), reducing the consumption of the reagents as well as avoiding the secondary pollution. However, in point of fact, the leaching efficiencies are not so satisfying, especially for NCM-based cathode. This is related to the component selection of DESs. Since the composition of the DESs varies a lot, screening suitable DESs for recycling valuable elements from spent LIBs is of priority. It is acknowledged that metal oxides with high valence metal elements are usually insoluble. The reducing power of DESs, therefore, plays a key role in dissolving transition metals. Zhang and colleagues proposed a novel, simple, and robust experimental methodology based on DFT calculation and CV analysis to identify the reducibility of DESs. After systematic investigations (Figure 2D and E), the good suitability of ChCl:urea-based DES was proved due to a more strong reducibility. And the prepared DES was successfully used to recycle Li and Co from LCO at a temperature of 180°C for 12 h. Results showed that the leaching efficiency was as high as those of hydrometallurgical methods. In the future, the following researches are expected to develop other types of DESs for efficient metal leaching at low cost.

DESs have also been used to solve the separation dilemma of aluminum (Al) foil and cathode materials in spent LIBs, alternating the commonly used NaOH solution and highly toxic organic solvents. Li et al. synthesized a green and cost-effective DES, choline chloride-glycerol, and achieved a rather high peeling percentage of 99.86 wt% under the condition of heating temperature 190°C, choline chloride: glycerol molar ratio 2.3:1, and heating time 15.0 min. In this study, authors further explained the inactivation of PVDF could be attributed to a defluorination process caused by the attack of the hydroxide of choline chloride on the acidic hydrogen atom in PVDF. The same group also explored the low-temperature molten salts, for example, an aluminum chloride-sodium chloride (AlCl$_3$-NaCl) system, to melt PVDF under a relatively moderate condition. The heat storage of the phase transformation of AlCl$_3$-NaCl molten salt caused the melting of the organic binder PVDF under 160°C which was lower than the melting point of PVDF (∼172°C). Through detailed characterization, it was confirmed that the crystal structure of the cathode peeled off from the Al foil remained unchanged, revealing that the AlCl$_3$-NaCl molten salt was a nondestructive separating medium.

Another kind of green solvent is the supercritical fluid (SCF). Owing to its extraordinary properties, SCF extraction has been explored as an ecofriendly technology to recycle heavy metals. Compared with traditional leaching process, the supercritical liquid provides an extreme environment which promotes the leaching reaction. Interestingly, in a work of Zhang et al., waste polyvinyl chloride (PVC) was introduced into the subcritical/supercritical water as an acid source to achieve a simultaneous detoxification of waste PVC and the metal recovery. Relatively high leaching efficiencies of over 95% Co and nearly 98% Li proved the high efficiency of this cotreatment process. Besides, Bertuol et al. performed leaching tests with supercritical carbon dioxide (CO$_2$) and cosolvents under conventional conditions. The introduction of the supercritical CO$_2$ successfully improved the leaching performance. In comparison with the conventional method, the leaching time was sharply decreased from 60 to 5 min and the required amount of H$_2$O$_2$ was also reduced to half when a similar extraction degree was achieved. Like the utilization of spent lead-acid battery electrolyte for metal extraction, which realizes the reuse of two wastes at the same time, more attempts and explorations are needed on the leaching systems for a more sustainable, low-cost yet efficient metal recovery.

3 | DIVERSIFICATION OF INNOVATIVE TACTICS

The previous route for recycling the spent LIBs, mainly the valuable cathode, is to dissolve the metals followed by separation and purification, expecting the final products to be high value-added reagents. The whole process goes through tedious steps, including leaching, filtering, solvent extraction, precipitation, and so on. Though this route has the feasibility for all kinds of compounds containing metals, but the drawbacks are also obvious. Even after a long-time optimization of the processes, some stubborn issues still remain unsolved. Therefore, green innovations alternating the conventional methods are greatly encouraged for better recycling of the spent LIBs.

Close-loop recycling of the spent LIBs is considered to be an ideal and specific recycling route. The whole process is started from a battery and ended in a battery, saving many steps of separation and purification, especially suitable for the recycling of NCM-based cathodes or a mixed type of cathodes. At early stages, close-loop recycling was usually carried out in either a coprecipitation or a sol-gel process. There are some problems to solve which exist in above-mentioned two methods including secondary pollution and difficulties in scale-up. In the work of Janek et al., an alternative process combining the thermal decomposition and dissolution was proposed for recycling NCM-based cathodes. The NCM811 was decomposed in a CO$_2$ atmosphere in the first step to produce Li$_2$CO$_3$, followed by a carbonated water leaching for Li extraction. Next, oxalic acid was adopted to convert the transition metal oxide residues into oxalate blends. The recycled NCM811
prepared from the product of Li$_2$CO$_3$ and oxalate mixtures delivered up to 80% of the capacity of commercial NCM811. This rapid and facile approach avoids the consumption of acids or alkaline and is promising for amplification in the industry. Lai et al. designed a novel strategy that an electrolysis system ((NH$_4$)$_2$SO$_4$ solution) was used for leaching Li$^+$ from the waste cathode. The separation of active materials, the recovery of Al foil, and the leaching of Li$^+$ were integrated in one-step electrolysis. Finally, the synthesized NCM material using the recovered products as raw materials possessed a rather good electrical performance, comparable to the pristine NCM cathode materials.

Direct regeneration is another type of close-loop process that is completely distinguished from conventional metallurgical processes. Conventional metallurgical methods always destruct the structure of the active materials in spent LIBs, while the direct regeneration process restores the performance of the degraded materials in a nondestructive way. A typical recycling process with direct regeneration of the active materials are shown in Figure 3A. Essentially, extra consumption of energy is necessary for destroying the structure of electrodes. In this sense, repairing the defects in both surface and bulk based on their original compound structure is more economic. According to a calculated result conducted by Li et al., direct regeneration of spent LiCoO$_2$ has great advantages over pyro/hydrometallurgical processes in terms of energy consumption (Figure 3B). Direct regeneration is also advantageous in reducing secondary pollution and simplifying recycling process. No waste gas or liquid will be emitted into the environment theoretically (Figure 3A). Along with the advantages of a nondestructive process and significant environmental benefits, direct regeneration is expected to be the optimum route for recycling of the spent LIBs. Compared with pyro/hydrometallurgy, direct regeneration contributes a specific route for giving spent LIBs a second life and attracts much attention since the rise of spent LIBs recycling. The capacity degradation of the cathode materials in LIBs is bound up with the free Li$^+$ loss, which is ascribed to the formation of the solid-electrolyte interface and undesired phase transformation (Figure 3C). The phase with a deficient Li content could be restored to the original one through a simple lithiation process.

In a typical regeneration procedure, the degraded cathode materials are first harvested from the discarded batteries via a physical or chemical method. The as-obtained powders are ready for a post-treatment to restore to its original composition and structure. As for regenerating methods, solid-state sintering was first reported and proved effective in restoring the capacity of the degraded cathodes. However, high-temperature sintering means much energy consumption, which is not in line with the principle of sustainability. In order to cut down the energy cost, a combination of hydrothermal treatment and short thermal annealing to recycle degraded NCM/LiCoO$_2$ cathode particles has achieved great success in restoration of stoichiometry composition and repair of the undesired phase on the surface. In the hydrothermal lithiation process, lithium was complemented in the degraded materials to certain degree, followed by a short annealing to fully recover the structure and composition. The results of the experiment demonstrate excellent cycling stability of recycled materials. This simple yet effective method lays a significant foundation for recovering the degraded cathode in a sustainable way. The lithiation techniques usually require accurate calculations of the Li loss to quantify the additional Li source for the individual spent LIBs. The determination of the Li content is generally carried out by using inductively coupled plasma (ICP) technologies, of which the procedure is quite tedious. Ban and colleagues developed a new methodology to quickly determine the lithium content in spent cathode materials via a thermal gravimetric analysis, facilitating the direct relithiation recycling with a large time and cost save. The similar methods using ionothermal lithiation or a ternary eutectic repair have also been developed recently. Luo and coworkers utilized the cost-effective Li halide (lithium chloride and lithium bromide) as Li sources and repaired the degraded LiNi$_{1/3}$Co$_{1/3}$Mn$_{1/3}$O$_2$ powder in different ILs. Besides, the ILs were successfully recycled and reused, indicating the sustainability of this process. A similar system of the molten LiOH–KOH–Li$_2$CO$_3$ was adopted by Wang and colleagues to compensate the Li$^+$ loss in the degraded LiCoO$_2$, and the damaged structure was repaired via a “dissolution-recrystallization” mechanism (Figure 3D). Guo and colleagues regenerated the degraded LFP via a hydrothermal method and synthesized the nano-structured LiFePO$_4$ (LFP)/graphene composite, making full use of the discarded cathode and anode in spent LFP batteries. The LFP particles were evenly distributed between the graphene layers (Figure 3E). In consequence, the regenerated composite displayed excellent electrochemical performance, indicating a satisfactory capacity recovery (Figure 3F). Moreover, the regenerated composite exhibited better properties than those of the restored LFP via a solid-phase sintering due to its exquisite structure.

In addition to Li$^+$ compensation driven by a thermodynamic process, electrochemical relithiation, a novel process with no need for precise amount of the Li source, has triggered the wide interests. In an aqueous electrolyte, both the degraded Li$_x$CoO$_2$ powder and end-of-life cathode sheet could be fully repaired through an electrochemical relithiation in an H-cell reported by Li and coworkers. Regarding the cycled LFP, as the impurity phase in the
FIGURE 3  (A) Schematic diagram of a typical recycling process via direct regeneration. Reproduced with permission from Ref. [48]. Copyright 2017, Elsevier. (B) Comparison of energy consumption of three different methods for the LiCoO$_2$ recycling. Reproduced with permission from Ref. [49]. Copyright 2019, Wiley. (C) Mechanism on the degradation and lithiation regeneration of the layered oxide cathodes. Reproduced with permission from Ref. [23]. Copyright 2018, American Chemical Society. (D) Illustration of the failure mechanism and the regeneration processes in a molten LiOH–KOH–Li$_2$CO$_3$ system of LiCoO$_2$. Reproduced with permission from Ref. [56]. Copyright 2020, The Royal Society of Chemistry. (E) Morphologies of the nano-structured LiFePO$_4$/graphene composite prepared from the spent LFP batteries. (F) Cycling performance of the resynthesized LiFePO$_4$/graphene composite and untreated materials at 0.5C. (E and F) Reproduced with permission from Ref. [57]. Copyright 2019, Elsevier.

degraded LFP is mainly FePO$_4$, which is electrochemically reversible to LFP when intercalated in Li$^+$. Based on this point, Guo and coworkers directly regenerated the spent LFP by a graphite prelithiation strategy. In this work, the spent LFP sheet was coupled with an electrochemically prelithiated graphite sheet to assemble the recycled battery, and after the first cycle of charging and discharging, excess Li$^+$ loaded in the graphite compensated the Li$^+$ loss in degraded LFP particles. This pollution-free, economic, and facile method successfully revived the spent LFP and achieved a satisfactory electrochemical performance in the full cell. This work also proves the feasibility of the electrochemical recovery for LFP, and breaks through the new pathway for the regeneration of spent LFP. As the studies on prelithiation of graphite or silicon have achieved great success, more electrochemical regeneration processes can be explored to find a better way for LFP recycling based on these strategies.
### TABLE 2  Examples of the direct regeneration of the cathode in spent LIBs

| Regenerated product       | Regenerated strategy                        | Initial discharge capacity (mAh/g) | Capacity retention | Reference |
|--------------------------|---------------------------------------------|-----------------------------------|--------------------|-----------|
| LiCoO₂                   | Solid-state synthesis                       | 152.4 (30 mA/g)                   | Attenuation rate of capacity during every cycle: 0.0313 mA h/g | 52        |
| LiCoO₂                   | Hydrothermal reaction + short annealing      | 153.1 (15 mA/g) 148.2 (150 mA/g)  | 91.2% at 150 mA g for 100 cycles                      | 22        |
| LiCoO₂                   | Electrochemical relithiation                | 136.0 (0.2 C)                     | No significant decay after 200 cycles                 | 60        |
| LiCoO₂                   | Electrochemical relithiation                | ~140.0 (0.1 C)                    | 93% at 0.1 C for 100 cycles                           | 49        |
| LiCoO₂                   | Thermal-chemical treatment                  | 144.5 (30 mA/g)                   | 92.5% at 30 mA g for 200 cycles                        | 56        |
| Al₂O₃-coated LiCoO₂       | Solid-state calcination and coating         | 136.8 (20 mA/g)                   | 90.1% at 20 mA g for 100 cycles                        | 61        |
| Nano Al₂O₃-coated LiCoO₂  | Ultrasonic separation and renovation        | 166.3 (not mentioned)             | 98% for 50 cycles                                     | 62        |
| LiNi₁/₃Co₁/₃Mn₁/₃O₂       | Hydrothermal reaction + short annealing      | 158.4 (150 mA/g)                  | 77.4% at 150 mA g for 100 cycles                       | 23        |
| LiNi₀.₃Co₀.₂Mn₀.₅O₂        | Solid-state synthesis                        | 161.25 (16 mA/g)                  | 95.25% at 80 mA g for 50 cycles                        | 53        |
| LiNi₀.₃Co₀.₂Mn₀.₅O₂        | Eutectic Li⁺ molten-salt + short annealing   | 149.3 (150 mA/g)                  | 90.2% at 150 mA g for 100 cycles                       | 24        |
| LiFePO₄                   | Doping with new LiFePO₄                     | 144.0 (0.1 C)                     | Not mentioned                                         | 63        |
| LiFePO₄                   | Electrochemical lithiation                  | 126.6 (0.5 C) in the full cell with graphite anode | 65.5% at 0.5 C for 200 cycles                          | 58        |
| LiFePO₄/graphene           | Hydrothermal method                         | 163.3 (0.2 C)                     | 99.63% at 0.2 C for 100 cycles                         | 57        |

As we can conclude from the above, compared with the processes for obtaining high-purity reagents, these resynthesis methods seem more simplified and cost-effective indeed. But as far as we can see, the limitation of this route is also obvious. The final products have barriers both in quality and market recognitions for applying in other fields except the LIBs’ upstream. Besides, the electrochemical performance of resynthesized cathodes is hard to match commercial materials. As summarized in Table 2, it is obvious to see that there is a gap between the performance of the regenerated materials and commercial materials. In the future, more endeavors should be devoted to design ingenious methodology, which has better quality control to enhance the regenerated performance and is convenient for scale-up production to achieve low-cost yet efficient recycling of spent LIBs in industry.

Apart from the recycling of electrode materials, electrolyte recovery, as another important source of Li, has also attracted wide attention of researchers. Conventional recycling processes based on pyrometallurgy and hydrometallurgy has reached good efficiency of electrolyte recovery. Li could be successfully recycled in the form of Li₂CO₃ or LiPF₆.⁶⁴–⁶⁶

Compared with the commonly used pyrometallurgy and hydrometallurgy, SCF/subcritical fluid extraction is a selective and environmentally friendly separation method. Nowak and his coworkers applied sub- and supercritical carbon dioxide to the extraction of electrolytes from the spent LIB earlier.⁶⁷–⁶⁹ They investigated that adding some solvents to carbon dioxide can effectively increase the recovery rate of each component of the electrolyte, especially lithium salt (LiPF₆).⁷⁰ In a series of studies by Dai et al., electrolyte recovery via supercritical carbon dioxide has also been conducted.⁷¹–⁷³ They reported the highest extraction yield of 85.07 ± 0.36% at 23 MPa and 40°C for 45 min. Furthermore, they presented a complete set of procedures which is composed of supercritical CO₂ extraction, resin, and molecular sieve purification, and component supplements for purification and characterization. The Li/LiCoO₂ battery based on recovered electrolyte delivered an initial discharge capacity of 115 mAh/g and the discharge capacity retention of 66% after 100 cycles at 0.2 C.⁷⁴

### 4 DEVELOPMENT OF MULTIFARIOUS APPLICATIONS

The “waste to wealth” concept is widely accepted in the recycling of spent LIBs.⁷⁵,⁷⁶ Other than the reutilization in the LIB manufacture, massive interests have been aroused...
in developing new applications in an economic manner, exploring suitable applications not only for the cathode materials but also for the relatively low value-contained graphite anode. Here, we briefly summarize and discuss the applications of the recycled materials in other fields.

Cobalt oxide (Co$_3$O$_4$) prepared from the spent LiCoO$_2$ was applied as a pseudocapacitor in supercapacitors by Freitas’s group. Similar to metallic Co, it has also been synthesized from the spent cathode and exhibited a specific capacitance of 625 F/g. Besides, catalysts are another important application field for spent LIBs. Transition metals in cathode are typical elements contained in catalysts. Metallic metals or their oxides could be recovered from the spent cathode. Natarajan et al. developed an easy strategy for the recovery of a valuable mixed metal oxide material in the form of spinel MnCo$_2$O$_4$ microspheres from spent LIBs and the prepared materials were used as a catalyst for OER electrocatalysis in alkaline medium. The results revealed that the recycled catalyst was advantageous in lower overpotential, lower Tafel slope, and excellent cycling stability compared with commercial catalysts. Wu et al. reported Li-Ni-Co-Mn oxides as an innovative anode catalyst for CO$_2$ electrolysis with high current efficiency, showing great potential for industrial use. In all, the key point of the recycled catalyst is the structure design and impurity control. Moreover, the cost of the fabrication process requires consideration as well.

Previous studies largely overlooked graphite recycling because of its relatively low cost compared with that of the cathode. The discard of waste graphite is obviously contrary to the purpose of green chemistry. Recently, apart from the preparation of battery-grade graphite from spent LIBs, there have emerged many studies revealing that recycled anode materials can be reused in many other fields. The graphite interlayer spacing becomes wider after continuous lithiation/delithiation, which may accommodate larger metal ions such as sodium ions and potassium ions. Wu et al. provided a new recycling concept that graphite from spent LIBs was reused as anode in Na-ion batteries (NIBs) and K-ion batteries (KIBs). The recycled graphite by pyrolysis process delivered reversible capacity of 162 mA h/g at 0.2 A/g and 94.6% capacity retention for 1000 cycles at 2 A/g in NIBs. Furthermore, recycled graphite was proved to have fewer defects and impurities by Raman spectra and X-ray photoelectron spectroscopy, respectively (Figure 4A). In addition, Divya et al. proposed a novel route to use recycled graphite as anode materials in the lithium-ion capacitor (LIC). The dual carbon LIC delivered a maximum energy density value of 185.54 Wh/kg and a good cycling performance at 10 and 25 C. Recycled graphite was also applied to sodium-ion capacitors.

Similar to sodium-ion battery, Na-insertion into graphite is extremely limited. The Na storage mechanism, including the percentage contribution of intercalation pseudocapacitance and diffusion-controlled intercalation, was thoroughly discussed in the report.

Graphene has been extensively studied because of its excellent mechanical, structural, thermal, and electrical properties. But there is still lack of efficient and low-cost preparation method. Due to the intercalation and deintercalation of Li$^+$ in the graphite during cycling, interlayer force decreases and the interlayer spacing increases. Zhang et al. proposed the battery cycling can be considered as a prefabrication step, which can largely enhance the productivity of graphene and its derivatives. They integrated the graphene fabrication with the recycling of LIBs and confirmed the assumption (Figure 4B). However, internal and external of graphite from spent LIBs were contaminated with organic and inorganic impurities, which seriously affected its subsequent implementation. Yu et al. established a characterization system with variable scales to analyze the distribution of impurities in the graphite electrode and revealed the mechanism of the modified Hummers method to remove impurities.

With the acceleration of social industrialization, the pollution of water used in production has become more and more serious. Therefore, some studies have applied graphite recovered from spent batteries to wastewater treatment. Zhang et al. reported the mesocarbon microbeads (MCMB) carbon was recycled as a highly efficient phosphate adsorbent by modifying it with nano-sized Mg(OH)$_2$ on the surface. The adsorbent showed a high phosphate adsorption capacity, up to 588.4 mg/g (Figure 4C). Hao et al. prepared an adsorbent for cadmium from anode materials through ball milling and in situ loading of MnO$_2$. Due to the increasing of surface area, the number of surface functional groups and defects, the obtained materials owned an adsorption capacity of 135.81 mg/g. In general, wide application of recycled anode has emerged and is booming, which shines new light on the future works of recycling of spent LIBs.

## 5 | SUMMARY AND PROSPECTS

With the worldwide popularity of the EVs and consumer electronics, and the extensive arrangement of large-scale energy storage system, the development of LIBs has reached a new level. However, much attention should be paid for conquering the troublesome situation of retired spent LIBs. With the first wave of spent LIBs on the road, recycling of spent Li-ion batteries has become a critical issue for alleviating resource anxiety and enabling economic and environmental sustainability of Li-based energy storage. Researchers in both industrial and academic fields are working hard on developing a sustainable and
effective route to recycle the spent LIBs. Different from the traditional ideas of metallurgy, some novelties have arisen in the recycling of spent LIBs. It is good to see that great improvements have been made in terms of the recovery efficiency, cost reduction, pollution control, and so on. However, these approaches are mostly in the laboratory stage, which means that few of them have truly realized the industrialization. Much consideration and effort for transforming laboratory researches into industrial applications is still in large need. Here are some perspectives for future research on recycling spent LIBs:

First of all, universal recycling strategies for commercial LIBs are necessary. Various cell chemistries are common presented in commercial systems, like LFP-, LCO-, NCM-, and LiNi_{1-x}Co_{x}Al_{y}O_{2} (NCA)-based cathode materials. Actually, the spent LIBs are usually a mixture of them. As the cost effectiveness of recycling mostly reflects in the energy saved and the final products with high added-value, whether it is cost effective for recycling the mixture without sorting is worth of well evaluation.

Second, in-depth researches on recycling need to be developed. There is a specific characteristic for the current recycling studies that the efficiency outweighs the mechanism to some degree. Nevertheless, in-depth investigations into the degradation mechanism and reactions within the substances in the battery system do make a difference. For example, digging the subtle difference of Ni, Co, and Mn elements do help separate them. Thus, more efforts need to be made for conducting the mechanism research and establish a theoretical basis.

Third, the recycling of the LIBs must catch up with the development of the LIBs manufacture. Nowadays, LIBs are changing rapidly both in material and battery system design. Generally, the lithium battery is moving towards a new generation with higher energy density, power density, and safety. Recycling of new types of LIBs and beyond requires corresponding adjustments. In solid-state lithium metal batteries, the separation of different components and the recycling of the lithium metal and the solid electrolytes is a new project for exploring.

Last but not the least, pollution control is always the first considering priority. More attention should be paid to the design of a green way instead of a secondary waste treatment to solve this problem fundamentally.

Recycling of the spent LIBs is a burgeoning field yet remains a lot of room for development. Researches in the
laboratory that take the lead should put more endeavors into designing distinguished routes for recycling the spent LIBs, taking full advantages of the residual value in the spent batteries. The overall target is to build an ecobenign, efficient, and cost-effective recycling system for the coming of spent LIBs wave. Appropriate treatment of spent LIBs is the best way for achieving sustainable and green development of LIBs industry.

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CONFLICT OF INTEREST
The authors declare no conflict of interest.

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