

Review

# The Sustainable and Green Management of Spent Lithium-Ion Batteries Through Hydroxy Acid Recycling and Direct Regeneration of Active Positive Electrode Material: A Review

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**Abstract:** The rapid increase in use of lithium-ion batteries in energy storage together with limited supply of critical metals used in batteries and environmental concerns have led to the urgent need for sustainable recycling technologies for these batteries. Li-ion battery chemistry, components, various designs, and two main approaches for recycling: pyrolysis and hydrometallurgical techniques are discussed in this review focusing on the novel, sustainable green approach of hydroxy acid leaching followed by a direct regeneration technique. This two-step emerging technique is compared with other conventional recycling methods in this critical review emphasizing simplicity and commercial potential. Current literature reporting rapid developments on this scalable process with pretreatment phases of sorting, discharging, disassembly of batteries, separation of electrode coatings from current collectors, leaching black mass with hydroxy carboxylic acids, separation of graphite, adjustments of Li, Ni, Mn, and Co compositions, and regeneration via co-precipitation or sol–gel formation techniques followed by pyrolysis are discussed in the detailed review. The conclusion section of this direct regeneration focused critical review gives an insight into challenges in hydroxy acid recycling and direct regeneration technology and practical solutions that may help in development into a mainstream technology.



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## 1. Introduction

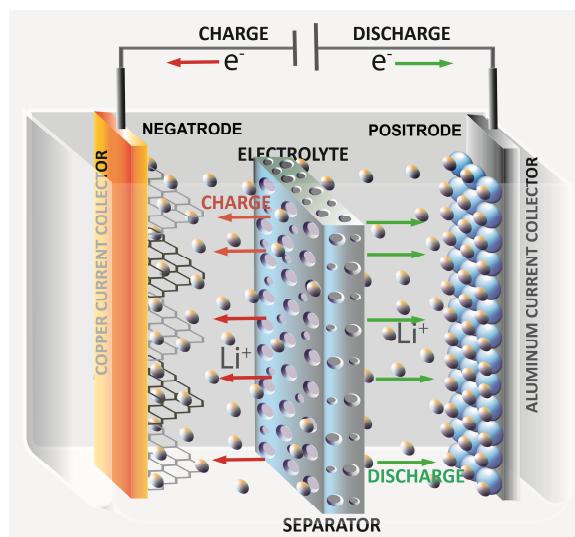
The use of lithium-ion batteries (LIBs) for energy storage has seen exponential growth in the last decade, and most of the progress is noted in mobile electronic devices, transportation, home solar power systems, and large-scale solar farm sectors [1,2]. The energy storage capacity of Li-ion batteries is based on the capability of lithium ion to intercalate between graphite and certain layered metallic frameworks. Intercalation of group I alkali metals such as Li, Na, and K in graphite has been known since 1926, and the formation of group I metal intercalation compounds with the layered dichalcogenides hosts was first recognized by Rudorff and Sick in 1959 [3,4]. Since these early observations, several researchers have noted the intercalation of small group I metals with metallic frameworks and graphite. Later, Whittingham summarized the early progress in this area in the 1978 landmark review titled “Chemistry of Intercalation Compounds: Metal Guests in Chalcogenide Hosts” drawing the initial attention to the intercalation process [5]. In the follow-up work, Whittingham and co-workers developed the first Li-ion rechargeable battery under an Exxon research

project, and Exxon manufactured the titanium disulfide positive electrode (positrode) and Li-Al negative electrode (negatrode) based early Li-ion batteries in the late 70s and early 80s; however, the original attempts were not commercially successful. Similarly, in the 1980s, Goodenough and co-workers corroborated the idea of energy storage using exchange of Li ions, expanding Whittingham's work by using more stable lithium cobalt oxide as a positive electrode material [6,7]. Nevertheless, the first direct precursor of the modern Li-ion battery was fabricated by Akira Yoshino in 1983 by using polyacetylene anode rather than a lithium metal negative electrode and lithium cobalt oxide positive electrode [8]. In recognition of their contributions to the development of Li-ion batteries, Stanely Whittingham, Akira Yoshino, and John Goodenough were awarded the Nobel prize in chemistry in 2019 [9].

A number of global reports are clearly pointing out the rapid growth expected in the use of lithium-ion batteries; for example, the world economic forum projected that the worldwide battery demand will rise to 2600 GWh in 2030 from the present demand of about 600 Wh kg<sup>-1</sup> [10]. Currently, NMC532 and NMC622 LiBs are state-of-the-art Li-ion battery cell types due to their excellent Wh kg<sup>-1</sup> energy densities and good charging/discharging rates. These batteries are known to offer up to 750 Wh kg<sup>-1</sup> [10] energy density; however, according to a study on future production of lithium-ion battery cells by Degen et al., lithium–iron–phosphate (LFP) and high-nickel content NMC811 battery cells are expected to dominate the market share in 2040 [1]. Likewise, the market segment of new types of LiBs, or the post lithium-ion battery (PLiB) cells are expected to be important in 2030s and may reach as high as 20–30% in 2040 [1]. The life expectancy of LiBs is an additional critical factor and generally varies based on the specific application. LiBs used in lightweight mobile electronic appliances such as cellular telephones, tablets and laptop computers are expected to last 2 to 3 years, while batteries used in electric automobiles are generally required to perform for 8 to 10 years [11]. Nonetheless, the wide use of Li-ion batteries in many applications inevitably results in a rapid accumulation of spent LiBs [12–14]. The estimation of worldwide accumulation of disposed Li-ion batteries is a challenging undertaking, and a 2017 assessment by Gu and co-workers forecasted that over 400 million tons of spent Li-ion batteries are estimated to be disposed by 2020 [15]. The wide use of electric automobiles has radically affected these estimates and the number of electric automobiles on our roads has been rapidly growing and reached over 7 million vehicles at the end of 2019 [16]. It is projected that by 2030, the global proliferation of electric automobiles will result in the availability of over 1000 GWh of Li-ion batteries that will soon require retirement due to their failure to fulfill the crucial performance standards for usage in electric automobiles [17].

## 2. Working Principle of Lithium-Ion Battery

The lithium-ion battery is based on the ability of Li<sup>+</sup> ions to intercalate in the mixed metal oxide positive electrode and graphite negative electrode as well as the ability to move back and forth between positive and negative electrodes through the electrolyte medium in the separator between the electrodes. The working principal of an LIB while charging and discharging the battery is shown in Figure 1. During the discharging/charging process there is a movement of lithium ions from the negatrode to positrode or vice versa and movement of electrons via an external circuit. The battery has two states: in the discharging mode, lithium becomes oxidized to Li<sup>+</sup>, releasing one that electron moves through an external circuit. In the discharging mode, Li<sup>+</sup> and electrons move towards the negatrode in the reverse direction [18,19].



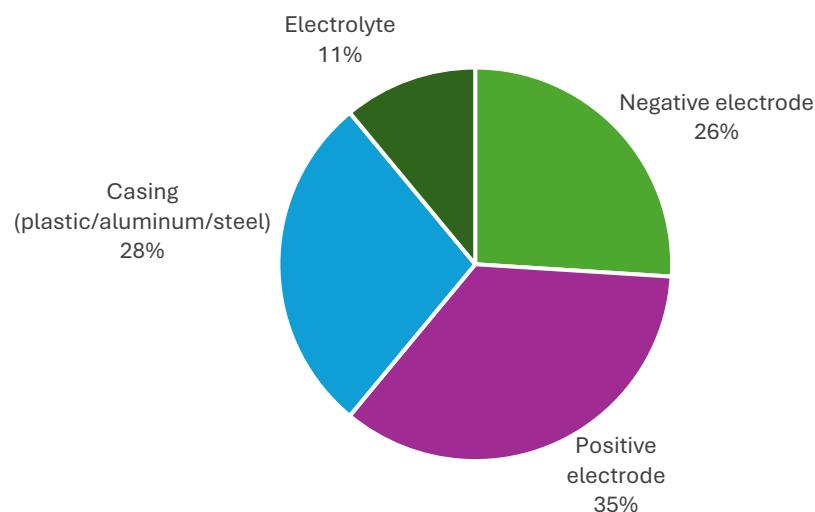
**Figure 1.** Working principle and components in the Li-ion battery.

The major components of the Li-ion battery include an active positive electrode material coated on an aluminum foil current collector, graphite negative electrode coated on a copper foil current collector, and a polymer membrane separator between the two electrodes, which is soaked with an electrolyte or polymer gel to facilitate the movement of lithium ions as shown in Figure 1 [20]. Furthermore, metal–organic framework (MOF) materials have also been studied as electrode coatings in attempts to improve the capacity, rate capability, and safety of LiBs [21]. Battery chemistry, design, charge/discharge characteristics, energy capacity, safety features, and the cost of lithium-ion batteries can vary across a broad spectrum for these Li-ion batteries. Mobile electronic devices such as cellular telephones use Li-ion batteries with polymer gel-type electrolytes where the weight of the battery is a critical factor. This type of Li-ion batteries uses a lithium cobalt oxide ( $\text{LiCoO}_2$ ) positive electrode material and a graphite negative electrode together with a polymer gel electrolyte in place of a liquid electrolyte offering a high energy density but a relatively shorter life span [22,23]. Lithium nickel manganese cobalt oxide ( $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ ), lithium iron phosphate ( $\text{LiFePO}_4$ ), and lithium manganese oxides such as  $\text{LiMn}_2\text{O}_4$  spinel or  $\text{Li}_2\text{MnO}_3$ -based lithium rich layered materials as positive electrode materials are known to offer high discharging rate capabilities and longer battery lives. This type of Li-ion battery with liquid electrolytes offers longer lifespans and are widely used in electric vehicles, power walls, and back-up power packs, medical equipment, and electric tools. In addition, an extensive selection of lithium mixed metal oxides with Li, Ni, Mn, and Co ratio variations are presently in use as LiB positive electrode materials in modern batteries [24]. The most common mixed metal oxide is  $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ , generally abbreviated as LiNMC, where the Li:Ni:Mn:Co ratio can be varied from one battery manufacturer to another, and positive electrode materials are named based on the Ni:Mn:Co ratio; for example, NMC 111 is the mixed metal oxide with Ni:Mn:Co 1:1:1. In addition, Al and Ti have also been used as a dopant or replacement for one of these metals in a smaller fraction of current lithium-ion batteries [25].

Positive electrode materials are mixed with acetylene carbon black to improve electrical conductivity; in addition, a binder like polyvinylidene fluoride (PVDF) and a dipolar aprotic solvent are used to disperse the solids while coating an aluminum positive electrode current collector. Acetylene black enhances the conductivity and electrode porosity, reduces agglomeration, and improves the electrode structure [20,26,27]. The electroactive materials and conductive additives in electrode coatings are held together using binders [28,29]. The common binders used in current LiBs are polyvinylidene fluoride (PVDF), carboxymethyl

cellulose (CMC), styrene-butadiene rubber (SBR), and polyacrylic acid (PAA) [30,31]. In addition, N-methyl-2-pyrrolidone (NMP) is a high-boiling organic solvent, commonly used in coating the active material on Al and Cu current collectors. NMP is the solvent of choice for this role due its low viscosity, toxicity, and flammability as well as its dipolar characteristic, which helps to dissolve the lithium salt and binders like PVDF and has good wetting properties that ensure good adhesion leading to uniform coatings [32].

The anode of LiBs is composed of a graphitic carbon coating, like the material that has been widely used as support in a heterogenous catalysis [33,34]. The conductive graphite is also used together with PVDF and NMP to facilitate the dispersion, achieving an even coating with good contact to the copper foil current collector [35]. Another important component of battery is the separator soaked with the electrolyte, which carries  $\text{Li}^+$  during the charging and discharging process. The commonly used electrolyte in LiBs is lithium hexafluorophosphate ( $\text{LiPF}_6$ ), which is diluted with an organic solvent like dimethyl carbonate, diethyl carbonate, ethyl carbonate, polypropylene carbonate, and dimethyl sulfoxide (DMSO). Similarly, lithium perchlorate ( $\text{LiClO}_4$ ), lithium triflate ( $\text{LiCF}_3\text{SO}_3$ ), lithium tetrafluoroborate ( $\text{LiBF}_4$ ), and *bis*(trifluoromethane) sulfonimide lithium salt ( $\text{LiN}(\text{SO}_2\text{CF}_3)_2$ ) are also being used as electrolytes [36–38]. The separator prevents the short circuit between the electrodes, and generally this middle component is a porous polypropylene, polyethylene, or glass fiber membrane [20,39]. Since there are numerous designs and chemistries, the composition of spent LiBs depend on the classification; nevertheless, the average weight distribution of common LiBs can be grouped into four major sectors as shown in Figure 2. The positive and negative electrode carries the most weight in Li-ion batteries, and electrolytes carry 11% of the weight as shown in Figure 2. Recycling expensive electrolytes [40] is another area of interest; however, this review focuses on the recycling of the electrode-coating active materials in Li-ion batteries.



**Figure 2.** Average weight percentages of components in common LiBs.

### 3. LiB Recycling Methods

Recycling LiBs is one of the present-day high-priority research areas in the sustainability and energy materials research field due to the urgency in the development of technologies that can help in mitigating climate change by decarbonizing the energy sector [41–44]. Recycling valuable critical metals such as Li, Ni, Mn, and Co used in LiBs is an essential module in this endeavor due to limited supply of essential metals and environmental concerns in disposing large quantities of used batteries [45,46]. Therefore, many research groups around the globe have taken the challenge in the development of industrially scalable and economical LiB recycling technologies over the past 5–6 years [47–50]. The LiB

recycling technologies, current trends, and market insights are reviewed in several reviews in recent times [51–53]. Firstly, He and co-authors reviewed the industrial market trend and current status of the global LiB recycling industry, and this review article summarizes the three most popular Li-ion battery recycling technologies, the current Li-ion battery recycling market trend, and global recycling magnates and industrial dynamics regarding this subject [51]. Secondly, Meegoda and co-authors have presented a comparative analysis of various recycling methods and assessments of these diverse recycling processes and capacities among key players in the industry [54]. In another review titled “Recycling of lithium-ion batteries: Recent advances and perspectives”, Huang identified a number of ways to establish efficient recycling systems such as (1) improvements in identification and sorting spent LIBs; (2) user-friendly labeling of LIBs facilitating the routing in recycling; (3) designing LIBs with recycling in mind and avoiding irreversible and complex assembling methods; (4) and accelerating legislation to standardize the recycling processes [55]. As common LiB recycling is reviewed in a number of recent review articles, this review will focus specifically on the hydrometallurgical process using hydroxy acids as leaching agents and direct regeneration of active cathode material from leachate solutions. This two-step emerging technique has certain advantages over other conventional recycling methods due to its simplicity and emerging commercial potential.

The LiB recycling methods can be grouped into two main divisions, pyrometallurgy and hydrometallurgy [56,57]. Pyrometallurgy involves the smelting of spent LiBs in a furnace above 1200 °C, usually with a reductant to produce metal alloys of Co, Cu, Fe, and Ni [58]. Basically, spent LIBs are burnt in a furnace without any type of pretreatment. The organic materials, electrolytes, solvents, plastics, and separators are combusted. This highly energy consuming process requires heating up to about 1500 °C, and burning organic electrolytes and plastics generates toxic gasses, such as dioxins, furans, hydrogen fluoride and halogens. The carbon in graphite tends to reduce metal oxides to metals, producing metallic alloys. The follow-up downstream hydrometallurgical process requires dissolution of the alloy in a concentrated mineral acid like nitric acid and separation of metals using precipitation and selective solvent extraction methods [53]. The main advantages of pyrometallurgy over hydrometallurgy are that this high temperature process does not require any pretreatments involving sorting under battery chemistries and cell types, discharging, mechanical dismantling, or separation of electrode coatings [53,59]. On the other hand, the disadvantages include the high energy costs, loss of all organic matter like plastics as well as some metals like Li and Al, and also the emission of harmful gasses during pyrolysis. Then, there are challenges in separating individual metals in their pure form or as salts for reuse in batteries or other applications [59]. In addition, deep eutectic solvents (DESs) like choline chloride–organic acid mixtures have also been used in recycling active positive electrode materials in LiBs, and Wang et al. have recently reviewed the progress in this area of LiB recycling technologies [60].

#### 4. Hydrometallurgical Process

The hydrometallurgy process involves the use of aqueous solvent system to leach out active positive electrode materials from spent LiB electrode coatings. Mineral acids, organic acids, or base solutions are often used under heating, sonication, or microwave conditions in hydrometallurgical processes. In addition, reductants such as hydrogen peroxide or ascorbic acid are frequently added to aqueous leaching medium to reduce Co(III) to Co(II) and improve the leaching. The hydrometallurgy process typically comprises three fundamental steps: pretreatment, leaching, and filtrations of metal ion containing leachate from graphite. The principal steps of the pretreatment process are sorting, discharging, dismantling, mechanical separation of electrodes, coating separation, pyrolysis, sieving to

give black electrode coating powder, and leaching of Li, Ni, Mn, and Co as illustrated in Figure 3. The primary objective of this process is to dissolve the electroactive materials in an aqueous solvent, thereby facilitating subsequent separation, purification, or regeneration steps. Mechanical agitation and ultrasonication are often employed in hydrometallurgical processes to accelerate the leaching and enhance recovery efficiency.



**Figure 3.** Pretreatment process and leaching of Li, Ni, Mn, and Co from spent Li-ion battery electrode coatings.

## 5. Pretreatment for Hydrometallurgical Process

### 5.1. Battery Classifications and Sorting

Sorting batteries based on size, shape, manufacturer, chemistry, and other identifiable physical characteristics is a crucial step in the hydrometallurgical method since these parameters significantly influence the final purification process and product yield. Additionally, categorizing batteries according to these criteria can reduce the dismantling efforts and time, improve leaching efficiencies by selecting the best leaching reagents and conditions for each class, as well as improving product quality and consistency. For instance, while ongoing research explores diverse options, six lithium-based metal oxides are currently employed as positive electrode material in today's LIBs. These include  $\text{LiCoO}_2$  (LCO),  $\text{LiFePO}_4$  (LFP),  $\text{LiNiMnCoO}_2$  (NMC),  $\text{LiNiCoAlO}_2$  (NCA),  $\text{LiMn}_2\text{O}_4$  (LMO), and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  (LTO) [61]. The separation of batteries according to battery chemistry is critical in the hydrometallurgical leaching process, particularly when direct regeneration without separation of metals is used downstream to avoid excessive adjustments in metal ion concentrations.

### 5.2. Discharging Spent LIBs

Discharging LIBs is an essential pretreatment step and a safety precaution to avoid electric short circuits and fires since spent LIBs may still have stored electrical energy instigating fast discharging, causing fires and explosions [62]. The common method of spent LiB discharging, reported in the literature, involves immersing LiBs in brine or other electrolyte solutions. Generally, 10% *w/v* sodium chloride or sodium sulfate is used for discharging spent batteries by immersion for 24–48 h [63]. In addition, solid electrical conductor discharge, cryogenic methods, and thermal deactivation are also reported in the literature [61].

### 5.3. Dismantling and Mechanical Separation of Spent LiBs

Dismantling or disassembly of spent LiBs involves the physical separation of the three main components in an LiB: positive electrode, negative electrode, and the separator from outer casing as shown in the scheme in Figure 3. In the laboratory experiment, scale disassembly can be carried out manually using tools like knives, saws, and scissors. However, industrial scale recycling operations require mechanical separation devices or machines. There are three common designs of LiBs: cylindrical, prismatic, and pouch types. A cylindrical LiB cell contains a sheet-like negative electrode, separator, and positive electrode that are sandwiched, rolled up, and packed into a cylindrical outer casing. This is the most common and first mass-produced type of LiBs and still the most popular design. The Li-ion prismatic cell contains a larger sheet of negative electrode, positive electrode, and separator sandwiched, rolled up, and pressed to fit into a hard plastic or metallic casing in the form of a cube. The third type, known as pouch LiB cell, does not have a rigid casing, but is used as a sealed and flexible foil as the cell enclosure. The disassembly technique may depend on the battery type as well as the size of the cell. The first step is the removal of the plastic, steel, or aluminum outer casing. Secondly, positive and negative electrodes and the separator are rolled back, separated, and collected for further processing [64].

The next step in the pretreatment processes is the removal of electroactive material coatings from positive electrode and negative electrode current collectors. Various solvents such as methyl-2-pyrrolidone (NMP), sodium hydroxide, N,N-dimethylacetamide (DMAC), dimethyl sulfoxide (DMSO), and N,N-dimethylformamide (DMF) are employed in this process, either individually or in combination with ultrasonication or heating [65]. Then, black mass collected from electrodes are usually pyrolyzed to remove binders and electrolytes sticking on to graphitic black mass. This thermal treatment enhances the leaching efficiency by weakening the bonding between graphite and mixed metal oxides, releasing the electroactive materials. Pyrolysis at temperatures between 150 and 900 °C for durations ranging from 0.5 to 5 h is common [66].

## 6. Mineral Acid Leaching

Acid leaching is one of the earliest methods used in the recovery of valuable metals from electrode coatings of LiBs. Common mineral acids HCl, H<sub>2</sub>SO<sub>4</sub>, and H<sub>3</sub>PO<sub>4</sub> have been used; however, the corrosive nature of these acids, need for large quantities of bases in neutralization, and environmental effects on disposal of large quantities are certainly negative factors in using mineral acids. Therefore, many researchers have turned their attention to organic acids as leaching agents for recovery of metals. The relatively low acidity, ease of handling, and chelation ability and the possibility of recovery of the metal by burning off the organic acid are advantages of organic acids. Among many organic acids tested, hydroxy acids with one or more neighboring hydroxyl groups are particularly effective in chelating with Li, Ni, Mn, and Co cations and promoting the leaching.

## 7. Carboxylic Acid Leaching

Organic acids generally form metal–organic chelated complexes by electron donation to metal cations present in electroactive materials. This method of extraction is environmentally friendly, safer, and produces minimal secondary waste while achieving high recovery rates. Organic acids are predominantly produced by microorganisms and plants. A number of monocarboxylic acids, such as formic and acetic acids, dicarboxylic acids like succinic, as well as -OH and NH<sub>2</sub> functionalized carboxylic acids, have been studied as leaching agents in hydrometallurgical process [67–69]. The smallest carboxylic acid, formic acid, is widely used as a leaching agent, which functions as a reducing agent as well [70–72]. Zeba and co-workers reported over 99% leaching of Li, Ni, and Mn using 10 mol L<sup>-1</sup> formic

acid at ~80 °C for 3 h. Furthermore, they have noted that formic acid played the dual role as a leaching agent and a reductant for Ni and Mn. However, only ~50% of Co was leached under these conditions. In addition, they reported that the insoluble cobalt could be rapidly dissolved by adding H<sub>2</sub>O<sub>2</sub> to the formic acid leaching solution [73]. In another example, Zeba et al. reported a 99.9% recovery of Li from spent LiFePO<sub>4</sub> batteries by using 2.5 mol/L formic acid under a 0.12 L/min oxygen flow and 25 mL/g solid loading at 70 °C for 3 h. Moreover, the leaching efficiency of Fe was only 1.7%, showing the selectivity of formic acid towards Li under the conditions employed [73]. In another example, Gao and co-workers also highlighted the use of formic acid for leaching Li at 99.93% yield while aluminum remained as the metallic form [74]. The same research group has used acetic as well, reporting comparable success in leaching Li (99.97%), Ni (92.67%), Mn (96.32%), and Co (93.62%) [75]. A rare example of using a carboxylic and sulfonic acid mixture was reported by Fu and co-workers, where a mixture of 1.5 M formic and 1.3 M benzenesulfonic acid was used to leach out 99% Li and 97% Co from a LiCoO<sub>2</sub>-type positive electrode coating at 50 °C [76]. Similarly, several other carboxylic acids, oxalic [77], benzoic [78], and trifluoro acetic [79], have been tested in leaching out electroactive materials from spent LiB black mass.

## 8. Hydroxy Carboxylic Acid Leaching

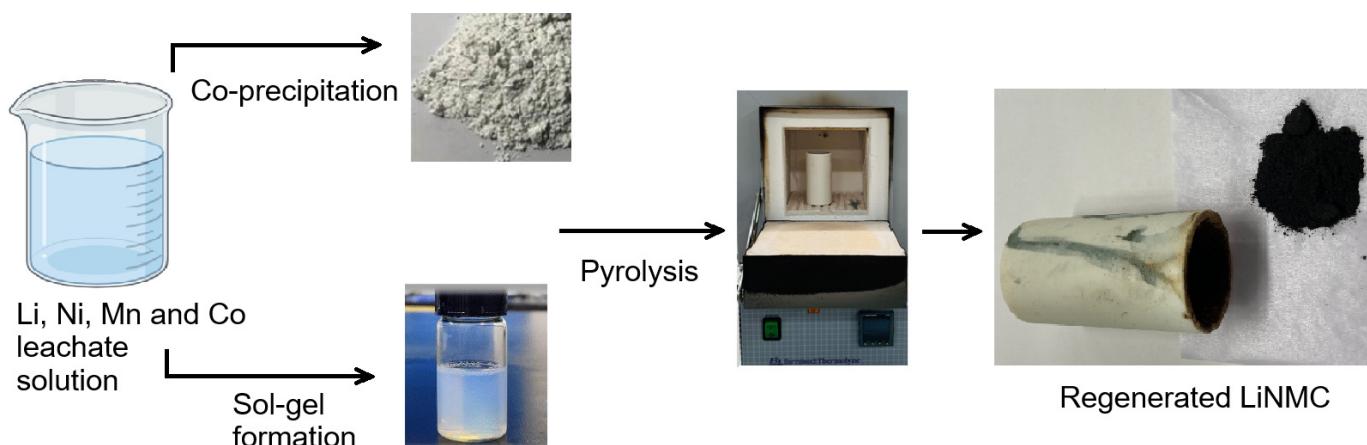
Hydroxy carboxylic acids are effective leaching agents due to their ability to chelate with Li, Ni, Mn, and Co ions through carboxylic and hydroxyl groups [69,80,81]. Hydroxy acids represent a wide class of naturally occurring and synthesized compounds, which have been widely used in biomedical applications, particularly in the cosmetic industry and therapeutic formulations [82]. These compounds are generally classified based on separation between -COOH and -OH groups and also by the number of functional groups such as  $\alpha$ -hydroxy acids,  $\beta$ -hydroxy acids,  $\gamma$ -hydroxy acids as well as polyhydroxy acids and generally produced via microbial reactions and enzymatic or chemical degradation of carbohydrates [82,83]. With the enhanced chelation ability of the hydroxyl group hydroxy poly-carboxylic acid, citric acid has been used without an oxidizing agent for leaching Li, Co, and Ni from lithium cobalt oxide as well as lithium nickel cobalt oxide battery electrode coatings [84,85]. In the LiCoO<sub>2</sub> example, Martins et al. reported quantitative leaching of Li and Co by using 1.0 M aq. citric acid without adding any other reagents [84]. In a comparable case, 80% leaching for Li, Ni, and Co from lithium nickel cobalt oxide is also known with citric acid without any additional reducing agents or reagents [85]. Hydroxy acid chelates of Li, Ni, Mn, and Co are known to form sol-gels at high concentrations, and these sol-gels can be decomposed to leave mixed metal oxides, paving the way for convenient regeneration of active positive electrode materials [86]. Additionally, lithium chelates with lactic acid, glycolic acid, malic acid, and tartaric acid, which are known to decompose at lower temperatures compared to lithium acetate, which lacks the hydroxyl functional group. This observation supports the notion that hydroxy derivatives of acids facilitate the regeneration of active materials by reducing the decomposition temperature [87]. With enhanced chelating ability over simple carboxylic acids and also with ease of regeneration of metal oxides at a lower temperature, several research groups have explored the application of hydroxy carboxylic acid as an emerging technology for recycling LiB waste. A representative list of hydroxy acids investigated as leaching agents, leaching conditions, and percentage yields in recovering LiB metals is provided in Table 1.

**Table 1.** Representative examples of leaching Li, Ni, Mn, and Co from spent LiB electrode coatings using hydroxy carboxylic acid leaching agents, showing acid concentrations, solid/liquid (S/L) ratios, and percentage recovery of metals.

| Hydroxy Carboxylic Acid Leaching Agent                                      | Leaching Conditions   | Percentage Recovery of Li, Ni, Mn and Co (%)         | References |
|---|---|--|------------|
| <br>Lactic acid<br>$pK_a = 4.76$  | 2 M acid, $H_2O_2$ 2 (vol %), $80\text{ }^\circ C$ , 40 min. S/L 10 (g/L)     | Li = 100<br>Ni = 92.8<br>Mn = 94.38<br>Co = 93.37    | [88]       |
|   | 1.5 M acid, $H_2O_2$ 4.60 (vol %), $61\text{ }^\circ C$ , 2 h. S/L 31.2 (g/L) | Li = 100<br>Co = 87.69                               | [80]       |
| <br>Gluconic acid<br>$pK_a = 3.76$  | 2.5 M acid, $80\text{ }^\circ C$ , 2 h. S/L 10 (g/L)                          | Li = 99.85<br>Co = 71.97                             | [89]       |
|   | 1.0 M acid, $H_2O_2$ 1 (vol %), $90\text{ }^\circ C$ , 90 min. S/L 20 (g/L)   | Li = 99<br>Ni = 99<br>Mn = 96<br>Co = 97             | [80]       |
| <br>Glycolic acid<br>$pK_a = 3.83$  | 2.0 M acid, $H_2O_2$ 2 (vol %), $70\text{ }^\circ C$ , 60 min. S/L 10 (g/L)   | Li = 97.54<br>Co = 97.83                             | [90]       |
| <br>Malic acid<br>$pK_{a1} = 3.40$<br>$pK_{a2} = 5.11$                      | 1.5 M acid, $H_2O_2$ 2 (vol %), $90\text{ }^\circ C$ , 40 min. S/L 20 (g/L)   | Li = 100<br>Co > 90                                  | [91]       |
|   | 1.0 M acid, $H_2O_2$ 4 (vol %), $80\text{ }^\circ C$ , 30 min. S/L 5 (g/L)    | Li = 98<br>Ni = 97.8<br>Mn = 97.6<br>Co = 97.3       | [92]       |
|   | 1.2 M acid, $H_2O_2$ 1.5 (vol %), $90\text{ }^\circ C$ , 30 min. S/L 40 (g/L) | Li = 98.9<br>Ni = 95.1<br>Mn = 96.4<br>Co = 94.3     | [93]       |
| <br>Citric acid<br>$pK_{a1} = 3.13$<br>$pK_{a2} = 4.76$<br>$pK_{a3} = 4.76$ | 1.5 M acid, $H_2O_2$ 2 (vol %), $95\text{ }^\circ C$ , 20 min. S/L 20 (g/L)   | Li = 96.2<br>Ni = 93.8<br>Mn = 94.3<br>Co = 89.5     | [94]       |
|   | 2.0 M acid, $H_2O_2$ 3 (vol %), $80\text{ }^\circ C$ , 90 min. S/L 20 (g/L)   | -  | [95]       |
|   | 1.25 M acid, $H_2O_2$ 1 (vol %), $90\text{ }^\circ C$ , 30 min. S/L 20 (g/L)  | Li = 100<br>Co > 90                                  | [96]       |
| <br>Tartaric acid<br>$pK_{a1} = 3.03$<br>$pK_{a2} = 4.37$                   | 2.0 M acid, $H_2O_2$ 4 (vol %), $70\text{ }^\circ C$ , 30 min. S/L 10 (g/L)   | Li = 99.07<br>Ni = 99.31<br>Mn = 99.31<br>Co = 98.64 | [97]       |
| <br>Ascorbic acid<br>$pK_{a1} = 4.17$<br>$pK_{a2} = 11.57$                  | 1M acid, $50\text{ }^\circ C$ , 1 h, S/L 100 (g/L)                            | Li = 88.40<br>Ni = 100<br>Mn = 88.9<br>Co = 85.4     | [98]       |
|   | 1.25 M acid, $70\text{ }^\circ C$ , 20 min. S/L 25 (g/L)                      | Li = 98.5<br>Co = 94.8                               | [68]       |

## 9. Regeneration of LiNMC Without Separation of Metals or Direct Regeneration

The classical and widely studied approach of recycling Li, Ni, Mn, and Co from spent LiB electrode coatings is the separation of individual metals in the form of their pure salts, usually as their nitrates or acetates. Once converted to stable salts, these salts can be repeatedly recrystallized to achieve a high purity like in the original materials used in the preparation of pristine active cathode materials. However, a few research groups have studied the possibility of regeneration of active positive electrode coating materials, such as lithium cobalt oxide (LiCO) and lithium nickel manganese cobalt oxide (LiNMC) without separation of individual metals [99,100]. This novel approach has the certain advantage of elimination of tedious separation of individual metals Li, Ni, Mn, and Co and conversion into their soluble salts like nitrates or acetates. The complexity and the cost of multi-step separation process is the driving force behind the search for simpler regeneration without separation or direct regeneration methods. Two general direct regeneration methods are currently under study, which involves co-precipitation of Li, Ni, Mn, and Co from leachate solution and sol-gel formation from the leachate solution as shown in Figure 4. These two approaches are discussed below under Sections 9.1 and 9.2.



**Figure 4.** Two routes of direct regeneration of LiNMC from a Li, Ni, Mn, and Co leachate solution: co-precipitation and sol-gel formation.

### 9.1. Regeneration Through Co-Precipitation Method

The majority of the regeneration methods described in the literature involves co-precipitation of Ni, Co, and Mn metal ions in the leachate solution, and this method normally leaves Li in the leachate, and incomplete precipitation of Li is another concern; however, some researchers have circumvented this issue by using stepwise precipitation methods. The common precipitation forms described in the recent literature are hydroxides [101–106], carbonates [92,107,108], and oxalates [109–113]. In general, in almost all reported cases, Ni, Co, and Mn content of the leachate solution was adjusted to the desired Ni, Co, and Mn ratio before the co-precipitation as there are losses in the leaching process. Normally, Ni, Co, and Mn acetates are used as supplements to adjust the metal ratios as necessary. At this stage, Li is also invariably added in the form of LiOH or  $\text{Li}_2\text{CO}_3$  to the co-precipitated Ni, Co, and Mn mixture to achieve the final metal composition; it is then dried, pyrolyzed, and sintered to regenerate the lithium nickel manganese cobalt oxide. A selected set of representative examples of the direct regeneration of LiB positive electrode materials through the co-precipitation method is shown in Table 2.

**Table 2.** Direct regeneration of LiB positive electrode materials through the co-precipitation method and their electrochemical performance.

| Leaching Reagents  | Co-Precipitation Reagents and Conditions                              | Formula of the Regenerated Positive Electrode Material                    | Electrochemical Performance: Discharge Capacity (mAh g <sup>-1</sup> ) and Capacity Retention % | Reference |
|--|---|---|---|-----------|
| H <sub>2</sub> SO <sub>4</sub> + H <sub>2</sub> O <sub>2</sub>   | aq. Na <sub>2</sub> CO <sub>3</sub> , NH <sub>4</sub> OH, 50 °C       | LiNi <sub>0.5</sub> Co <sub>0.2</sub> Mn <sub>0.3</sub> O <sub>2</sub>    | 161.4 (0.2 C) 92.4% (0.2 C, 100 cycles)   | [108]     |
| H <sub>2</sub> SO <sub>4</sub> + H <sub>2</sub> O <sub>2</sub>   | aq. Na <sub>2</sub> CO <sub>3</sub> , NH <sub>4</sub> OH, pH 8, 55 °C | LiNi <sub>0.5</sub> Co <sub>0.2</sub> Mn <sub>0.3</sub> O <sub>2</sub>    | 143.5 (1 C) 87% (1 C, 100 cycles)   | [107]     |
| H <sub>2</sub> SO <sub>4</sub> + H <sub>2</sub> O <sub>2</sub>   | aq. Na <sub>2</sub> CO <sub>3</sub>                                   | LiNi <sub>0.33</sub> Co <sub>0.33</sub> Mn <sub>0.33</sub> O <sub>2</sub> | 165.7 (0.1 C) 85.91% (1 C, 200 cycles)  | [104]     |
| H <sub>2</sub> SO <sub>4</sub> + H <sub>2</sub> O <sub>2</sub>   | aq. NaOH, NH <sub>4</sub> OH, pH 10, 50 °C                            | LiNi <sub>0.33</sub> Co <sub>0.33</sub> Mn <sub>0.33</sub> O <sub>2</sub> | 148.8 (0.2 C) 97% (0.2 C, 100 cycles)   | [106]     |
| (NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> + NH <sub>4</sub> OH + Na <sub>2</sub> SO <sub>3</sub> | aq. NaOH, pH 11.2   | LiNi <sub>0.6</sub> Co <sub>0.2</sub> Mn <sub>0.2</sub> O <sub>2</sub>    | 151.1 (0.5 C) 92.19% (0.5 C, 100 cycles)  | [103]     |
| H <sub>3</sub> PO <sub>4</sub> + Citric acid   | aq. Oxalic acid, NH <sub>4</sub> OH, pH 1.98                          | LiNi <sub>0.5</sub> Co <sub>0.2</sub> Mn <sub>0.3</sub> O <sub>2</sub>    | 135.35 (1 C) 85.45% (1 C, 100 cycles)   | [110]     |
| H <sub>2</sub> SO <sub>4</sub> + Citric acid   | aq. Oxalic acid, NH <sub>4</sub> OH                                   | LiNi <sub>0.6</sub> Co <sub>0.2</sub> Mn <sub>0.2</sub> O <sub>2</sub>    | (0.1 C) 86% (1 C, 100 cycles)   | [109]     |
| DL-Malic acid + H <sub>2</sub> O <sub>2</sub>  | aq. Na <sub>2</sub> CO <sub>3</sub> , pH 8–9, 60 °C                   | LiNi <sub>0.6</sub> Co <sub>0.2</sub> Mn <sub>0.2</sub> O <sub>2</sub>    | 168.3 (0.1 C), 91.9% (1 C, 50 cycles)   | [92]      |
| Oxalic acid  | aq. Na <sub>2</sub> CO <sub>3</sub> , then oxalic acid, 80 °C         | LiNi <sub>0.33</sub> Co <sub>0.33</sub> Mn <sub>0.33</sub> O <sub>2</sub> | 168 (0.2 C), 91.5% (0.2 C, 150 cycles)  | [112]     |
| Ascorbic acid  | aq. Na <sub>2</sub> CO <sub>3</sub> , 90 °C, then oxalic acid         | LiNi <sub>0.33</sub> Co <sub>0.33</sub> Mn <sub>0.33</sub> O <sub>2</sub> | 164.9 (0.2 C), 91.3% (0.2 C, 100 cycles)  | [113]     |
| H <sub>2</sub> SO <sub>4</sub> + H <sub>2</sub> O <sub>2</sub>   | aq. NaOH, 25 °C   | LiFePO <sub>4</sub>   |   | [114]     |
| H <sub>2</sub> SO <sub>4</sub> + H <sub>2</sub> O <sub>2</sub>   | aq. NaOH, NH <sub>4</sub> OH, 50 °C                                   | LiNi <sub>0.5</sub> Co <sub>0.2</sub> Mn <sub>0.3</sub> O <sub>2</sub>    | 173.9 (0.2 C), 91.9% (0.5 C, 100 cycles)  | [115]     |
| Fruit peel lixiviant   | aq. Ammonium oxalate  | LiCoO <sub>2</sub>  | 98 (0.67 C), 93% (0.67 C, 140 cycles)   | [116]     |

The regeneration without separation is known with Ni, Co, and Mn in the leachate solutions prepared from mineral acid as well as with organic acid leachates [117]. In one of the mineral acid leaching examples using sulfuric acid together with hydrogen peroxide, Zheng and co-workers [106] adjusted the molar ratio of Ni, Co, and Mn in the leachate solution to 1:1:1 by adding the corresponding metal ions as sulfates. The metals were then co-precipitated as their hydroxides by adding a mixture of aq. sodium hydroxide and ammonium hydroxide [106]. The mixed hydroxide LiNi<sub>0.33</sub>Co<sub>0.33</sub>Mn<sub>0.33</sub>(OH)<sub>2</sub> was filtered, dried, pyrolyzed, and sintered to give a regenerated LiNi<sub>0.33</sub>Co<sub>0.33</sub>Mn<sub>0.33</sub>O<sub>2</sub> product. Furthermore, Zheng et al. have observed a well-developed layered structure comparable to pristine material in regenerated materials, and the discharge capacities of regenerated LiNMC reached 150.6, 148.8, 141.6, 132.9, and 120.5 mAh g<sup>-1</sup> at 0.1 C, 0.2 C, 0.5 C, 1 C, and 2 C, respectively, with acceptable charge/discharge and cycling characteristics [106]. The LiNMC re-synthesis is known from a basic medium leaching solution as well. In this instance, Zhu and co-workers used an aqueous NH<sub>3</sub>-(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>-Na<sub>2</sub>SO<sub>3</sub> system as the leaching medium on LiB black mass [103]. Then, metal molar ratios were adjusted to the formula LiNi<sub>0.6</sub>Co<sub>0.2</sub>Mn<sub>0.2</sub>O<sub>2</sub> by adding appropriate amounts of metal salts; afterwards, ammonia was evaporated, leaving the mixed metal hydroxide. In addition, they have been able to control the ammonia concentration to avoid stable complexation that may hinder the precipitation. The basic media leaching has the advantage of avoiding neutralization, and the regenerated LiNi<sub>0.6</sub>Co<sub>0.2</sub>Mn<sub>0.2</sub>O<sub>2</sub> material displayed good discharge capacities of 178.6, 171.7, 148.5, and 120.3 mAh g<sup>-1</sup> at 0.1 C, 0.2 C, 0.5 C, and 1.0 C, respectively. Furthermore, Zhu and co-workers reported the capacity retention rate as 92.19% after 100 charge/discharge cycles at 0.5 C [103].

In another example, Ning and co-workers used 1.0 M aq. DL-malic acid together with 4 vol % hydrogen peroxide as the leaching medium under ultrasonic irradiation at 80 °C for 30 min, achieving leaching efficiencies 98.0, 97.8, 97.3, and 97.6% for Li, Ni, Mn, and Co, re-

spectively. As in the earlier examples, the molar ratio of Ni, Mn, and Co was adjusted to 3:1:1 by adding acetate salts:  $\text{CH}_3\text{COONi}\cdot 4\text{H}_2\text{O}$ ,  $\text{CH}_3\text{COOMn}\cdot 4\text{H}_2\text{O}$ , and  $\text{CH}_3\text{COOCo}\cdot 4\text{H}_2\text{O}$ . Then, Ning and co-workers added 0.6 M aqueous  $\text{Na}_2\text{CO}_3$  to the leachate solution while maintaining the pH at 8–9 for the co-precipitation of metals as their carbonates. The precipitate was filtered out and mixed with  $\text{LiOH}\cdot \text{H}_2\text{O}$  at a Li/M (M = sum of Ni, Co and Mn) molar ratio of 1.05 to compensate for the lithium loss. Then, the mixture was dried and pyrolyzed in air at 800 °C for 15 h to produce regenerated  $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ . In a performance test, the regenerated  $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$  showed a specific capacity of 168.32  $\text{mAh g}^{-1}$  at the first discharge at 0.1 C, and the capacity retention was 85.0% after 50 cycles [92]. Zhang and co-workers have developed an interesting approach for leaching and precipitation of metals from LiB waste using a single reagent, where oxalic acid was used as the leaching agent as well as the precipitation reagent for Ni, Mn, and Co [112]. Lithium dissolved into the solution and the transition metals precipitated as oxalates could be recovered upon cooling. Then, a stoichiometric amount of  $\text{Li}_2\text{CO}_3$  was added to the oxalate precipitate and directly calcined to regenerate  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$  [112]. The regenerated positive electrode material produced after 10 min of leaching time showed the highest electrochemical performances, delivering the peak initial discharge capacity of 168  $\text{mAh g}^{-1}$  at 0.2 C and 153.7  $\text{mAh g}^{-1}$  after 150 cycles, with a retention of 91.5% [112].

Shang and co-workers used a mixture of inorganic and organic acids for leaching, and after adjustment of metal ratios, all four metals were precipitated together by adding a mixture of oxalic acid and ammonium hydroxide [109]. The regenerated positive electrode material,  $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ , delivered an original specific capacity of 180.6  $\text{mAh g}^{-1}$  at 0.1 C [109]. An 86% capacity retention was accomplished after 100 cycles at 1.0 C. Furthermore, Shang et al. have noted the importance of controlling the pH of the medium in the oxalic acid co-precipitation procedure to avoid the dissolution of the generated manganese oxalate at a low pH [109]. A few researchers have noted that electrochemical performance of regenerated positive electrode material is strongly influenced by size and morphology of the precursor particles of mix metal precipitate, and they have paid attention to this important consequence [118]. As Zheng and his co-workers reported, all hydroxides or carbonates may not precipitate at the same time in co-precipitation techniques. In addition, they have proposed a mechanism for co-precipitate particle formation based on their experiments. According to their explanation, the primary particles of hydroxides or carbonate agglomerate into porous secondary particles in the initial stages. These uneven secondary particles may lead to spherical particles in the next stage due to collisions and inter-particle friction. The spherical particles may continue to grow uniformly keeping the spherical shape. However, at the end strong hydrodynamic shear forces stabilizes the growth rate of larger particles, where the shape and size of the secondary particles slowly even out resulting in the co-precipitate of the mixed metal hydroxides or carbonates. As described in Zheng and co-workers' precipitation mechanism, the morphology, particle size distribution, and tap density of the mixed metal hydroxide co-precipitate generally stabilizes after about 48 h. Furthermore, Zheng and co-workers claimed that as a result of this even co-precipitate particle formation, regenerated  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$  exhibited even better electrochemical performance than the commercial pristine material at high charge/discharge rates of 5 C and 10 C [118]. An additional critical element influencing the electrochemical functioning of the regenerated LiB positive electrode materials is the purity of precursors. Even though additional elements or impurities such as Al, Cu, and Fe are unavoidably present in the primary leachate solution, many research reports claim that the amounts of the impure metal ions in the leachate solution and precursors are very low or insignificant after reasonable pretreatments [92,110,118]. Therefore, impurities would

generally not significantly impact the electrochemical performances of the regenerated positive electrode material samples [110,118].

### 9.2. Regeneration Through Mixed Metal Sol–Gel Method

The sol–gel method is an alternative to co-precipitation and is gaining attention in direct regeneration of positive electrode materials from leachate solutions [88,119,120]. One or more chelating agents are required in the sol–gel method to assist the uniform mixing of metal ions and the formation of homogeneous sol–gels [100]. Polycarboxylic acids, hydroxy acids, polyols, and amines chelating with Li, Ni, Mn, and Co ions have been used to prepare mixed metal sol–gels as an alternative to co-precipitates [121,122]. As in precipitation methods, the metal ratios can be adjusted to the required stoichiometry before the sol–gel formation. The chelating agents are later added to the leachate solution or can be the same as the reagent used in the leaching process. Adjusting and maintaining the appropriate pH is critical for complexation, and this is an important consideration for the sol–gel formation; undisturbed chelate solutions or sol–gel precursors are normally allowed to stand at room temperature or higher in closed containers for the sol–gel formation process. Then, the sol–gel is dried at around 100–105 °C to remove water, pyrolyzed, and sintered to produce the regenerated cathode material. The main advantage is that organic chelate can be burned off, regenerating the positive electrode material [88,119–122]. Selected examples of regeneration of LiB positive electrode materials through sol–gel formation, pyrolysis route, and their electrochemical performance are shown in Table 3.

**Table 3.** Regeneration of LiB positive electrode material through sol–gel method and their electrochemical performance.

| Leaching Reagents   | Sol–Gel Formation Reagents and Conditions          | Formula of the Regenerated Positive Electrode Material                                       | Electrochemical Performance: Discharge Capacity (mAh g <sup>−1</sup> ); Capacity Retention | Reference |
|---|--|--|--|-----------|
| DL-Malic acid + H <sub>2</sub> O <sub>2</sub>   | aq. NH <sub>4</sub> OH, pH 8, 80 °C                | LiNi <sub>0.33</sub> Co <sub>0.33</sub> Mn <sub>0.33</sub> O <sub>2</sub>                    | 147.2 (0.2 C), 95.06% (0.5 C, 100 cycles)  | [120]     |
| Lactic acid + H <sub>2</sub> O <sub>2</sub>   | aq. NH <sub>4</sub> OH, pH 7, 80 °C                | LiNi <sub>0.33</sub> Co <sub>0.33</sub> Mn <sub>0.33</sub> O <sub>2</sub>                    | 143.9 (0.5 C), 96% (0.5 C, 100 cycles)   | [88]      |
| Citric acid + H <sub>2</sub> O <sub>2</sub>   | aq. NH <sub>4</sub> OH, pH 8, 80 °C                | LiNi <sub>0.33</sub> Co <sub>0.33</sub> Mn <sub>0.33</sub> O <sub>2</sub>                    | 147 (1 C), 93% (1 C, 50 cycles)  | [119]     |
| Lactic + Glycolic   | Evaporation, 100 °C                                | LiNi <sub>0.03</sub> Mn <sub>0.02</sub> Co <sub>0.11</sub> O <sub>0.30</sub>                 | -  | [86]      |
| HNO <sub>3</sub> + H <sub>2</sub> O <sub>2</sub> + glucose                            | aq. Citric acid, glucose or sucrose, 80 °C         | LiNi <sub>0.6</sub> Co <sub>0.2</sub> Mn <sub>0.2</sub> O <sub>2</sub>                       | 176.9 (0.2 C), 95.8% (0.2 C, 50 cycles)  | [122]     |
| NH <sub>4</sub> OH + NH <sub>4</sub> HCO <sub>3</sub> + H <sub>2</sub> O <sub>2</sub> | aq. Citric acid, NH <sub>4</sub> OH, pH 7–8, 80 °C | Li <sub>1.2</sub> Mn <sub>0.54</sub> Ni <sub>0.13</sub> Co <sub>0.13</sub> O <sub>2</sub>    | 246.5 (0.1 C), 66.2% (0.1 C, 80 cycles)  | [121]     |
| DL-Malic acid + H <sub>2</sub> O <sub>2</sub>   | aq. NH <sub>4</sub> OH, pH 8, 85 °C                | LiNi <sub>0.04</sub> Co <sub>0.33</sub> Mn <sub>0.33</sub> Al <sub>0.04</sub> O <sub>2</sub> | 148.1 (0.2 C), 90% (0.2 C, 100 cycles),  | [123]     |
| Citric acid + H <sub>2</sub> O <sub>2</sub>   | aq. NH <sub>4</sub> OH, pH 6.5, 90 °C              | LiCoO <sub>2</sub>   | 120 ± 8 (0.2 C), 93% (0.2 C, 20 cycles)  | [124]     |

The sol–gel method can be applied in combination with mineral acid leaching or with organic acid leaching. For instance, Li et al. demonstrated the sol–gel pyrolysis regeneration technique on a nitric acid leachate of metal ions [122]. In addition, they studied the effect of chelating agent structure on the performance of the regenerated LiNMC [122]. In this case, waste lithium nickel manganese cobalt positive electrode materials were dissolved in nitric acid hydrogen peroxide solution to leach out all metal ions. Then, the Li, Ni, Co, and Mn concentrations in the leachate were analyzed and the Li:Ni:Co:Mn mole ratio was adjusted to the desired 1.05:0.6:0.2:0.2. ratio by adding Li<sub>2</sub>CO<sub>3</sub>, Ni(NO<sub>3</sub>)<sub>2</sub>, Mn(NO<sub>3</sub>)<sub>2</sub>, and Co(NO<sub>3</sub>)<sub>2</sub>. Next, a mixture of citric acid, glucose, or sucrose was added as a gelling agent, and the pH was adjusted to 6.0 using ammonia. The resulting viscous gel was dried at 120 °C for 12 h,

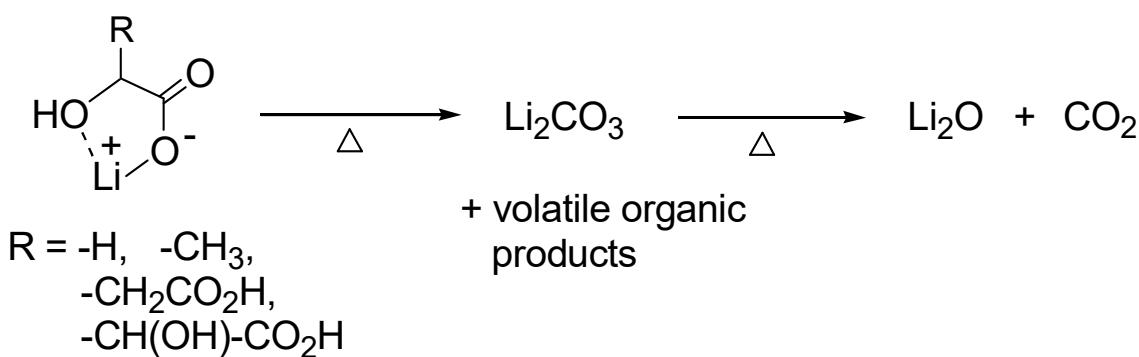
subsequently pyrolyzed, and calcined to produce the regenerated  $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ . This regenerated positive electrode material showed an original discharge capacity of  $176.9 \text{ mAh g}^{-1}$ , original coulombic efficiency of 87.0%, and after 50 cycles, the discharge capacity retention rate was 95.8% at 0.2 C [122]. Furthermore, Li and co-workers have noted that the best electrochemical performance could be obtained when D-glucose was used as a gelling agent, due to minimum cation mixing, relatively complete hexagonal crystal structure, minimum impedance, and the best reversibility of the redox reaction [122]. Ammonia leaching using a mixture of  $\text{NH}_4\text{OH}$ ,  $\text{NH}_4\text{HCO}_3$ , and  $\text{H}_2\text{O}_2$  was also reported as an alternative to acid leaching [122]. Then, the metal-containing leaching solution was subjected to the sol–gel process; afterwards, pyrolysis and sintering were used to produce the regenerated  $\text{Li}_{1.2}\text{Mn}_{0.54}\text{Ni}_{0.13}\text{Co}_{0.13}\text{O}_2$  positive electrode material [122].

In one of the early attempts of the sol–gel approach, Li and co-workers used a mixture of 1.5 M lactic acid together with 0.5 vol %  $\text{H}_2\text{O}_2$  as the leaching agent with a solid/liquid ratio of  $20 \text{ g L}^{-1}$ , at  $70^\circ\text{C}$ , for leaching Li, Ni, Mn, and Co from cathode coatings. Under the optimized conditions, they reported high leaching efficiencies of 97.7, 98.2, 98.9, and 98.4% for Li, Ni, Co, and Mn, respectively. Next, Li and co-workers used the leachate solution for the re-synthesis of  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$ . In preparation of the regenerative synthesis, the  $\text{Li}^+$ ,  $\text{Ni}^{2+}$ ,  $\text{Co}^{2+}$ , and  $\text{Mn}^{2+}$  molar ratio was adjusted to 3.05:1:1:1 by adding stoichiometric amounts of pure salts,  $\text{CH}_3\text{COOLi}\cdot 2\text{H}_2\text{O}$ ,  $(\text{CH}_3\text{COO})_2\text{Ni}\cdot 4\text{H}_2\text{O}$ ,  $(\text{CH}_3\text{COO})_2\text{Co}\cdot 4\text{H}_2\text{O}$ , and  $(\text{CH}_3\text{COO})_2\text{Mn}\cdot 4\text{H}_2\text{O}$ . Next, the pH was adjusted to 7 with ammonium hydroxide and heated at  $80^\circ\text{C}$  to form a transparent sol–gel, which was dried at  $100^\circ\text{C}$  for 24 h, followed by heating at  $450^\circ\text{C}$  for 5 h and calcination at  $900^\circ\text{C}$  for 12 h, producing the regenerated  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$ . Electrochemical performance testing showed that the regenerated  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$  positive electrode materials deliver a highly reversible discharge capacity,  $138.2 \text{ mAh g}^{-1}$  at 0.5 C, with a 96% capacity retention after 100 cycles, which is comparable to the freshly synthesized  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$  [88].

Hydroxy acids are excellent reagents for the dual role of leaching and sol–gel formation through their documented chelating ability with Li, Ni, Mn, and Co ions [87,125–127]. For example, Yao and coworkers used D,L-malic acid as the leaching agent as well as the chelating ligand in the following sol–gel method based regeneration of  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$  [120]. After adjusting the metal ion ratio and pH of leachate, a D,L-malic acid chelate of the metal mixture was formed and then pyrolyzed to regenerate  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$ . The electrochemical tests showed the initial charge and discharge capacities of the regenerated material as  $152.9 \text{ mAh g}^{-1}$  and  $147.2 \text{ mAh g}^{-1}$  ( $2.75\text{--}4.25 \text{ V}$ , 0.2 C), respectively. The capacity retention at the 100th cycle remains 95.06% of the original value ( $2.75\text{--}4.25 \text{ V}$ , 0.5 C) [120]. Aqueous lactic acid with 0.5 vol % hydrogen peroxide was the organic acid of choice in the studies by Li and co-workers, in which they reported leaching efficiencies of Li, Ni, Mn, and Co as high as 97.7, 98.2, 98.4, and 98.9%, respectively [88]. The ideal leaching conditions were lactic acid concentration of  $1.5 \text{ mol L}^{-1}$ , a solid/liquid ratio of  $20 \text{ g L}^{-1}$ , a leaching temperature of  $70^\circ\text{C}$ , and a time of 20 min. The Li:Ni:Mn:Co molar ratio was then adjusted to 3.05:1:1:1 by adding acetate salts:  $\text{CH}_3\text{COOLi}\cdot 2\text{H}_2\text{O}$ ,  $(\text{CH}_3\text{COO})_2\text{Ni}\cdot 4\text{H}_2\text{O}$ ,  $(\text{CH}_3\text{COO})_2\text{Mn}\cdot 4\text{H}_2\text{O}$ , and  $(\text{CH}_3\text{COO})_2\text{Co}\cdot 4\text{H}_2\text{O}$ . Next, the pH was adjusted to 7 by adding ammonium hydroxide, and the resulting solution was heated to  $80^\circ\text{C}$  for the sol–gel formation [88]. The dried gel was pyrolyzed at  $450^\circ\text{C}$  for 5 h and then calcined in air at  $900^\circ\text{C}$  for 12 h to yield the regenerated  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$  positive electrode material. This regenerated LiNMC delivered a highly reversible discharge capacity of  $138.2 \text{ mAh g}^{-1}$  at 0.5 C and 96% capacity retention after 100 cycles. Furthermore, Li and co-workers claimed that these values are comparable to those of freshly synthesized  $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$  [88]. The use of a glycolic and lactic acid mixture is also known, and the synergistic effect of two acids were reported recently in recovering and regenerating LiNMC from spent Li-ion

laptop batteries by using the sol–gel process followed by pyrolysis [86]. In this instance, an aq. mixture of 0.35 M lactic and 0.15 M glycolic acid displayed the highest leaching efficiencies of 100, 100, 100, and 89% for Li, Ni, Mn, and Co, respectively, for a leaching experiment carried out at 120 °C for 6 h. Afterward, the leachate solution was concentrated by evaporation of the leachate at 100 °C using a steam bath to produce a chelate gel. Which was dried in an oven at 105 °C, and then pyrolyzed at 800 °C for 15 h; burning off organic hydroxy acids, regenerating LiNMC [86]. The Li, Ni, Mn and Co metal ratio in the regenerated LiNMC was comparable to the metal ratio in the initial combined electrode coating material used for leaching and displayed similar powder X-ray diffractograms, signifying the suitability of  $\alpha$ -hydroxy carboxylic acids such as lactic and glycolic acid mixtures as leaching and sol–gel formation agents in regeneration of positive electrode material via pyrolysis of the dried chelate sol–gel [86].

The  $\alpha$ -hydroxy carboxylic acid chelates of positive electrode material metals are known to decompose at temperatures much lower than their metal carboxylates, allowing facile recovery of the metal as metal oxides by pyrolysis of the  $\alpha$ -hydroxy carboxylic acid gels [87]. The same research group studied the thermal decomposition process of a series of selected lithium chelates of  $\alpha$ -hydroxy acids: glycolic, lactic, malic, and tartaric acids and compared them with decomposition of lithium acetate ( $\text{CH}_3\text{CO}_2\text{Li}$ ) to study pyrolysis as an effective metal recovery method from  $\alpha$ -hydroxy carboxylic acid chelates [87]. In thermogravimetric analysis (TGA), the glycolic, lactic, malic, and tartaric acid chelates studied showed lower initial decomposition temperatures in the range of 313–356 °C, in comparison to  $\text{CH}_3\text{CO}_2\text{Li}$ , with an initial decomposition temperature of 444 °C [87]. The Li  $\alpha$ -hydroxy acid chelate pyrolysis residue formed above 300 °C was identified as lithium carbonate ( $\text{Li}_2\text{CO}_3$ ). Further heating of the  $\text{Li}_2\text{CO}_3$  residue above 700 °C resulted in a decomposition of  $\text{Li}_2\text{CO}_3$  to lithium oxide ( $\text{Li}_2\text{O}$ ) as shown in Figure 5. This process allows the recovery of lithium as  $\text{Li}_2\text{CO}_3$  or  $\text{Li}_2\text{O}$  as needed. Furthermore, facile decomposition of  $\alpha$ -hydroxy carboxylic acid metal chelates were explained as a result of their favorable decomposition pathways [87]. This easy decomposition of metal hydroxy carboxylate chelates to metal oxides at lower temperatures is a certain advantage of using  $\alpha$ -hydroxy carboxylic acids, in addition to their ability to behave as good leaching reagents complexing with LiB metal ions under mild leaching conditions.



**Figure 5.** Pyrolysis of Li  $\alpha$ -hydroxy carboxylic acid chelates of glycolic, lactic, malic, and tartaric acids to produce lithium carbonate ( $\text{Li}_2\text{CO}_3$ ) and lithium oxide ( $\text{Li}_2\text{O}$ ). Reprinted with permission from ref. [87]. Copyright 2024 Elsevier.

## 10. Current Challenges and Future Directions

Lithium ion battery recycling is an essential component in decarbonization through transformation into renewable energy technologies and electrical energy storage. A number of established recycling companies and startups have already started recycling LiBs on a limited scale, realizing the vast commercial potential in this industry [51]. Various pyromet-

allurgy and hydrometallurgy-based technologies are discussed in this review emphasizing the advantages of hydroxy acid-based leaching and direct regeneration methods. This approach has distinct benefits over other recycling technologies such as pyrometallurgy and classical hydrometallurgy with separation of individual metals as salts. Lower carbon number organic acids, particularly formic acid and acetic acid, have demonstrated efficacy in the leaching process in early studies. However, in later studies, a number of researchers have pointed out the advantages of using hydroxy acids over simple carboxylic acids. Specifically, a-hydroxy acids with a bidentate chelating ability can form more stable complexes with critical metals at optimal pH and temperatures, with significantly improved leaching efficiencies. Furthermore, multidentate chelating agents such as citric and malic acids also have shown much promise in leaching out metals and gel formation allowing easy regeneration of active cathode materials. The most remarkable benefits are its simplicity, excellent recovery, low energy consumption, as well as the alignment with green chemistry principles. Although several recent review publications have discussed the use of organic acids in LiB recycling, there is a notable absence of a comprehensive review focusing on the use of hydroxy acids in leaching and direct regeneration; moreover, there are certain challenges in this breakthrough technology, and some of the workable solutions are discussed in the follow-up section.

#### *Challenges in Hydroxy-Acid Leaching—Direct Regeneration Technology and Solutions*

- (1) Battery chemistries: Even within the same type or class of batteries, there are wide variations in battery chemistries in currently used LiBs. Sorting batteries based on chemistry is a major challenge in direct regeneration technology. Processing LiBs of similar Li, Ni, Mn, and Co compositions is an important consideration for achieving high leaching efficiencies in a batch process, where adjusting the parameters like selection of the hydroxy acid, pH, acid concentration, solid loading, temperature, and time is mandatory in achieving the optimum leaching efficiency. In addition, a batch of spent batteries with the same Li, Ni, Mn, and Co compositions processed together will give leachate solution reflecting the original composition and can be used to regenerate LiNMC of the matching composition without significant adjustments of metal concentrations. Better labeling and classification systems and reliable composition data from battery manufacturers can help in resolving some of these issues. However, these composition data can be trade secrets of battery manufacturers, and they may be reluctant to disclose the compositions due to stiff competition in the field.
- (2) Disassembly of spent LIBs: Separation of positive electrode and negative electrode films without mixing coatings and contamination with other metals such as aluminum and copper in current collectors and iron in casing are major challenges. Mechanical separation of each of these components require complex machines and skilled handling of components. Standardization of battery design is one possible solution. In addition, designing batteries with recyclability in mind is an important consideration for future battery designs. Disassembly of electrodes without contamination and damage to electrode foils can be the way to recover copper and aluminum, generating a secondary income stream for the recycling industry.
- (3) Custom selection of hydroxy acids and leaching parameters: The structure, geometry,  $pK_a$  of acids, and  $K_a$  of complexes are crucial factors to consider when choosing hydroxy acid or acids for leaching positive electrode materials. The leaching mechanisms as well as technical details such as the effects of reaction parameters like pH, acid concentration, and temperature required to achieve optimal metal leaching efficiency are still not well-studied areas that need in-depth studies. Furthermore, a combination

of hydroxy acids may offer an effective approach for recovering these materials, giving improved yields due to synergistic effects of hydroxy acids. For instance, a mixture of lactic and glycolic acids has been shown to be an effective leaching agent without the need for a reducing agent. Likewise, there are many hydroxy acids that have yet to be explored as leaching agents, and further research on new hydroxy acids and mixtures may reveal an optimized leaching system for each type of LiB chemistry.

- (4) Leaching of Co: Reducing agents like  $H_2O_2$  or ascorbic acid are frequently added to the leaching solution to reduce Co(III) to Co(II), since Co(III) compounds are mostly insoluble. This is a critical consideration for Co leaching. However, there is a handful of reports on hydroxy acid leaching of Co without reducing agents. Further research into Co leaching may help in the development of efficient systems to minimize the leaching reagent costs.
- (5) Adjusting metal ratio: In most reported work on direct regeneration through precipitation or sol–gel formation methods, the Li:Ni:Mn:Co ratios are adjusted to the desired ratio by adding pure acetate or nitrate salts of these metals before regeneration. Adjusting the metal ratios by adding supplements can be minimized or altogether eliminated by choosing one type or class of spent batteries, optimizing the leaching conditions to achieve high leaching yields. This technique can minimize or avoid the use of expensive pure salts, regenerating valuable positive electrode materials in minimum operations.
- (6) Precipitate structure: In the precipitation technique, the particle size and shape of precipitate can affect the layered structure of LiNMC and the electrochemical properties such as capacity and charge discharge characteristics. This is still an unexplored research area that requires detailed studies to regenerate high quality LiNMC from spent batteries.
- (7) Processing sol–gel: Pyrolysis of metal hydroxides, carbonates, and carboxylates is well known in the synthesis of metal oxides and mixed metal oxides. Drying and pyrolysis of mixed metal chelate gels is an unexplored area of interest, where the gel structure, pyrolysis temperature, kinetics, and atmosphere may influence the structure and performance of regenerated materials in the direct regeneration process. In a recent study on comparison of decomposition kinetics of lithium acetate with lithium hydroxy acid chelates, the hydroxy acid chelates were found to decompose at a much lower temperatures in comparison to the corresponding acetate. This finding has consequences in the development of an efficient mixed metal hydroxy acid chelate pyrolysis technique in easy direct regeneration of mixed metal oxide LiB cathode materials. Further research on metal–hydroxy acid gel pyrolysis kinetics is required in optimization of this vital regeneration technique.

The highlighted technique should be applicable as a general method for all types of batteries such as LCO, NCM, and LFP. However, the hydroxy acid used and the leaching conditions should be customized to each type of battery, and it is recommended to process only one type of batteries at a time in the batch process. This is a practical challenge in large-scale industrial applications; nevertheless, it may be resolved by proper labeling, identification, and sorting of batteries based on their chemistries. The reviewed hydroxy acid-based green chemical recycling processes without adding extra chemicals like reducers, minimizing adjustments of metal ratio with supplementary salts, and the direct regeneration of active positive electrode materials without separation of metals, have certain advantages over conventional spent lithium-ion battery recycling technologies. More importantly, this straightforward technology has the potential to develop into a profitable industry. Overall, this review concludes that the use of hydroxy acid chelating agents and direct regeneration offer a promising environmentally friendly approach to recycle spent

lithium-ion batteries; nevertheless, more research is needed to optimize these processes and realize their full potential.

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