

Review

Recycling of Lithium-Ion Batteries via Electrochemical Recovery: A Mini-Review

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Abstract: With the rising demand for lithium-ion batteries (LIBs), it is crucial to develop recycling methods that minimize environmental impacts and ensure resource sustainability. The focus of this short review is on the electrochemical techniques used in LIB recycling, particularly electrochemical leaching and electrodeposition. Our summary covers the latest research, highlighting the principles, progress, and challenges tied to these methods. By examining the current state of electrochemical recovery, this review intends to provide guidance for future advancements and enhance LIB recycling efficiency.

Keywords: lithium-ion battery; battery recycling; electrochemical recovery; electrochemical leaching; electrodeposition

1. Introduction

Lithium-ion batteries play a crucial role in advancing the development of electric vehicles and consumer electronic devices. As these markets expand, there is a growing demand for lithium-ion batteries, emphasizing their essential role in modern technology and sustainable energy systems [1–3]. However, the increasing reliance on lithium-ion batteries brings challenges related to natural resource limitations and sustainable development. The limited availability of natural resources, such as lithium, cobalt, and nickel, necessitates the development of efficient and low-cost recycling processes [4–6]. These processes are crucial for recovering valuable materials, with a particular emphasis on cathode materials, which are essential components of lithium-ion batteries. Currently, lithium-ion battery recycling is still in its early stages at the industrial scale, with significant challenges related to the quality of regenerated materials and the cost of the process. Recycling methods for lithium-ion batteries are typically classified into pyrometallurgical, hydrometallurgical, and direct recycling processes [4,7]. Within these categories, various techniques have been applied for cathode recycling, including acid leaching [8–10], direct delamination [11–13], solvent extraction [14], hydrothermal treatment [15], membrane separation [16,17], and electrochemical method [18–20]. Among these approaches, the electrochemical method is gaining attention because of its potential for energy efficiency, scalability, and selectivity and reduced environmental impact with less harmful emissions and waste products.

The electrochemical method for battery recycling uses electrochemical reactions to recover critical metals from battery scraps and end-of-life batteries. Recent advancements in the electrochemical recovery of lithium-ion batteries are divided into two main approaches: electrochemical leaching and electrodeposition [21–23]. For electrochemical leaching, the electric current is applied to the battery materials, thus achieving the dissolution of metal ions in the solution. As for electrodeposition, the electric current is applied to the solution with dissolved metal ions and promotes the reduction of the dissolved metal ions, thus enabling the recovery of high-purity metals on the electrode. Electrodeposition was applied for pure metal recovery, the separation of metals, and cathode relithiation. In this work, we summarize the most recent advancements in electrochemical leaching and electrodeposition



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for cathode material recycling. Additionally, we provide insights into the challenges of, potential improvements in, and future directions for enhancing the efficiency and sustainability of these electrochemical recycling methods.

2. Electrochemical Leaching

Electrochemical leaching is the extraction of metal ions from the anode electrode into the solution through an external current. Currently, inorganic acid combined with hydrogen peroxide for cathode leaching is the most commonly used method for lithium-ion battery recycling. However, the usage of inorganic acid with hydrogen peroxide involves the usage of a large amount of water and leads to a complicated separation and precipitation process. Electrochemical leaching provides an alternative method for effectively extracting metal ions from the cathode electrode in a greener process [24–29]. Recently, Adhikari et al. conducted a lifecycle assessment to compare the electrochemical leaching process with the traditional hydrometallurgical leaching method. The results showed that the electrochemical leaching process greatly relieved the environmental impact by 80–87%, owing to its reduced usage of acid and hydrogen peroxide [30]. Table 1 provides a summary of recent research studies on electrochemical leaching for cathode recycling. In 2020, Diaz et al. proposed an electrochemical method for cathode leaching to recover valuable metals from mixed cathodes. In this work, the three-electrode system was used with nickel foam as the anode electrode, a stainless-steel mesh as the cathode electrode, and Ag/AgCl as the reference electrode. KOH solution (1 M) was used as an anolyte and a bipolar membrane was used for separating the anode and cathode. The researchers applied this electrochemical leaching process to an industry LIB metal oxide filter cake (MOFC), which is composed of mixed anode graphite and different cathode materials, including LiCoO_2 , $\text{LiMn}_x\text{Co}_y\text{O}_2$, $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$, $\text{LiCu}_x\text{MnO}_2$, and $\text{LiNi}_x\text{Co}_x\text{Al}_z\text{O}_2$. The MOFC was mixed in sulfuric acid (H_2SO_4) solution (0.5–2 M) with a low concentration of FeSO_4 (10 mM) as a reducing agent. The electrochemical leaching was operated at a controlled voltage of -0.3 V vs. Ag/AgCl. During the process, the water was oxidized to produce protons and hydroxyl ions. The produced protons were transferred to the cathode and facilitated the leaching process, while the hydroxyl ions were transferred to the anode for oxygen evolution in alkaline media. With this electrochemical design, the leaching efficiencies of active metals, including Li, Ni, Mn, and Co, reached over 96%. The economic analysis showed that the energy and chemical costs of this process are about 80% less than that of the conventional hydrometallurgical process [31]. In 2021, Lei et al. applied an electrochemical method for co-leaching the Li, Ni, Mn, and Co from a reduced $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ (NMC) cathode. Before the electrochemical leaching process, the NMC cathode materials were first annealed at 1600°C for 120 mins to obtain the reduced cathode mixture, which included Li_2CO_3 , NiO , Co_3O_4 , Mn_2O_3 , and MnO_2 . The mixture was kept in a polypropylene bag and immersed in a sulfuric acid electrolyte. In this work, a two-electrode system was used with titanium as an anode and graphite as the cathode. An anion exchange membrane was set in the middle for separating the cathode and anode electrodes. The H_2SO_4 concentration, electric current, and reaction time were adjusted and optimized. The leaching efficiencies of the metal ions showed a trend of first increasing and then decreasing with the increase in the H_2SO_4 concentration and the electric current, as more H^+ existed in the solution and the higher electric current promoted the electrochemical leaching reaction, while the extra-high concentration of H^+ and electric current promoted the hydrogen evolution reaction, thus decreasing the electrochemical leaching performance. Regarding the leaching time, the leaching efficiencies of the metals reached equilibrium after the reaction for about 150 mins. Therefore, the optimal conditions for electrochemical leaching were set as 1.5 M H_2SO_4 solution, 0.8 A of electric current, and 150 min of leaching time. Under this condition, the leaching efficiencies of Li, Ni, Mn and Co reach 100%, 90.59%, 66.40%, and 90.53%, respectively [26]. Besides the electrochemical co-leaching of metals, including Li, Mn, Ni, and Co together, Yang et al. recently proposed an electrochemical leaching approach for selectively extracting Li from spent cathode

materials. In this study, spent $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ (NMC622) cathode materials were used as research subjects and a K_2SO_4 solution was used as the electrolyte solution. The impact of the applied voltage (1.0–3.0 V) on the delithiation performance was detailed. The SEM images of the delithiated samples are displayed in Figure 1a–d, and the leaching efficiency of the Li and a comparison of the Li with Co, Ni, and Mn under different operation voltages are presented in Figure 1e. The voltage of 2.5 V gave the best Li leaching performance as the low voltage led to low efficiency, while the high voltage resulted in a strong side reaction, which led to the leaching of the transition metals (Ni, Co, and Mn) instead of the selective leaching of Li. The working mechanism is displayed in Figure 1f. Under optimized voltage, the NiO_x releases electrons synchronously, which further facilitates the Li leaching process [32].

Table 1. A summary of electrochemical leaching studies for cathode recycling.

Leaching Items	Operation System	Leaching Agents	Applied Voltage/Current	pH	Leaching Efficiency	Reference
LIB metal oxide filter cake	Threeelectrode system	H_2SO_4 solution (0.5–2 M), FeSO_4 (10 mM) as a reducing agent	−0.3 V	>1	Li, Co, Mn, and Ni: >96%, Fe: >40%, Cu: >70%, Zn: >80%, Al: >30%	[31]
Reduced NMC cathode	Twoelectrode system	H_2SO_4 solution (0.5, 1, 1.5, and 2 M)	0.2–1.0 A	\	Li: 100%, Ni: 90.59%, Co: 90.53%, and Mn: 66.40%	[26]
NMC622 cathode	Two-electrode system	K_2SO_4 solution	1.0–3.0 V	9–12	Li: 95.02%	[32]
NMC cathode	Twoelectrode system	Malic acid solution	2.0–10.0 V	<6.6	Li: 100%, Ni: 99.87%, Co: 99.58%, Mn: 99.82%	[28]
LiCoO_2 (LCO) cathode	Twoelectrode system	Malic acid solution	2.0–10.0 V	<6.22	Li: 94.17%, Co: 90.45%,	[24]

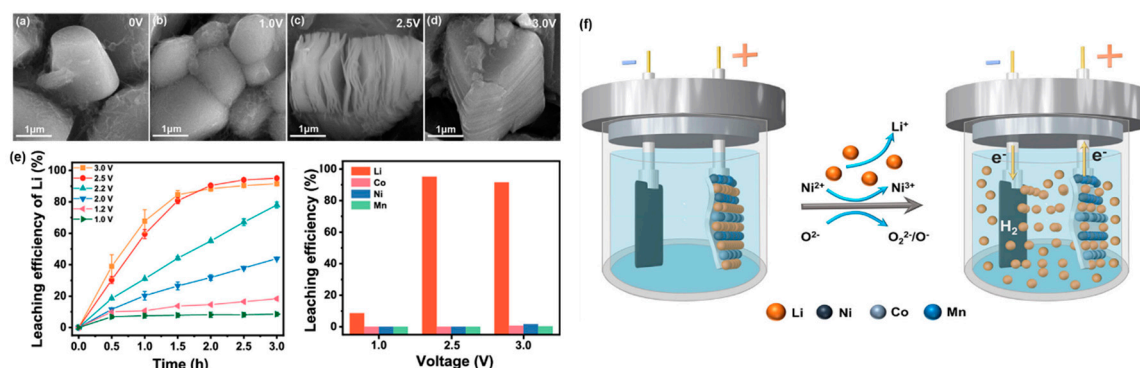


Figure 1. (a–d) SEM images of spent NMC622 cathodes after lithium released under different voltages, (e) leaching efficiencies under various voltages, and (f) schematic diagram of the electrochemical leaching for delithiation of spent cathode electrode [32].

3. Electrodeposition

Electrodeposition is an electrochemical process wherein the metal ions within the solution are reduced with an external electric current and subsequently deposited onto the electrode surface. This technique is used for battery recycling for precipitation of the critical transition metals from the solution. The order and yield of electro-deposited metals are significantly influenced by factors such as the characteristics of the metal ions, the composition and concentration of the electrolyte solution, the pH levels, the operating temperature, and the applied electric current. Consequently, the electrodeposition process can be precisely controlled by adjusting these parameters.

3.1. Lithium Electrodeposition

As an essential element for batteries, lithium recovery has received significant attention [33]. Generally, lithium recovery via electrodeposition from various resources, including ores, brine, and seawater, has been well explored [34,35]. With the emerging market for electric vehicles, a substantial number of end-of-life batteries will accumulate in the future. Consequently, lithium recovery from spent batteries is particularly appealing to battery manufacturers. Commercially, the hydrometallurgical method is applied for spent battery recycling and results in the leachate. Jang et al. conducted electrodeposition for selective lithium recovery from an acidic LIB leachate. In this work, an electrode system was employed with MnO_2 as the working electrode and activated carbon (AC) as the counter electrode. The LIB leachate was applied as an electrolyte with various cations dissolved, including Li, Ni, Co, and Mn ions. The researchers conducted a detailed investigation about the effect of current density and reaction time on the selective recovery of Li. With the increased electric current, the recovery capacity of the lithium improved, while the recovery capacities of Ni, Mn, and Co decreased. Therefore, the selective recovery of lithium from coexisting ions was achieved at a higher current density. Additionally, the delithiated MnO_2 demonstrated high selectivity for Li over other coexisting cations, probably because of the intercalation of the lithium into the MnO_2 lattice during the electrodeposition process. Regarding the reaction time, the longer the reaction proceeded, the more lithium was recovered, while other ions were not. Thus, extending the reaction time enhanced both the purity and the recovery capacity of lithium. Restricted by the electrochemical stability of the leachate, the reaction time was optimized at 15 min and the maximum lithium recovery capacity of 3.51 mmol g^{-1} was delivered under a current density of 0.2 A g^{-1} [36]. Furthermore, Jang et al. also used the MnO_2 //AC system for lithium recovery from pre-treated ammonium LIB leachate. The lithium recovery capacity was 1.39 mmol g^{-1} , with a purity of 96.8%, under a current density of 0.2 mA cm^{-2} [37].

Besides lithium recovery, lithium electrodeposition has been proposed as an effective method for cathode relithiation. Spent cathode materials often suffer from damage, particularly Li vacancy defects, after the cycling process. The traditional hydrometallurgy process involves cathode breakdown and resynthesis processes, which add complexity to the overall recycling process. Relithiation via lithium electrodeposition provides an option for closed-loop recycling by simplified process. Recently, researchers have applied electrochemical relithiation to various types of spent cathode chemistries for direct regeneration [38–42]. Liu et al. conducted electrochemical relithiation of a spent LiFePO_4 (LFP) cathode. The Li-containing solution was adopted as an electrolyte. A three-electrode system was used with platinum as the counter electrode, spent LFP as the working electrode, and Ag/AgCl as the reference electrode. As illustrated in Figure 2a, the Li ions would be intercalated into a spent LFP cathode under applied voltage, and the effect of the current densities on the relithiation performance is detailed in this work. As shown in Figure 2b, the potential vs. time curves of the electrodeposition process for the Li ions were collected under various current densities, ranging from 0.25 to 0.45 mA cm^{-2} . As the relithiation proceeds, the potential of the relithiated electrode rapidly drops and gradually stabilizes at a plateau, indicating the completion of the relithiation process. With increased current densities, less time was required to reach the plateau, showing that the Li ions were easier to insert into the spent LFP electrode under high current densities. The crystal structures of the relithiated LFP cathodes under different current densities were characterized by XRD (Figure 2c,d). The phase transformation of FePO_4 into LFP confirmed that the Li ions were successfully lithiated into the spent LFP cathodes. However, the high current density led to excess lithium and introduced Li_3PO_4 impurity phases. The Li/Fe ratio was further examined through ICP measurement, revealing a rise in the Li/Fe ratio with increasing current densities (as illustrated in Figure 2e). This observation confirmed the feasibility of electrochemical relithiation for spent LFP cathodes. The electrochemical performance of the obtained relithiated LFP cathodes was assessed by a half-cell configuration. These cathodes delivered a discharge capacity of 154.2 mAh g^{-1} at C/2 and maintained 91% of the capacity

after 300 cycles at 1 C [42]. Zhang et al. performed electrochemical relithiation on a spent LCO cathode using a three-electrode system. In this setup, a platinum plate served as the anode, the spent LCO electrode as the cathode, and Ag/AgCl as the reference electrode. A Li_2SO_4 solution (0.1–1 M) was used as electrolyte and various current densities (-0.12 to -0.42 mA/cm^2) were applied and compared for relithiation performance. The results indicated that the higher concentration of electrolytes and current density lead to a more efficient relithiation process. The relithiated LCO cathode exhibited a capacity of 136 mAh/g , which is slightly lower than the commercial LCO cathode capacity of 140 mAh/g [39].

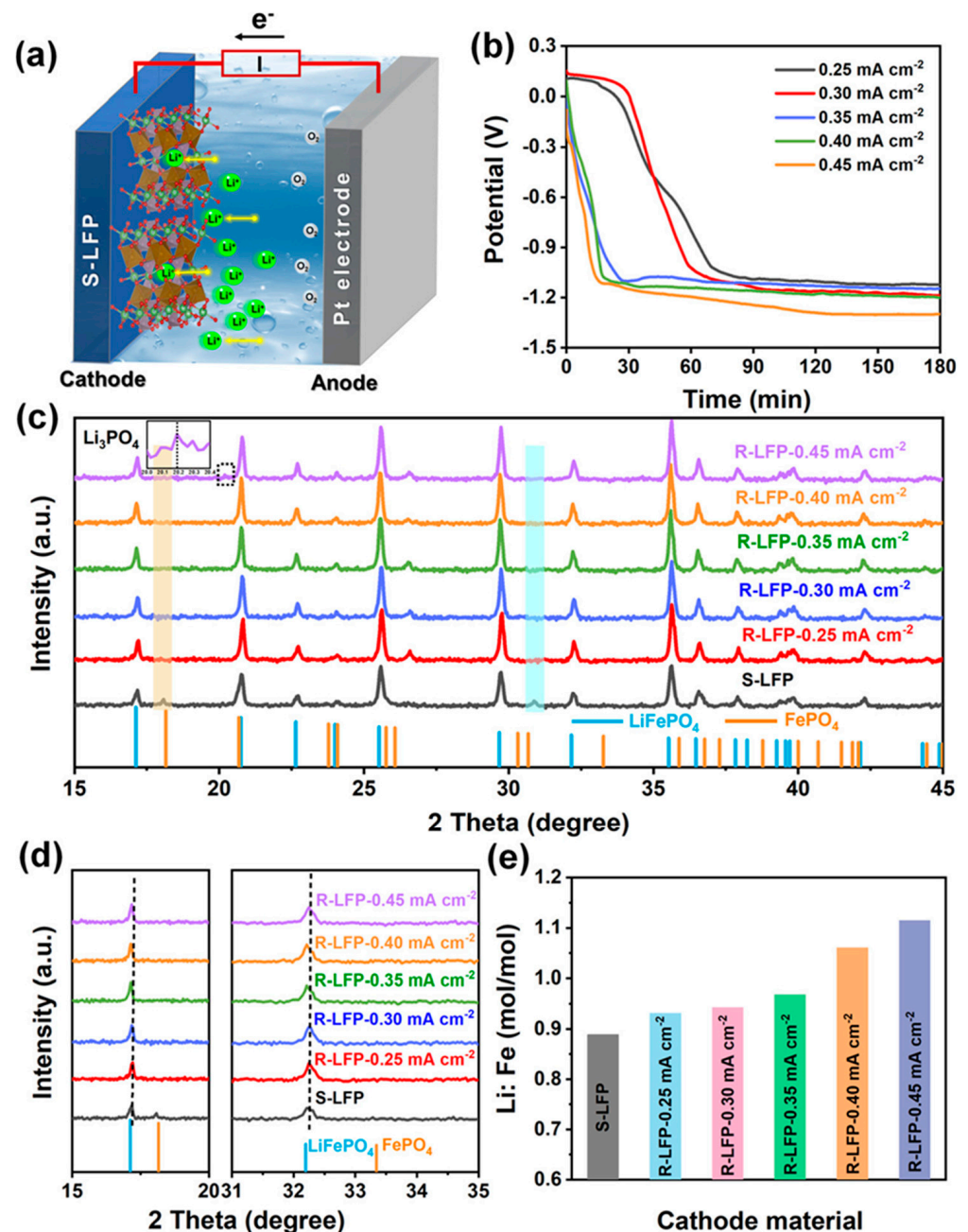


Figure 2. (a) Schematic diagram of the electrochemical system for LFP relithiation; (b) voltage–time curves of relithiation process at various current densities; (c,d) XRD spectra of relithiated LFPs at various current densities; (e) Li/Fe ratios of electrode samples [42].

3.2. Cobalt Recovery

Cobalt is a strategic material that typically contributes to the cathode and enables lithium-ion batteries to achieve high energy densities and stability over cycling. However, it is relatively expensive and has limited natural resources [43]. Several research studies have delved into the utilization of the electrodeposition method as a means of cobalt recovery [44,45]. Tran et al. applied electrodeposition for Co recovery from an obtained metal leachate. The deep eutectic solvent (DES), which is a combination of ethylene glycol and choline chloride, was applied for LCO cathode leaching. With the obtained leachate, the electrodeposition of Co was conducted with a three-electrode setup. A stainless-steel mesh was used for both the working and the counter electrodes, and Ag/AgCl served as the reference electrode. The cobalt was deposited and recovered as $\text{Co}(\text{OH})_2$ under a constant current of -4.5 mA for 1 h [43]. Recently, Cheng et al. also applied electrodeposition for co-precipitation from metal leachate. The DES, composed of choline chloride, tartaric acid, and ethylene glycol, was first used for $\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$ (NMC811) cathode leaching. The separation of cobalt and nickel was achieved as the nickel preferred to complex with the ethylene glycol and form precipitates while the cobalt ions were still dissolved in the DES. The electrodeposition of cobalt was conducted with a three-electrode system, with carbon cloth as the counter electrode, a stainless-steel mesh as the working electrode, and Ag/AgCl as a reference electrode. H_2O was added to decrease the viscosity of the DES, thus decreasing the resistance for charge transfer during the electrodeposition process. The cobalt was recovered as $\text{Co}(\text{OH})_2$ with a purity of 97.3% and a recovery yield of 98.1%, with an optimized potential of -1.3 V and a reaction time of 4 h [46].

3.3. Electrodeposition of Ni-Co Alloy

In many scenarios, Ni^{2+} and Co^{2+} always coexist in the leachate of lithium-ion batteries, while there still a lack of an effective method to separate nickel and cobalt due to their similar reduction potentials [20,46,47]. In other words, the electrodeposition of Ni-Co alloy is feasible [48]. In 2020, Landa-Castro et al. used a choline-chloride-based DES for Ni-MH battery leaching. The collected leachate was used as an electrolyte for the electrodeposition of a Ni-Co alloy. A three-electrode system was employed in that work, with glassy carbon as the working electrode, platinum wire as the counter electrode, and silver wire as the reference electrode. The deposited Ni-Co alloy contained 64.09% nickel and 35.90% cobalt in the form of Ni and Co hydroxyl complexes [49]. Recently, Liu et al. conducted a one-step electrodeposition of a Ni-Co alloy from LIB leachate and detailed the effect of the potential (-0.7 to -1.5 V) and pH (2–6) on the electrodeposition of the Ni-Co alloy. The initial LIB leachate was obtained after sulfuric acid leaching, and it was directly obtained from industry. The concentrations of the Li^+ , Ni^{2+} , Co^{2+} , and Mn^{2+} in the LIB leachate were 2430, 71,560, 15,840 and 24,160 mg/L, respectively. The workflow process is shown in Figure 3a. A three-electrode system was used here, with Ag/AgCl as the reference electrode, and both the counter electrode and the working electrode used carbon cloth. As shown in Figure 3b–e, as the reduction potential increased, the concentrations of Ni^{2+} and Co^{2+} greatly decreased, indicating Ni-Co electrodeposition. The -1.3 V gave the best performance for Ni-Co electrodeposition and the amount of electrodeposited Ni-Co even decreased at a higher potential at -1.5 V , probably because of the severe hydrogen precipitation on the cathode, which affects the deposition of Ni-Co alloys. Regarding the pH values, the optimal removal rates of Ni^{2+} and Co^{2+} , resulting in the best Ni-Co electrodeposition performance, were achieved at pH = 3. At pH = 2, the positive potential (as shown in Figure 3f) due to the protonation on the cathode surface led to a repulsive effect on the Ni^{2+} and Co^{2+} , which inhibited the deposition of the Ni-Co alloy. For electrolytes with pH values of 4 to 6, the experimental results show that the pH of the leachate quickly fell below 2 within 0.5 h of the reaction (as shown in Figure 3g). This severe drop in pH led to increased protonation on the surface of the cathode, reducing electrostatic attraction, thereby inhibiting the reduction of the Ni^{2+} and Co^{2+} . Additionally, the presence of Mn^{2+} acted as an electron donor, which was found to be a crucial factor contributing to the Ni-Co

alloy's electrodeposition. By supplementing the Mn^{2+} ions in the solution, the Mn^{2+} was oxidized at the anode, which further promoted the cathodic electrodeposition. With this proposed method, the purity of the electrodeposited MnO_2 at the anode reached 99.52%, and that of the Ni-Co alloy at the cathode reached 99.80% [47].

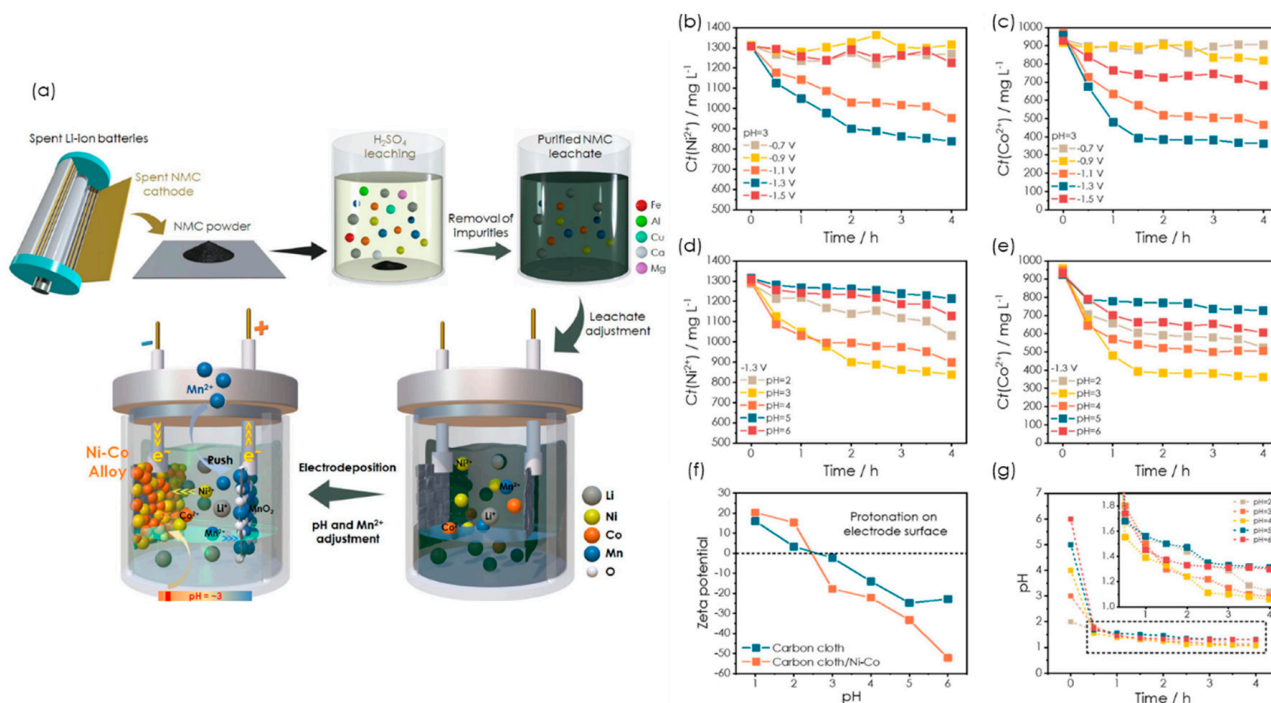


Figure 3. (a) Schematic diagram of electrodeposition of Ni-Co alloys; the concentration of Ni^{2+} (b) and Co^{2+} (c) at reduction potentials (-0.7 to -1.5 V) under $\text{pH} = 3$, and the concentration of Ni^{2+} (d) and Co^{2+} (e) at $\text{pH} 2\text{--}6$ under -1.3 V; (f) zeta potential of carbon cloth and carbon cloth/Ni-Co under pH values of $1\text{--}6$; and (g) changes in electrolyte pH as the reaction proceeded [47].

3.4. Ni and Co Separation via Electrodeposition

As previously mentioned, it is challenging to separate Ni^{2+} and Co^{2+} via electrodeposition because of their similar reduction potentials. Over the past few years, many researchers have been investigating methods to achieve Ni/Co separation. In 2021, Kim et al. proposed a strategy to achieve nickel and cobalt separation via electrodeposition through the combined function of interfacial engineering and electrolyte control. In this work, the researchers employed 10 M hydrochloric acid as a leaching agent for the NMC cathode and manipulated the pH value of the collected metal leachate solution by adding LiOH . LiCl was added to obtain highly concentrated chloride as the electrolyte, which helped the formation of negatively charged CoCl_4^{2-} and positively charged $[\text{Ni}(\text{H}_2\text{O})_4\text{Cl}]^+$. With the formed nickel and cobalt complex, the LSV curves of the cobalt and nickel showed significant differences, with reduction potentials of -0.68 V vs. Ag/AgCl and -0.59 V vs. Ag/AgCl , respectively (shown in Figure 4a–c). The researchers compared the electrodeposition with different chloride concentrations (0.1 M Li_2SO_4 , 0.1 M LiCl , and 10 M LiCl) and the results showed that the anomalous deposition of Co/Ni was highest in the 10 M LiCl at -0.75 V vs. Ag/AgCl (shown in Figure 4d–f). Subsequently, as shown in Figure 4g, with the controlled PDAMA polymer loading on the Cu substrate, the electrode potential could be fine-tuned, thereby directly separating the opposite charged nickel and cobalt complexes. The final purities of the cobalt and nickel were $96.4 \pm 3.1\%$ and $94.1 \pm 2.3\%$ [20].

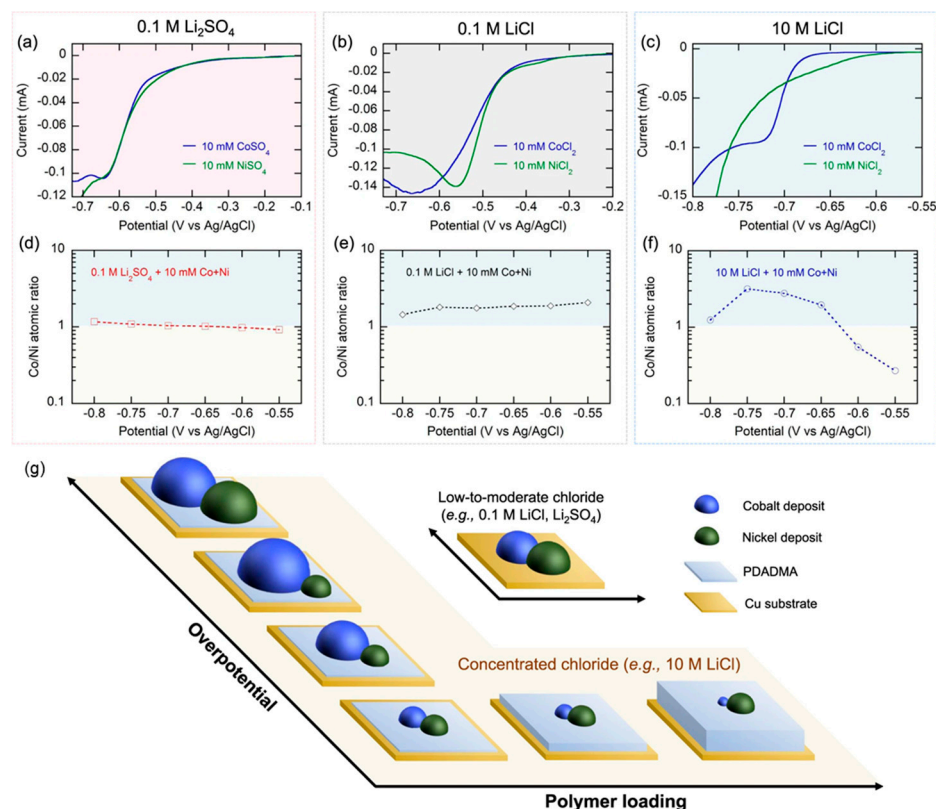


Figure 4. (a) Linear sweep voltammograms of a 10 mM Co²⁺ or Ni²⁺ in (a) 0.1 M Li₂SO₄, (b) 0.1 M LiCl, and (c) 10 M LiCl; electrodeposition of Co/Ni atomic ratios with the electrolyte of (d) 0.1 M Li₂SO₄, (e) 0.1 M LiCl and (f) 10 M LiCl; and (g) schematic diagram of selective deposition of Ni and Co by synergistic electrolyte and interfacial control [20].

4. Conclusions and Perspectives

This mini-review outlined the recent progress in electrochemical techniques for recycling lithium-ion batteries. The discussion was divided into two main directions: electrochemical leaching for dissolving cathode materials and electrodeposition for metal precipitation. Effective processes have been developed for co-leaching lithium, nickel, cobalt, and manganese from cathode materials, as well as for the selective leaching of lithium. In terms of electrodeposition, efficient methods have been established for recovering cobalt and cobalt–nickel alloy, electrodepositing lithium for cathode relithiation, and nickel and cobalt separation. Significant progress has been made in developing electrochemical methods tailored for lithium-ion battery recycling. However, these processes are still in the early stages, with many challenges remaining. Further research and exploration are necessary, as outlined below:

- (1) The reaction mechanisms behind electrochemical leaching and electrodeposition need further clarification. Advanced characterization techniques, such as neutron scattering and in situ X-ray analysis, should be used for more in-depth investigation.
- (2) There is still a lack of efficient methods for selectively leaching or depositing metals. It remains challenging to separate nickel and cobalt via electrochemical methods because of their similar chemical reactivities.
- (3) The process requires fine-tuning with precise control over purities. Maintaining purity levels remains a significant hurdle in both the electrochemical leaching process and electrodeposition.
- (4) Further advanced research is needed for investigating and controlling the morphology of the obtained metals through electrodeposition.
- (5) There is a crucial need for efficient, customized electrochemical methods tailored to various battery chemistries. A comprehensive investigation is required to elucidate

the mechanism behind relithiation through lithium electrodeposition, and to develop cathodes that exhibit the desired electrochemical performance.

- (6) Electrochemical methods for recycling lithium-ion batteries primarily target cathode materials. However, the pretreatment process involves complexities, such as battery dismantling and electrode delamination. Additional research is required to develop efficient pretreatment methods.
- (7) The current electrochemical recycling process is limited to the laboratory scale. Extensive further research is necessary to upscale the process for commercialization. Rigorous testing and validation procedures are essential to showcase the scalability, reliability, cost-effectiveness, and sustainability of the technology.

In summary, electrochemical methods show promise for recycling lithium-ion batteries. The ongoing research and development in this field offers great potential for advancing battery technology while promoting sustainability.

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