

Review

# Emerging and Recycling of Li-Ion Batteries to Aid in Energy Storage, A Review

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**Abstract:** The global population has increased over time, therefore the need for sufficient energy has risen. However, many countries depend on nonrenewable resources for daily usage. Nonrenewable resources take years to produce and sources are limited for generations to come. Apart from that, storing and energy distribution from nonrenewable energy production has caused environmental degradation over the years. Hence, many researchers have been actively participating in the development of energy storage devices for renewable resources using batteries. For this purpose, the lithium-ion battery is one of the best known storage devices due to its properties such as high power and high energy density in comparison with other conventional batteries. In addition, for the fabrication of Li-ion batteries, there are different types of cell designs including cylindrical, prismatic, and pouch cells. The development of Li-ion battery technology, the different widely used cathode and anode materials, and the benefits and drawbacks of each in relation to the most appropriate application were all thoroughly studied in this work. The electrochemical processes that underlie battery technologies were presented in detail and substantiated by current safety concerns regarding batteries. Furthermore, this review collected the most recent and current LIB recycling technologies and covered the three main LIB recycling technologies. The three recycling techniques—pyrometallurgical, hydrometallurgical, and direct recycling—have been the subject of intense research and development. The recovery of valuable metals is the primary goal of most recycling processes. The growth in the number of used LIBs creates a business opportunity to recover and recycle different battery parts as daily LIB consumption rises dramatically.

**Keywords:** Li-ion battery (LIB); recycling; leaching; electrode; electrolyte; remanufacturing



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## 1. Introduction

Climate change is intimately tied to the potential for energy production and the demand for energy. There is currently a notable drive to progress renewable energy technology in light of the enormous energy demand for the expansion of the economy and the excessive use of nonrenewable energy resources such as coal, oil, and fossil fuels [1,2]. Moreover, economic development has become more challenging because of the unsustainable nature of nonrenewable energy resources [3,4]. This would result in a significant rise in environmental pollution, which will worsen existing issues including acid rain, ozone depletion, and global warming brought on by the atmospheric release of dangerous gases such as carbon dioxide (CO<sub>2</sub>) [5–7].

Solar cells [8], fuel cells [9–13], supercapacitors [14], and metal-based batteries are a few examples of clean and sustainable energy technologies that are so pervasive [15].

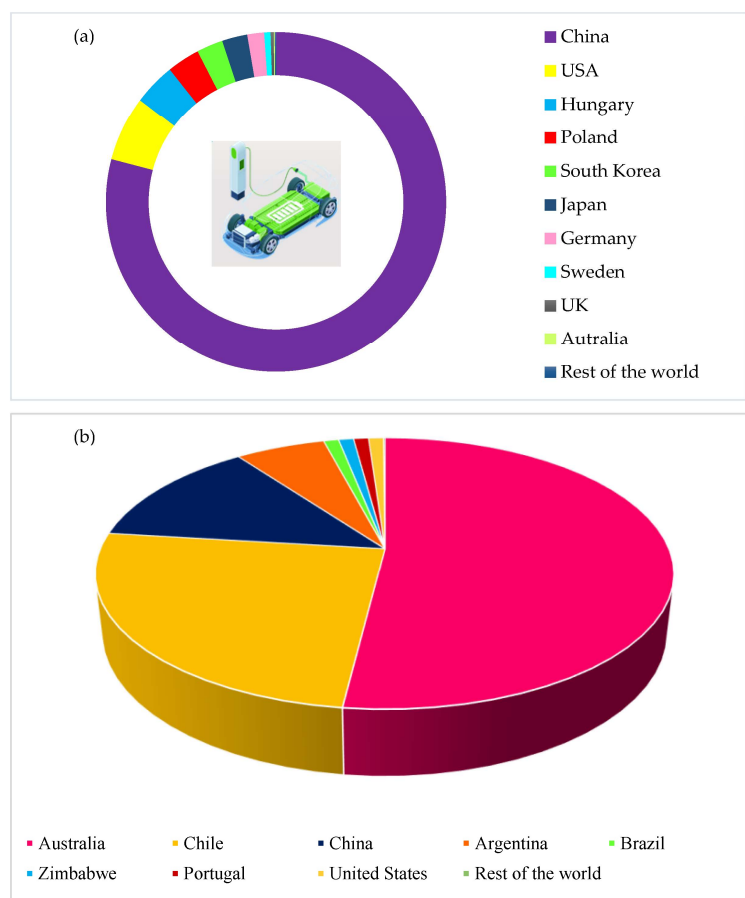
Furthermore, the unpredictable availability of renewable electricity sources necessitates the rapid development of energy storage technologies and proper energy distribution [16]. These requirements are the outcome of an effort to advance an eco-friendly economy that favors the use of clean energy, rather than continually depleting nonrenewable natural resources, in order to support a healthy environment [17]. This has led to tremendous recent advances in energy storage in terms of adaptability, high energy density, and efficiency [18]. More self-sustaining approaches to energy management are being looked into as a result of the world's rapidly expanding population, which will result in a higher rate of resource use [19].

The metal-ion battery is one of the most important inventions for assisting with energy supply and management [20]. In 1749, Benjamin Franklin was known to be the first person to create the term "battery" [21]. In the 19th century, Michael Faraday was the first to discover electromagnetic induction [22]. However, the first galvanic battery was constructed by Alessandro Volta, which he named the Voltaic pile [23]. Batteries are seen to be significantly valuable due to the discovery of the telegraph [24]. Hence, the importance of batteries skyrocketed [23]. Since the lead battery was first developed in 1859, years of study and development have gone into the development of these metal-ion batteries [25]. As a consequence, numerous technologies, including NiMH (nickel–metal hybrid), NiCd (nickel–cadmium), NMC (nickel–cobalt–manganese), LCO (lithium–cobalt oxides), NCA (nickel–cobalt–aluminum), LFP (lithium–iron–phosphate), and lithium-ion batteries, have been developed [26]. Batteries may also be broadly categorized into three groups: rechargeable batteries, flow batteries, and ultra-batteries [27,28]. Rechargeable batteries or secondary batteries are known for numerous charges and discharges of a load from one form of energy (electrical) to another (chemical) and back [29]. Moreover, by using reversible electrochemical reactions, the energy can be collected and stored; compared to using nonrenewable energy, this will reduce the environmental impacts [30]. However, an increase in production of the number of applications can lead to an increase in cost [31]. Flow batteries consist of a membrane that separates the two electrolyte reservoirs (positive and negative) [30]. The vital component of the flow batteries is the membrane because these components determine the performance and manufacturing cost [32]. Flow batteries offer several advantages by separating the electrolytes reservoir such as a flexible layout, high energy efficiencies, long cycle life, and easy scalability [33]. However, the energy density is quite low and the construction of the battery is complex [34]. At the turn of the 20th century, as a result of the steady improvement of renewable energy, the use cases for lithium-ion batteries (LIBs) have expanded to encompass electric vehicles, electric bicycles, household appliances, energy storage, and a variety of other things [35].

The development of lithium-ion batteries has inspired researchers globally for many years. Consequently, the 2019 Nobel Prize in Chemistry was given to J. B. Goodenough, M. S. Whittingham, and A. Yoshino, who are the creators of wireless devices including mobile phones and laptops [36]. LIBs have been explored as one of the most developed power supply technologies with high charge–discharge current [37]. The greatest lithium deposits in the world are in Australia, where there are more than 55,416 tons of lithium reserves. The list of deposits covers not just Australia, but also other nations such as Chile, China, Argentina, etc., that are producing a significant amount of lithium, leading the global lithium output to top 100,000 tons for the first time in 2021 [38,39]. Figure 1b displays the countries in the world with the highest lithium output. A substantial portion of the worldwide market is claimed by a few nations that are major producers of battery technology. Large battery manufacturers are located in these countries, which frequently have well-established supply chains and infrastructure to enable the large-scale production of batteries. China produced 13.98 billion Li-ion batteries in 2018, an increase of 12.9% over the previous year; of these, 56.9 GWh of installed power batteries had a growth rate of 56.3% annually [40], and according to the China Association of Automobile Manufacturers (CAAM), 19.1% of Chinese vehicles will be electric by December 2021 [41]. Rapidly increasing demand for lithium-ion batteries has also resulted in increased interest in using

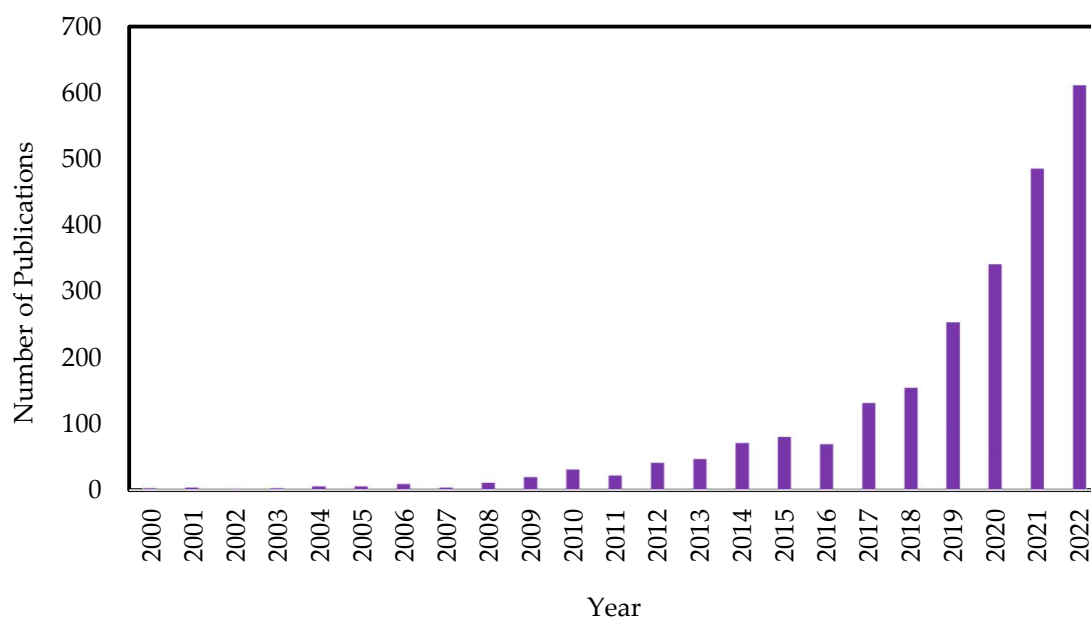
batteries for electric cars (EVs). The power and energy density performances of LIBs have been significantly improved for larger-scale implementations with electric and hybrid electric vehicles [42,43]. EVs have just started to gain popularity in international markets, including China, the USA, and Europe, and are expected to produce 9300 gigawatt-hours (GWh) by 2030 (Figure 1a).

At present, gasoline-fueled cars have been replaced with lithium-ion batteries; this substitution reduces carbon gas emissions to the atmosphere hence, improving the ecological impact [44]. Moreover, the high electrical performance of LIBs may enable their usage in a variety of applications for the electric grid, such as improving the quality of energy obtained from renewable sources such as wind, solar, geothermal, and others [45]. Therefore, LIBs have become the prime focus of interest for both industry and government funding agencies [45].



**Figure 1.** (a) The largest battery producers are situated in areas where there is a considerable demand for EVs and (b) world countries with the highest lithium production [39,46].

In order to encourage the growth of LIBs in various nations, a significant amount of resources is committed to R&D [47]. LIBs are known to be lightweight, more compact, design flexible, able to operate at a voltage of 3.6 to 4 V, and have a capacity ranging from 700 to 2400 mAh for a single cell (battery) [48,49]. To avoid wasting energy and to keep the modules at the desired state of charge (SOC) for storage, a partial discharge test that measures the partial capacity during the discharge process was developed to determine the remaining capacity of retired battery modules from electric vehicles (EVs). A nominal voltage of 3.2 V and a capacity superior to 20 Ah was found for this LIB [50]. LIBs are far more advanced than other traditional batteries in terms of their volumetric and gravimetric energy and power densities [51]. According to the amount of research papers by publication numbers by year, there has been a noticeable growth in the study of different LIB sectors in the pursuit of high-performance rechargeable LIBs, as shown in Figure 2.



**Figure 2.** A plot of publication numbers by year based on lithium-ion batteries (source of data: [www.scopus.com](http://www.scopus.com)) [52].

Although lithium metal batteries theoretically have far higher energy densities than LIBs, overuse and vulnerability will cause an explosion because of their low recharge ability [53]. For the rechargeable battery market, LIBs are known to be dominant compared with different rechargeable batteries [53]. Moreover, regarding availability, lithium is located at the outside layer of the Earth and has an adequate amount of power to operate automobiles globally [54]. Thus, rechargeable batteries have been an immediate economical product and are considered necessary globally [55]. However, lithium-ion batteries (LIBs) are majorly required in numerous electrical devices such as mobile phones and automobiles [56]. Thus, there is an increase in the prices of lithium [57,58]. The market for lithium-ion batteries is anticipated to expand greatly as well, given that the demand for these batteries will climb by an astounding elevenfold globally. China was the world leader in lithium-ion battery production in 2020, generating more than 70% of the batteries that were sold worldwide [59]. It should come as no surprise that China was home to the majority of the largest battery facilities in the world. By 2030, both Europe and the USA aim to make substantial investments in lithium-ion battery production facilities to fulfill domestic battery demand with entirely local supply chains [60]. The export volume will peak at 439.32 Gigawatt hours (GWh) by 2025, and the global demand for LIBs for energy-storage networks and EVs will reach USD 99.98 billion [61].

The development of materials for use in LIB devices has also been the subject of extensive research during the past several years [62]. The nanoscale ranges in engineering electrode materials are known to be important in the field of electrochemical energy storage [63]. Presently, a variety of nanostructures, including zero-, one-, two-, and three-dimensional ones, have been developed using complementary electrode materials [64,65]. Generally, the thermodynamics and kinetics of electrochemical reactions can be modified based on the size of the nanostructured materials [66]. Additionally, there are ways to improve nanostructures, such as increasing the capacity (Q) of the active material and boosting the working voltage (V) by reducing polarization [67]. Moreover, nanostructures can increase the power density by reducing the electronic transport and ionic diffusion distance [68]. Despite this, significant side reactions will occur when using a large surface area of electrode or electrolyte. Meanwhile, nanomaterial that has a small packing density can lead to lower volumetric density [69]. Thus, the improvement of nanostructured material can widen the scope and challenges based on improving energy storage. Cleaner alternative battery technologies are therefore more appealing, especially those that employ

components that are more available, could have a greater energy density, and might have better safety measures.

Most LIBs now in use have a shelf life of one to three years, after which they are either disposed of in landfills or transferred to the municipal waste stream. The worldwide electric vehicle sales have reached 140 tonnes by 2040, with over 11 million tonnes of expended energy by 2030. Less than 5% of LIBs are being recycled presently [70]. The number of LIBs that are decommissioned or abandoned will increase even more as a result of battery issues and the fact that many battery storage systems are nearing the end of their useful lives. The majority of lithium batteries employ lithium (Li), nickel (Ni), cobalt (Co), manganese (Mn), and other valuable metals. They have the same advantages as natural ore if these metals are successfully extracted on a big scale from extinct batteries. The recovery of valuable metals from used LIBs and their recycling to create new rechargeable LIBs and other rechargeable battery technologies is a lucrative business venture for many business persons. Recycling would thereby enhance supply chain integrity, lessen reliance on these materials, and lessen the negative effects these batteries have on people and the environment [71,72]. Recycling would therefore result in less reliance on new materials while having a minimal impact on the environment [73]. In addition to recycling the batteries, three main methods—pyrometallurgy, hydrometallurgy, and direct regeneration methods—are typically used to recycle the cathodic materials in LIBs.

The present work will mainly focus on the development of next-generation Li-ion batteries, the underlying ideas, the highlighting of recent advancements in materials, and an analysis of the challenges that Li-ion batteries encounter. Thereupon, we move to the most recent advances in battery technology. To guarantee that the huge quantities of LIBs produced are not damaging to the environment or human health after consumption after being produced at an increased pace to fulfill the energy needs of daily living, recycling these precious materials used in LIBs is essential. To address such a challenge, we present a brief overview of these cutting-edge recycling solutions for LIB materials based on the most recent methodologies established for recycling LIB battery materials. We anticipate that this analysis will not only give a thorough overview of recent developments in the battery business, from material selection to technological innovation, but will also throw light on the potential future development of unique battery recycling technologies. This will illuminate how to boost the economic and environmental sustainability of the batteries.

## 2. Lithium-Ion Battery (LIB)

Due to their effectiveness in storing energy, ease of upkeep, and extended lifespan, lithium-ion batteries (LIBs) are the most popular and successfully developed type of metal-ion batteries now on the market (Figure 3) [74]. In the early 1990s, a major development of technology based on lithium battery has improved significantly, where negative electrodes were utilized on different ranges of high specific energy lithium batteries [75]. Since their introduction to the general public in 1991, LIBs have found widespread application in electric cars and a variety of portable devices [30]. Due to their light weight and high energy density, LIBs have also come to be recognized as one of the most important energy storage technologies [48].

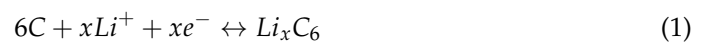
To generate an LIB, Li-ion cells are connected in a number of different ways, including in parallel, series, or combination. Using a parallel setup is one technique to enhance the current. Meanwhile, series wiring is used to raise voltage [76,77]. An anode, a cathode, and a solid-state electrolyte that holds lithium ions are the three primary parts of the LIB (Table 1). These lithium ions are then observed [30]. Lithium ions can travel back and forth between the positive and negative electrodes thanks to a microporous polymer membrane that acts as a separator between them [53]. The lithium-ion battery has a larger capacity while being smaller than a typical nickel–metal hydride battery [78]. Furthermore, because it can be charged with high electrical conversion efficiency, it provides efficient energy consumption with low energy loss [45]. For this reason, lithium-ion batteries are considered to be an appropriate basis for discussing the recycling technology processes [79].



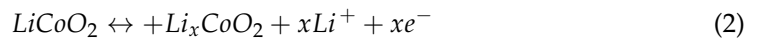
**Figure 3.** Typical commercial lithium-ion batteries.

Figure 4a shows the basic schematic and working fundamental of the lithium-ion battery cell. Commercial LIBs are constructed by using layer-type compounds; the positive electrode, which is the cathode, uses lithium cobalt oxide (LiCoO<sub>2</sub>), LiNiO<sub>2</sub> (LNO), LiMn<sub>2</sub>O<sub>4</sub> (LMO), LiFePO<sub>4</sub> (LFP), LiNi<sub>x</sub>Co<sub>1-x</sub>AlO<sub>2</sub> (NCA), and LiNi<sub>x</sub>Mn<sub>y</sub>Co<sub>z</sub>O<sub>2</sub> (NMC) [36]; the negative electrode, which is the anode, use graphite (C); and meanwhile, the electrolyte is a nonaqueous Li-ion [48]. For the charging process, the electrical supply is connected to the two electrodes as shown in Figure 4a. The electrons will be driven to leave the cathode and go to the anode by this. When moving from cathode to anode through the electrolyte, the lithium ions do so concurrently in the same direction. The anode and cathode parts of the battery, therefore, contain electrochemical energy [53]. The reactions which occur through the lithium-ion and electron generation are as follows:

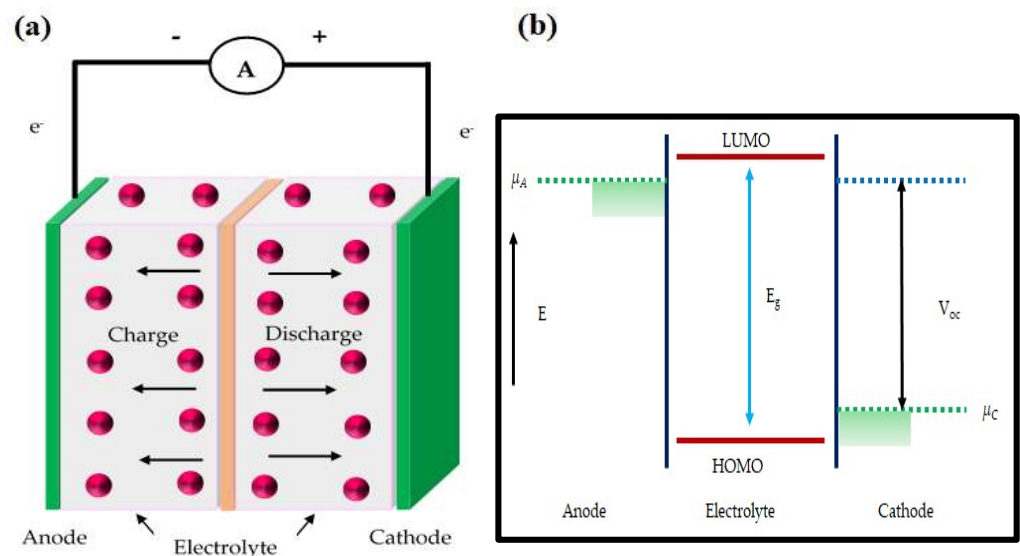
At the anode



At the cathode



To transmit energy during the discharge process, the electron from the anode travels to the cathode through a wire that passes through an external load [80]. Simultaneously, Li ions from the anode travel through the electrolyte to the cathode. Shuttle chair refers to the movement of Li ions between the anode and cathode during charge and discharge cycles [81].



**Figure 4.** (a) Schematic diagram of a lithium-ion battery and (b) electrode potentials and the electrolyte energy gaps relative energies in LIBs [82,83].

A battery pack originates from a single battery cell. The process involves numerous battery cells merging to form a module which results in a battery pack [53]. Furthermore, optimal efforts can be acquired based on the electrode materials, especially for the anode and cathode [84]. This depends upon their electrochemical potential values, in which  $\mu_A$  is known as the anode and  $\mu_C$  is known as the cathode [52]. To achieve a stable cell,  $\mu_A$  is required to have a lower energy compared to the LUMO of the electrolyte. Thus, the electrolyte will be reduced, as illustrated in Figure 4b, it would have a greater energy than the HOMO of the electrolyte [83]. Moreover, high lithium intercalation power and the greatest electrochemical potential difference between the anode and the cathode may be used to achieve high energy storage density [85]. A variety of factors, including specific energy, volumetric energy, particular capacity, abuse tolerance, and discharging rate, affect how effectively LIBs function [53].

**Table 1.** Typical lithium-ion battery cell components [86].

Cell Component	Material	Composition/wt%	Cost/%
Cathode active material	Layered structures, e.g., LiCoO <sub>2</sub> (LCO)/Li(Ni <sub>x</sub> Mn <sub>y</sub> Co <sub>1-x-y</sub> )O <sub>2</sub> (NMC)/Li(Ni <sub>1-x-y</sub> CoAl <sub>y</sub> )O <sub>2</sub> (NCA), Spinel structures, e.g., LiMn <sub>2</sub> O <sub>4</sub> (LMO), Olivine structures, e.g., LiFePO <sub>4</sub> (LFP)	22–25	65–70
Cathode foil	Al	4–5	1
Anode active material	Materials made of silicon or carbonaceous substances (such as graphite or hard carbon)	24–26	8–9
Anode foil	Cu	3	2
Binder	Polyvinylidene fluoride (PVDF)/polytetrafluoroethylene (PTFE)/polyvinyl alcohol (PVA)/carboxymethyl cellulose (CMC)/styrene butadiene rubber (SBR)	2–3	8–9
Electrolyte	Mixtures of ethylene carbonate (EC)/propylene carbonate (PC)/dimethyl carbonate (DMC)/ethyl methyl carbonate (EMC)/diethyl carbonate (DEC) and additives, e.g., fluoroethylene carbonate (FEC)/vinylene carbonate (VC)	10–12	1
Conductive additive	Acetylene black (AB)	1	0.1
Conductive salt	LiPF <sub>6</sub>	1.5–2	8
Separator	Polyethylene (PE)/polypropylene (PP)	4–5	4
Cell case	Varies (metal or laminate)	4–6	4

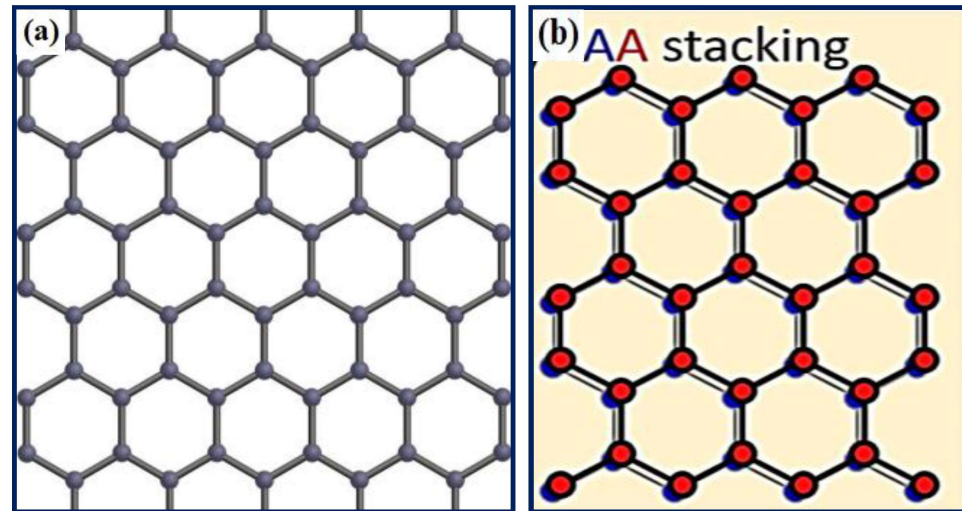
## 2.1. Anode

In electrochemistry, the term “anode” refers to the location of an oxidation process [87]. Typically, anions or negative ions that are located at the anode are forced due to electrical potential which will react chemically and distribute electrons [67]. Thus, the electrons will flow upward and move toward the driving circuit. Nowadays, there is a wide range of research that has been conducted based on anode material. The electrochemical efficiency of Li-ion batteries in terms of cycle life, rate of charging, and energy density has a considerable bearing on the choice of anode materials [88].

### 2.1.1. Carbon-Based Anode

The carbon anode or graphitic carbon has been known as the dominant material in commercial Li-ion batteries for more than 20 years [45]. Unfortunately, the major problem with using a metallic anode is that the surface of the metal can form a dendrite which can lead to short-circuiting, and produce a thermal runaway reaction on the cathode [45]. Using a minimal potential intercalation electrode will prevent the lithium anodes from dendrite formation associated with cycling and safety issues [89]. Nowadays, there are two types of anode material that are used, which are carbon (primarily graphite) and lithium alloyed metals [90]. Graphite is known as the most effective anode material due to

it being a highly available material and having high reactivity with alkali metal anodes [91]. Moreover, it was discovered that the graphite intercalation alloy has the capability to shield the lithium, thus formulating the anode to be less reactive towards electrolyte [88]. A variety of graphene sheet types are organized in graphite either in hexagonal (AB) or rhombohedral (ABC) arrangements [89]. Figure 5 illustrates the basic graphene sheet.



**Figure 5.** (a) Schematic structure of graphene sheet and (b) atomic configuration simulations of the AA arrangement in graphene sheets [92].

As the lithium ion enters the anode, the formation of graphene sheets will automatically recombine into the AA position; they form layers on top of each other, and this process is known as staging [93,94]. Li-ion batteries utilize carbon as the material for anode due to several beneficial in terms of low cost, abundant availability, and combined properties such as low delithiation potential compared to Li. It has a high diffusivity, is a great conductor of electricity, and during lithiation or delithiation the volume change is quite low, as shown in Table 2.

**Table 2.** Properties of different anode materials.

Material	Lithiation Potential (V)	Delithiation Potential (V)	$D_{Li}$ ( $cm^2 s^{-1}$ )	Ref.
Graphite	0.07, 0.10, 0.19 0.08, 0.11, 0.20	0.1, 0.14, 0.23 0.11, 0.15, 0.22	$10^{-11}$ – $10^{-7}$	[95–100]
LTO	1.55	1.58	$10^{-12}$ – $10^{-11}$	[101–104]
Si	0.05, 0.21 0.07, 0.22	0.31, 0.47 0.3, 0.49	$10^{-13}$ – $10^{-11}$	[105–108]
Ge	0.2, 0.3, 0.5	0.5, 0.62	$10^{-12}$ – $10^{-10}$	[109–111]
Sn	0.4, 0.57, 0.69	0.58, 0.7, 0.78	$10^{-16}$ – $10^{-13}$	[112–114]
Al	0.19	0.45	-	[115]
Ga	0.025, 0.52, 0.82	0.25, 0.73, 0.90	-	[116]
Zn	0.06, 0.19, 0.21	0.16, 0.22, 0.25	-	[117]
P	0.61, 0.71, 0.8	0.99, 1.16	-	[118]
Sb	0.78, 0.83	0.99	-	[119]
Ag	0.04, 0.12	0.04, 0.26, 0.38	-	[120]
Mg	0.05	0.24	-	[121]
Bi	0.73, 0.78	0.85	-	[122]



Regardless of the beneficial factors mentioned above, graphite carbon has poor lithium intercalation capacity compared to Li-ion alloys [123]. Hence, the anode material is replaced by utilizing graphitic carbon [90]. Graphitic carbon has two features, which are large graphite grains and a layered structure. Thus, this will produce minimum irreversibility alongside the movement of lithium ions within the lattice space, resulting in excellent cyclability [124]. Moreover, carbonaceous anodes illustrate significant improvements in terms of charge–discharge efficiency and discharge capacity by modifying the structural and surface [90].

It is worth noting that there are more materials that are typically used to produce the anode, including metallic lithium [125], hard carbon [126], synthetic graphite [127], lithium titanate [128], alloys [129], and silicon-based materials [130].

### 2.1.2. Hard Carbon

It has been noted and noticed in the electric industry that graphite electrodes have good capacity and high energy density, but coke or hard carbon, as an anode, has outstanding cycle stability. The first lithium-ion secondary battery was constructed in 1991 using hard carbons, which the Kureha Corporation (Japan) produced. These materials have a large reversible capacity (more than 500 mAh/g<sup>1</sup>) in the potential range of 0–1.5 V compared to Li/Li<sup>+</sup> [131,132]. As a result of the random alignment of the graphene sheets in hard carbon, there are numerous spaces that may hold lithium; however, because of how lithium diffusion happens inside hard carbons, this material has a very low rate capacity. Undesirably, poor initial coulombic efficiency and poor tap density, however, are the two key shortcomings of hard carbons [133].

### 2.1.3. Alloy Anode

Whilst compared to Li-ion alloys, graphite carbon has a lower capacity for intercalating lithium, alloy anodes, such as those made of aluminum (Al), tin (Sn), magnesium (Mg), silver (Ag), and antimony (Sb), can be great LIB anode choices. It is well known that alloy anodes have a specific capacity that is two to ten times greater than anodes composed of carbon [134]. The volume needs to be accommodated within a battery since all alloys expand significantly during lithiation. Consequently, in order for volumetric capacity values to be connected to performance in actual cells, they must be computed with regard to alloy volume at complete lithiation [135,136]. Alloy anodes are a fascinating issue with promise for LIB progress because of these features. For tin oxide, the specific theoretical capacity varies from 783 mAhg<sup>−1</sup> to 4211 mAhg<sup>−1</sup> for silicon [137,138]. While these alloy-based materials can offer greater specific capacities than graphite (372 mAhg<sup>−1</sup>) and LTO (175 mAhg<sup>−1</sup>), their main limitations are low cycling life caused by fast volume expansion/contraction, and a higher irreversible capacity during the early cycles [139,140].

### 2.1.4. Metallic Anode

Currently, the LIB research effort proposes the use of several metal oxides as viable anode choices. Transition metal oxides (MO<sub>x</sub>, where M is the transition metals such as iron, cobalt, nickel, copper, and zinc) have several appealing properties that have sparked much interest in them as top possibilities for LIB anode materials due to their high performance with a non-toxic nature.

Tin sulfide (SnS), molybdenum disulfide (MoS<sub>2</sub>), iron disulfide (FeS<sub>2</sub>), and cobalt sulfide (CoS<sub>2</sub>) are all chalcogenides that have emerged as appealing anode materials with a high theoretical capacity [141]. Despite how chalcogenides have a high theoretical capacity, highly conductive and stable materials such as polyaniline and graphene are applied as coatings on the surface of the sulfides to improve the cycling stability and rate performance of the chalcogenides (especially cobalt sulfide), which are hampered by significant changes in the bulk dimensions of the material during charging–discharging [142].

Transition metal oxalates are another intriguing metal anode because of the extraordinary progress made by researchers in their electrochemical characteristics [143,144]. During

Li-ion deployment/extraction, zinc oxalates ( $\text{ZnC}_2\text{O}_4$ ), cobalt oxalates ( $\text{CoC}_2\text{O}_4$ ), manganese oxalates ( $\text{MnC}_2\text{O}_4$ ), nickel oxalates ( $\text{NiC}_2\text{O}_4$ ), and iron oxalates ( $\text{FeC}_2\text{O}_4$ ) experience large volume expansions, resulting in poor cycling performance.

In addition, MXenes such as titanium carbide ( $\text{Ti}_3\text{C}_2\text{T}_x$ ) and selenides such as  $\text{CoSe}$ ,  $\text{FeSe}_2$ , and  $\text{NiSe}_2$  may be used.  $\text{Ti}_3\text{C}_2\text{T}_x$  might be used as an anode material for LIBs due to its increased electrical conductivity, excellent chemical stability, and low lithium-ion diffusion impedance [145]. Ding et al. reported that by combining  $\text{Ti}_3\text{C}_2\text{T}_x$  MXene with a transition metal selenide ( $\text{Co}_{0.85}\text{Se}$  clad with carbon) to generate hybrids, anode materials with outstanding volumetric stability, high electrical conductivity, and reversible capacity up to  $700 \text{ mAhg}^{-1}$  could be developed [146].

### 2.1.5. Silicon-Based Anode

Among the potential anode materials, silicon has the largest volumetric and gravimetric capacities [147–149]. The crust of the planet contains plenty of it. Due to its low working potential, it also does not pose as many safety issues as graphite electrodes. There is considerable academic and commercial interest in silicon development for active anode materials, which are regarded as the most fascinating and promising materials for the implementation of the next generation of LIBs. Although intermetallic Li–Si binary compounds such as  $\text{Li}_{12}\text{Si}_7$ ,  $\text{Li}_7\text{Si}_3$ ,  $\text{Li}_{13}\text{Si}_4$ , and  $\text{Li}_{22}\text{Si}_{15}$  have high specific capacity values, they still have some drawbacks that prevent the use of Si as an anode in LIBs, such as the large volume modification (400%) during the charge–discharge process that results in poor cycling life and irreversible capacity [150]. During its initial cycle of charging and discharging, silicon normally undergoes a transition from the crystalline to the amorphous phases. In order to prevent silicon from transitioning back to crystalline Si, the voltage at which lithium intercalation takes place in amorphous silicon must be kept above 50 mV [151].

## 2.2. Cathode

Currently, there are numerous types of research on cathode material that have been investigated for Li-ion batteries [51,152–154] because the vital source of all Li ions in a lithium-ion battery is located at the cathode (positive electrode) [155]. Therefore, the decision on selecting positive electrodes relies upon rechargeable Li-metal or Li-ion batteries as shown in Figure 6. Li-metal will serve as a negatively charged electrode in rechargeable Li batteries, preventing lithiation with the positive electrode [156,157]. However, for Li-ion batteries, the positive source needs to act as a source of Li, due to the unfilled Li located in carbon-negative electrodes [158]. Nowadays, there are several types of cathode material that are used as shown in Table 3. The most commonly used cathodes are lithium cobalt oxide ( $\text{LiCoO}_2$ ) and lithium iron phosphate ( $\text{LiFePO}_4$ ) [159].

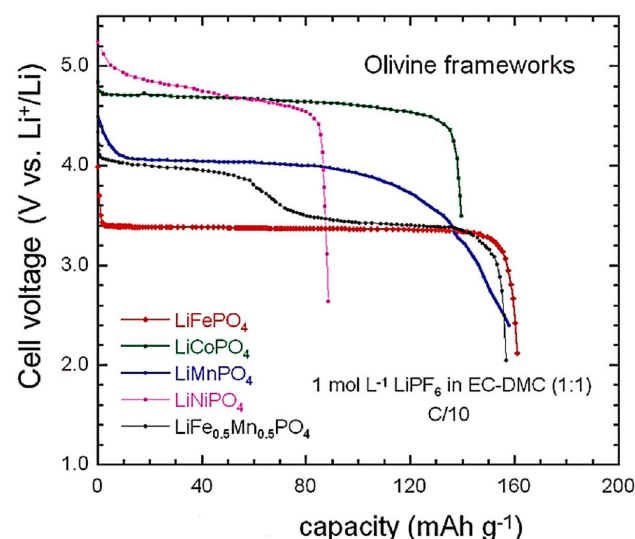


Figure 6. Voltage versus the capacity of various lithium-ion batteries [160].

**Table 3.** Characteristics for different types of battery electrode materials.

Crystal Structure	Compound	Potential vs. Li/Li <sup>+</sup> (V)	Specific Capacity (mAh/g)	Ref.
Layered	LiCoO <sub>2</sub>	3.9	274/155/140	[155,161,162]
	LiMnO <sub>2</sub>	3.3	285/140	[163,164]
	LiTiS <sub>2</sub>	1.9	239/210	[165]
	Li <sub>2</sub> MnO <sub>3</sub>	3.8	458/180	[166]
	LiNiO <sub>2</sub>	3.8	275/150	[167]
Olivine	LiFePO <sub>4</sub>	3.4	150/160/170	[168–170]
	LiCoPO <sub>4</sub>	4.2	167/125	[171]
	LiMnPO <sub>4</sub>	3.8	171/168	[172]
Spinel	LiMn <sub>2</sub> O <sub>4</sub>	3.9	148/120	[173,174]
	LiCO <sub>2</sub> O <sub>4</sub>	4.0	142/84	[175]

### 2.2.1. LiCoO<sub>2</sub>

In 1991, LiCoO<sub>2</sub> was utilized as a cathode material with the carbon anode, thus this marked the first successfully commercialized lithium-ion battery, which was introduced by Sony [155]. In a cubic close-packed configuration of transition metal, LiCoO<sub>2</sub> has O<sup>2-</sup> and Li<sup>+</sup> structured in alternating layers occupying an octahedral site [176]. However, the Li<sup>+</sup> ions can also be eliminated through an electrochemical procedure, separating themselves from the octahedral composition [83]. Thus, this will lead to the formation of a hexagonal close-packing oxygen layer in CoO<sub>2</sub> [177]. In addition, there are several advantages when using LiCoO<sub>2</sub> such as high theoretical specific capacity (274 mAh/g), high theoretical volumetric capacity (1363 mAh/cm<sup>3</sup>), low self-discharge, and high discharge voltage, and good cycling performance [178]. Despite that, there are several disadvantages that have to be conceded when using LiCoO<sub>2</sub> such as the cost of the material being relatively expensive, limited cobalt resources, low thermal stability, and toxicity caused by cobalt [179,180].

### 2.2.2. LiFePO<sub>4</sub>

LiFePO<sub>4</sub> (known as olivines) have a flat discharge plateau structure and sufficient capacity. Moreover, there are several benefits when utilizing LiFePO<sub>4</sub> in terms of stability, temperature tolerance (−20 to 70 °C), harmlessness, and minimal reduction in performance over the life of a battery [181–183]. Furthermore, LiFePO<sub>4</sub> provides significant protection compared to using Li–cobalt as a cathode; thus, this will allow them to be used for higher-level applications [184]. In addition, the iron (Fe) transition metal is inexpensive, accessible, and environmentally sustainable [185]. Regardless of the beneficial factors mentioned above, LiFePO<sub>4</sub> is a poor conductor of electricity and the charging voltage is below 4 V for each cell [170]. In spite of this, two methods were implemented in order to improve the problems. The particle size may be reduced using the first technique, and the conductive carbon nanostructure of LiFePO<sub>4</sub> can be used for the second [170]. Even Though the development of olivines with transition metals can be challenging and can have smaller capacity, they are able to show a high discharge potential and energy output values [186,187].

### 2.2.3. LiMnO<sub>2</sub>

One of the most promising cathode materials for the upcoming lithium-ion battery generation is LiMnO<sub>2</sub>, since it demonstrates a number of benefits, including the following: large energy density, environmental friendliness, plentiful manganese resources, high discharge capacity, etc. [188,189]. These materials outperform commercially available cathode materials in terms of capacity and are anticipated to meet the needs of applications involving electric vehicles. According to Freire et al., a mechanochemical synthesis process might produce a rock-salt structure of Li<sub>4</sub>Mn<sub>2</sub>O<sub>5</sub> with a high capacity of 355 mAhg<sup>−1</sup> [190]. However, the Jahn–Teller phenomenon linked to high-spin Mn<sup>3+</sup>, which results in significant structural deterioration and quick capacity fading, has prohibited the practical use of

LiMnO<sub>2</sub> cathodes in LIBs [187,191]. During charge–discharge cycles, LiMnO<sub>2</sub> displays low structural stability and irreversible phase changes. LiMnO<sub>2</sub> cannot be used as a suitable substitute for LiCoO<sub>2</sub> because of the structural instability brought on by Jahn–Teller distortion. The layered structure might be significantly stabilized by Co and/or Ni replacements for Mn, such as LiNi<sub>0.5</sub>Mn<sub>0.5</sub>O<sub>2</sub> and LiNi<sub>0.33</sub>Co<sub>0.33</sub>Mn<sub>0.33</sub>O<sub>2</sub>; however, this method removes the benefit and makes the cathode thermally unstable [192,193].

#### 2.2.4. LiNi<sub>x</sub>Co<sub>y</sub>Mn<sub>1-x-y</sub>O<sub>2</sub> (NCM)

LiNi<sub>0.33</sub>Mn<sub>0.33</sub>Co<sub>0.33</sub>O<sub>2</sub> (NMC) is a layered oxide cathode material with improved structural stability at higher voltages, greater environmental compatibility, reversible capacity, stable structure, cheap cost, and good thermal stability [194,195]. The electrochemical performance of typical bulk micro-sized cathode material was restricted, and it was unable to satisfy the needs of batteries. Alternative methods for preparing porous materials include nanoparticles, hollow spheres, nanowires, and porous materials. These methods are intended to maintain good particle–particle interaction and the appropriate particle size while boosting rate capability [196,197]. As the Mn<sup>3+/4+</sup> band sits above the Ni<sup>2+/3+</sup> band in NMC, Mn<sup>3+</sup> has a tendency to be oxidized during synthesis to Mn<sup>4+</sup> by reducing Ni<sup>3+</sup> to Ni<sup>2+</sup>. This implies that Mn<sup>4+</sup> aids in the integration of Ni as a stable Ni<sup>2+</sup> into NMC and acts as a structural stabilizer without taking part in the charge–discharge process. In comparison to the other samples, the NCM sample of the cathode at 850 °C for 12 h exhibits a maximum discharge capacity of 207.6 mAhg<sup>−1</sup> and 93.1% coulombic efficiency (CE) [194].

#### 2.2.5. V<sub>2</sub>O<sub>5</sub>

As a well-known transition-metal oxide, vanadium pentoxide (V<sub>2</sub>O<sub>5</sub>) has been thoroughly researched. Three Li ions may be stored in one formula unit of V<sub>2</sub>O<sub>5</sub> [198]. Along with the benefits of high output voltage, simple access to numerous sources, excellent safety, and admirable energy density, it also features Li-ion intercalation characteristics [199,200]. In addition, unlike LiFePO<sub>4</sub>, V<sub>2</sub>O<sub>5</sub> falls into the 2D category in terms of both structural and ionic transport characteristics, and at 25 °C, it has a conductivity in the order of 103 S cm<sup>−1</sup> [201]. The key benefits of V<sub>2</sub>O<sub>5</sub> are that it is simple to access the lower oxidation states of vanadium (+4 and +3), and that it can conduct both ions and electrons after being intercalated with electron-donating ions [200]. Delmas et al. demonstrated for the first time in 1991 that deep discharge with intercalation of more than two Li ions causes an irreversible loss of crystallinity with the structural change to ω-Li<sub>x</sub>V<sub>2</sub>O<sub>5</sub> [202]. The electrochemical characteristics of ω-Li<sub>x</sub>V<sub>2</sub>O<sub>5</sub> are dictated by its atomic-scale structure and the structural transition between the Li<sub>x</sub>V<sub>2</sub>O<sub>5</sub> phases, which also determine its potential as a cathode material.

### 2.3. Electrolyte

In addition to the electrodes, the third main part of a battery is the electrolyte; batteries typically depend on a vigorous electrolyte such that the electrode can achieve high efficiency [203]. Despite that, the electrolyte must be deliberately selected to prevent degradation or decomposition in the redox environment, which involves both the anode and cathode side and the voltage range [204]. Moreover, the design and composition of the electrolytes should be closely aligned with the surface quality of the electrodes [205]. One of the easiest solutions to the issue with high-energy electrodes may be to adjust the electrolytes [206]. For example, fluoroethylene carbonate has been found to limit dendritic development by creating a stable LiF solid electrolyte interphase (SEI) layer on the Li anode [207]. In addition, there are several types of electrolytes as listed in Table 4.

**Table 4.** List of basic electrolyte characteristics.

Electrolyte Properties	Ionic Conductivity	Contact Properties	Thermal Stability	Electrochemical Stability	Safety	Ref.
Liquid	High	Good	Poor	Poor	Poor	[208,209]
Solid	Low	Poor	Good	Good	Good	[209]
Gel	Medium	Medium	Poor	Poor	Medium	[209]

### 2.3.1. Liquid Electrolyte

Liquid electrolyte is considered to have the most important properties in terms of the ability to preserve steady contact with electrodes and high ionic conductivity [210]. In addition, liquid electrolytes were the most essential for the creation of nanostructured electrodes due to the combination of bulk and interfacial properties [211]. Thus, these will achieve a high specific surface area from the steady interaction between electrolyte and electrodes [212]. Despite that, there are several drawbacks to utilizing liquid electrolytes such as stability, flammability, and leakage [213]. Furthermore, the liquid electrolyte is the most commonly used since the first development of Li-ion batteries [214]. There are several forms of liquid electrolytes: organic, aqueous, water-in-salt, or ionic liquid. All of them have particular benefits and drawbacks, as mentioned below.

### 2.3.2. Organic Electrolyte

The most well-known types of electrolytes are organic electrolytes, which activate the batteries in mobile phones, laptops, and even automobiles [215]. Typically, an organic electrolyte consists of an inorganic solute dissolved in a very low electrical conductivity organic solvent. Moreover, organic solvent ionic conductivity is about 10 mS/cm [216]. The large electrochemical window of electrolytes, as opposed to water, makes it possible to identify their propensity for organic solvents [217]. Previously, there were several organic carbonates that have been utilized for Li-ion batteries, such as ethylene carbonate (EC), propylene carbonate (PC), and dimethyl carbonate (DMC) [218]. For EC, passivation layers and its high temperature resulted in it becoming an integral part of the mixture of carbonate solvents. Moreover, for operating Li-ion batteries, the required voltage is 4 V [219]. Unfortunately, due to the anodic stability limit, it is difficult to utilize carbonate anodes for high voltage (>5 V) systems [220]. There are also a number of chemical substances that have been used in Li batteries, including esters, ethers, and sulfones [221–223]. Furthermore, organic solvents are selected where different solvents are examined based on physicochemical properties, environmental impacts, and manufacturing paths for Li-ion batteries [220]. However, flammability is the main drawback of utilizing organic electrolytes leading to overcharging of batteries, internal shortening, or high-temperature exposure [224]. Hence, safety is a significant concern when manufacturing a battery and the selection of an electrolyte that is not potentially unsafe is of paramount importance [91].

### 2.3.3. Aqueous Electrolyte

When utilizing aqueous electrolytes, there are several advantages in terms of inexpensive and high ionic conductivity compared to organic counterparts [225]. In addition, the aqueous electrolyte can absorb significant amounts of heat and have greater thermal capacitance [226]. During overcharge, the positive electrode will emit oxygen, which then disperses through the separator and reduces to H<sub>2</sub>O at the negative electrode [227]. This process is known as the oxygen cycle, and it reduces the battery overcharge tolerance [228]. Thus, water is an excellent electrolyte solution since the electrolyte is not polluted by the decomposition products, hydrogen and oxygen [11,229]. Moreover, there are numerous benefits when utilizing water as an electrolyte solution, such as being environmentally friendly, easily accessible, and plentiful [230]. Regardless of the beneficial factors mentioned above, the primary drawback of utilizing aqueous electrolytes is that the decomposition voltage is extremely low (=1.23 V) compared to organic electrolytes (>3.0 V) [231]. Furthermore, all

aqueous electrolyte battery systems that have a higher voltage of 1.23V are classified as hazardous and hence have a certain degree of self-discharge [232]. To summarize, selecting electrode materials that are compatible with water is one of the main problems; thus, the redox reaction takes place within a restricted electrochemical window [91].

#### 2.3.4. Water-in-Salt Electrolyte

An aqueous electrolyte resolves the problem of utilizing an organic electrolyte, in which in their structures are made of flammable and poisonous materials [233]. However, there are a variety of drawbacks for aqueous electrolytes in the electrochemical window, including its severe reduction, restricted energy, and lack of economic viability [234]. Thus, water-in-salt electrolytes are more beneficial in terms of the electrochemical window compared to aqueous electrolytes [235]. For instance, the electrochemical window of an aqueous electrolyte is 1.23 V, thus the majority of the electrochemical couples (anode and cathode material) utilized in Li-ion batteries can be difficult to sustain. When utilizing water-in-salt electrolytes, the electrochemical window is about 3 V [236]. Moreover, the term “water-in-salt electrolyte” refers to a substance in which salt, rather than water, dominates both the total mass and volume [235]. Thus, lithium-bis(trifluoromethane sulfonyl)imide (LiTFSI) was selected as the salt due to several benefits, such as easily dissolving in water and high hydrolysis durability [237]. Hence, compared to the aqueous electrolyte, the number of water molecules available that are part of ion solvation shells will allow higher voltage. However, it is significantly expensive to use water-in-salt electrolytes because lithium is a rare element that is allocated in the Earth’s crust [238]. Moreover, LiTFSI is harmful upon touch or absorption and may inflict damage on organs [91]. Nevertheless, by utilizing water-in-salt electrolytes, they offer several major potential benefits such as being non-flammable, cost-competitive, and with a high electrochemical stability window [239].

#### 2.3.5. Ionic Liquid Electrolyte

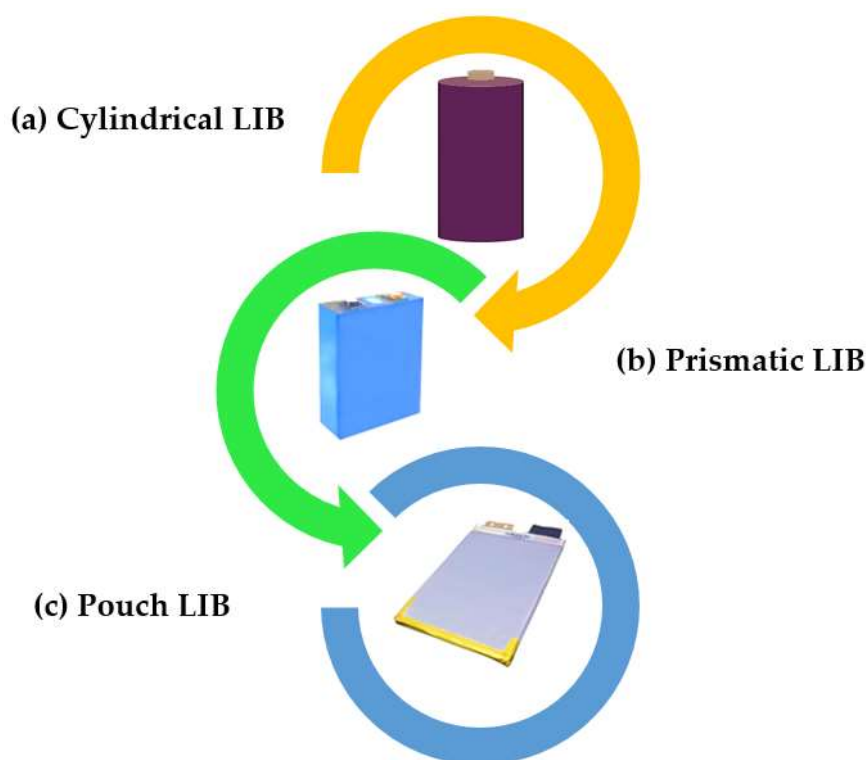
Ionic liquids (ILs) are molten salts with an exceptionally low melting point of less than 10 °C and are normally in liquid form at room temperature [240]. Nevertheless, ILs offer numerous beneficial properties such as low vapor pressure, and being thermal stable, environmentally friendly, non-poisonous, and non-flammable [241]. Moreover, ILs are able to generate higher electricity conductance compared to organic electrolytes because they are a pure ionic mixture [242]. ILs have a comparable overwhelming excess of ions compared to water-in-salt electrolytes. Thus, there are no significant differences with the water-in-salt electrolyte, whereas ILs are greater compared to organic counterparts [217]. In contrast, ILs are able to obtain a comparatively high electrochemical stability window of 4 V [243]. There are a number of prerequisites needed to create high conductivity [244], including flowing liquid electrolytes and cations and anions that are atypically generated [91]. In addition, although having ions that are similar in size to those in highly regular salt, they are capable of generating lower melting points [245]. The most significant drawback of utilizing ILs is their high viscosity, which can lead to poor electrode surface wettability, and they are relatively expensive [246]. The positively charged ion can intercalate interlayers that cause mechanical instability of the electrodes and may require additives to improve the electrochemical performance of lithium-ion batteries [247]. All of the different types of electrolytes are summarized in Table 5.

**Table 5.** Different types of electrolytes.

Electrolyte	Toxicity	Low-Temperature Performance	Thermal Stability	Ion Conductivity	Ref.
Organic	Medium/High	Relatively good	Poor	High	[248,249]
Aqueous	Low	Poor	-	High	[205,249]
Water-in-salt	Low	Good	Low	-	[250,251]
Ionic liquid	Low	Poor	High	Good	[205,248,249]
Solid polymer	Low	Poor	Excellent	Low	[248,252]
Gel polymer	-	High	Good	High	[205,248]
Inorganic solid	-	-	High	High	[253]

### 3. Fabrication Technique of LIBs

There are three different cell types used in the production of Li-ion batteries: cylindrical, prismatic, and pouch (Figure 7). Additionally, the manufacturing methods for these cells are one of the primary emphases of this work [254].



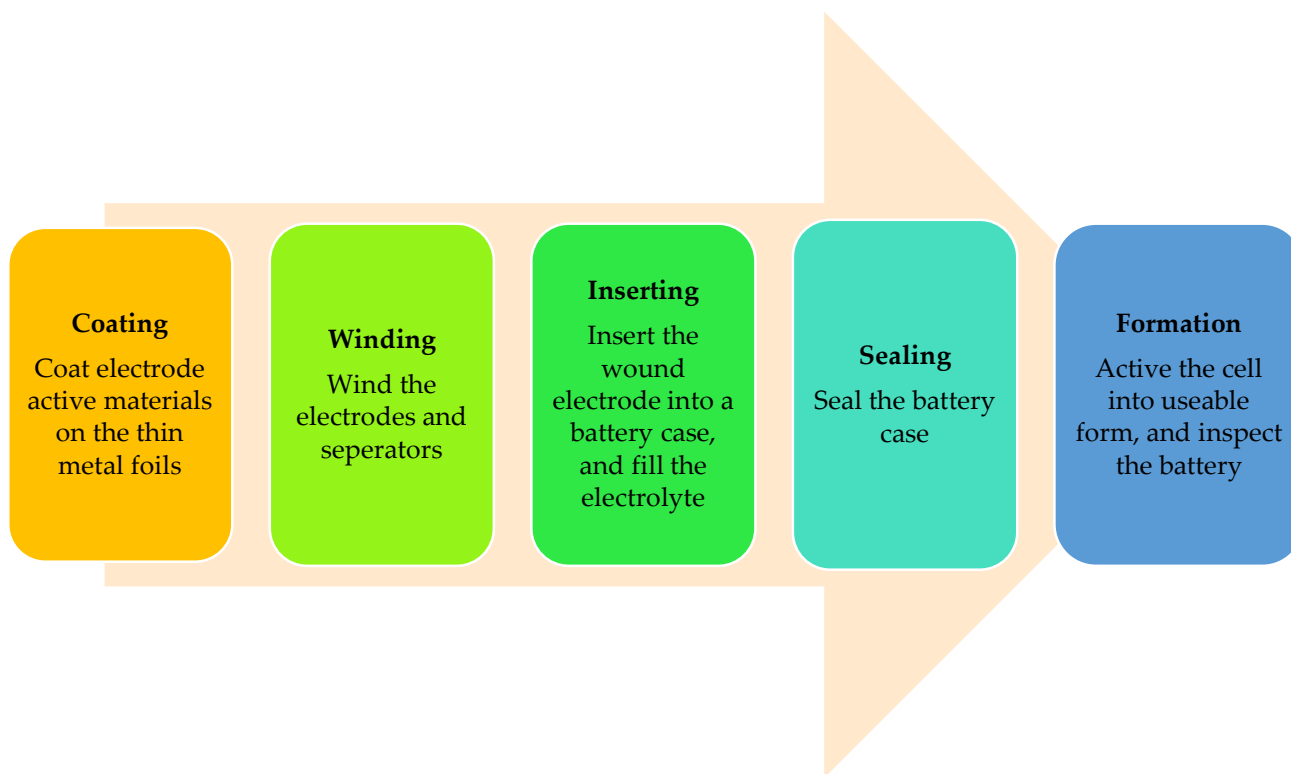
**Figure 7.** Various cell designs include (a) cylindrical; (b) prismatic; (c) pouch [255].

A number of methods are used to create Li-ion batteries, including coating the active components of the positive and negative electrodes with thin metal foils, winding the electrodes with separators in between them, placing the wound electrodes in a battery case, filling the electrodes with electrolytes, and sealing the battery case to complete the process [256]. Figure 8 depicts the overall development process from a high-level perspective for the Li-ion batteries.

Li-ion battery manufacturing may be broken down into five primary steps: (1) mixing, pressing, coating, and slitting the positive electrode and negative electrode material on thin metal foils; (2) spooling the separator, positive, and negative electrodes; (3) inserting and charging the wound cell core with electrolyte into the battery case; (4) sealing or closing the battery case; and (5) formation, aging, and inspection as shown in Figure 9 [257].



**Figure 8.** A general description of the Li-ion battery development process [258].



**Figure 9.** Li-ion battery component production procedure [259].

### 3.1. Cylindrical Cell Fabrication

The first step for manufacturing a cylindrical cell is mixing and coating [257]. The cathode electrode is made of active materials such as  $\text{LiCoO}_2$ ,  $\text{LiFePO}_4$ , or  $\text{LiMnO}_2$ , whereas the anode electrode is either carbon or graphite [260]. During the manufacture of the electrodes, the active electrode materials are coated and calendared on both sides of metal foils [261]. The two electrode components are combined in a mixer with a conductive binder to create a slurry that is spread out across the surface of the foil as it goes through the slitting device [262]. An edge of a knife is positioned above the foil. The thickness of the coating between the knife edge and the foil can thus be checked using this method, and it will also guarantee that all the pieces fit into the case during final assembly. After that, the coated foil is heated in a drying oven so that the solvent is vaporized and the active ingredients are exposed [263]. Moreover, the optimal electrode thickness for high-energy Li-ion batteries is between 70  $\mu\text{m}$  and 320  $\mu\text{m}$  [264]. The coated foil is then transferred to a slitting machine where measurement strips will be cut off using a cell-shaped cutting tool [265]. In addition, the slitting procedure needs to be precise, since any burr edge on the foil strips might result in a short circuit in the cell.

In order to create a reel with a cylindrical mandrel, the second step involves loading the winding machine with both electrode strips [257]. As a result, before the strips are utilized, the winding machine functions automatically [259]. Additionally, the separator is applied to prevent short circuits by disconnecting the anode and cathode. For constructing a cylindrical cell, the cell design requires two strips of electrodes [266]. Consequently, both terminals of electrodes are joined independently with a single tab welded to the bare section. To prevent mechanical damage to the battery, such as shock or vibration, the center



pin is placed into the reel afterward [267]. The central pin is used to assist the assembly of the electrodes and acts as a passage for the discharge of internal reactions in gas [259]. A welded electrode is put through the center pin’s hole to solder the anode tab into the cell cover. The positive temperature coefficient (PTC) thermistor that is built into the casing and vents is used to protect the electrode arrangement subassembly as it is joined to the terminals [268].

In the third phase, the electrolyte is loaded into the cell under a vacuum [269]. The suitable location for conducting the process is in a dry room to avoid any water reacting to the electrolyte. In addition, some moisture exposure may lead to the decomposition of the electrolyte by the emission of toxic gases. In order to load the electrolyte, a device known as a precision pump is utilized, then a vacuum is utilized to ensure that the separator pores and electrodes are saturated and fully loaded with electrolytes. Once the loading procedure is complete with any necessary protection measures, as indicated above, the cap is then put on.

In the last phase, the cell must be sealed using polymer compression. Secondly, using a heat welding procedure, the safety vents may be affixed to the top of the positive terminal. In addition, this will allow the release of gases within the battery and reduce the effects of excessive heat, such as fire or explosion [269]. In the fifth phase, the length of the cell is measured (cycle aging), and any design problems such as misalignment during the winding process are found [259]. Lastly, a serial number label that offers particular information from cell manufacture is applied to the casing in order to categorize the cell.

### 3.2. Prismatic Cell Fabrication

Prior to applying the final manufacturing step, the coating and winding processes used to create Li-ion batteries for prismatic cells are identical to those used to create cylindrical cells [265]. However, the method varies from the cylindrical cell geometry because of the properties of the prismatic geometry (Table 6). The prismatic cell utilizes an aluminum cell shell to minimize the weight, and to reduce the battery thickness ( $\leq 4$  mm) an aluminum-laminated material is utilized [270]. On the bottom of a cylindrical case, an insulating layer is inserted and an insertion device is utilized to position the wound coil in the case. Thus, the positive electrode tab for the prismatic cell is soldered to the aluminum case [271]. The negative electrode tab for the cylindrical cell is soldered at the bottom of the can. In addition, laser-welding is utilized to assemble the top cap and a tab to the cell case for the prismatic cell. The cells must be vacuum-dried overnight or for 24 h to remove all water if the laser-welding technique is not carried out in a dry chamber [272]. Finally, a cap insertion device is utilized to put the cap into the casing. Thereafter, a laser-welding device is used to partly solder the cap to the casing. The frameworks of the case and cap are then entirely laser-fused together. After that, with laser welding equipment, solder is used to seal the electrolyte fill tube [273].

**Table 6.** Specific energy, specific power, energy density, and power density.

Types	Can Type	WCell (g)	VCell (l)	Specific Energy (Wh/kg)	Energy Density (Wh/l)	Specific Power (Wh/kg)	Power Density (W/l)	Ref.
Cylindrical (18,650)	Steel	42.3	0.0165	166	425	168	430	[274]
Prismatic (103,450)	Al	38.5	0.017	173	391	171	387	[274]

For a cylindrical cell, it has a cylindrical structure and more space is occupied. In addition, the manufacturing cost for the cylindrical cell is low [255]. Thus, they are widely utilized battery cells that can be allocated in laptop batteries, electrical vehicles, etc. For prismatic cells, the designed structure is thin and light. Moreover, to make the prismatic

cell more stable, they will use casings made of either steel or aluminum. However, the manufacturing cost for the prismatic cell is expensive.

According to Table 6, there are a number of factors that can affect the specific energy and energy density for both cell types, including cell design, cell geometric form, and can material. Additionally, even if a prismatic cell has a larger volume than a cylindrical cell, the geometric variances between cylindrical and prismatic cells may lead to changes in the specific energy of the cells and differences in the density of cells [255]. Furthermore, compared to cylindrical cells made of steel, the material of the container may have a major role in improving the cell-specific energy of prismatic cells made of aluminum. According to Table 6, the cylindrical cell has a higher energy density than the prismatic cell because it radiates heat and regulates temperature more efficiently, allowing it to store more energy. The cylindrical cell has a higher power density compared to the prismatic cell; as a result, the cylindrical cell can output large amounts of energy depending on its mass.

Cylindrical cell production can have a better automation method and techniques that can improve consistency and can withstand high internal pressures without deforming [275]. Therefore, the cylindrical cell is significantly cheaper and can be produced at a much more rapid rate than prismatic cells. Moreover, the design of cylindrical cells has greater temperature tolerance than prismatic cells [276]. This is because of how all of the prismatic cells are packed together, which may lead to short-circuiting and a greater chance of design inconsistency. However, the main downfall of utilizing cylindrical cells is the limited usage space.

Despite that, prismatic cells are lighter, thin, and manufactured to conserve space. Therefore, the thin prismatic cell's rectangular shape makes it simpler to attach four cells and construct a 12 V battery pack [277,278]. In certain circumstances, the larger size of the prismatic cell may be attractive, but it also lessens the likelihood that such a battery could be utilized in a highly automated environment. However, there are several disadvantages to using prismatic cells; when one of the cells fails, the extra power will have an impact on the entire battery pack, making it more challenging for the battery management system to accurately monitor heat and prevent battery overcharging. Additionally, the production and manufacture of prismatic cells is costly compared to cylindrical cell batteries [279]. For this study, cylindrical cells are the preferred cell due to several beneficial outcomes such as their lower cost, better performance, longevity with less concern for space usage, higher power density, and higher energy density compared to prismatic cells.

### 3.3. Pouch Cell Fabrication

Due to their high energy density, excellent power performances, and other design advantages, pouch cells are the primary choice for many manufacturers. An aluminum-coated plastic film encloses the majority of the components of the cell, which is generally a type of soft battery. Only two tabs are visible, and they are all joined to the pouch's current collectors via welding. The positive and negative connector tabs are carried by these extremely conductive tabs, which also enable the extraction of electric energy from the pouch cell. Electrode cutting/trimming, electrode stacking, tab welding, pouch sealing, electrolyte injection, formation, and final degassing and resealing are all steps in the process of creating a pouch cell [280].

The cell parts are stacked repeatedly with numerous layers inside the pouch. Anode, cathode, and separator sheets must be separated from the roll material in order to produce the pouch cell, and this process is referred to as separation. Ions move through the liquid electrolyte between the cathode and the anode during the charging and discharge of the cell. To prevent moisture from harming the battery cell components, the electrode sheets are prepared and the pouch cell is constructed in a controlled atmosphere. Welding a metal strip to the current collector constitutes the first stage of assembly. After that, the cell stack is put inside the pouch foil. The bag foil is deep-drawn for this purpose in a previous production phase. In most cases, an impulse or contact sealing technique is used to gas-tightly close the pouch cell on three sides. The liquid electrolyte is subsequently injected

into the cell using an electrolyte-filling mechanism. However, the pouch cell fabrication is finished after the cell is vacuum-sealed [281,282].

Comparing pouch cells to cylindrical cells, a pouch cell can store much more energy in a given physical area. Pouch cells typically weigh less than a prismatic or cylindrical battery of the same size. Although the energy density of flat pouch cells is higher than that of other cell shapes, making stacking pouch cell batteries is significantly more challenging.

#### 4. Advancement in Battery Technology

Researchers are working hard to advance battery technology, trying to increase capacity, speed up charging, and improve performance at any temperature. The new era in battery technology begins with the development of nano-sized lithium-ion batteries. One day, owing to this remarkable technology, we will be able to shape synthetic materials into the shape of a house using nanotools to construct one. This grain-sized lithium-ion battery has the potential to transform portable power sources and pave the way for lithium-ion batteries smaller than a grain of salt to power tiny devices in the future. This idea inspired Wang et al. [283] to propose generating micrometer-sized Si/C particles by embedding Si nanoparticles into graphene oxide (GO)-based micro rolls, which not only have a higher volumetric capacity of  $2506 \text{ mAh cm}^{-3}$  and a higher initial coulombic efficiency of 85.1%, but also have a higher tap density of  $0.62 \text{ g cm}^{-3}$  and higher cycling performance of  $660 \text{ mAh cm}^{-3}$  after 500 cycles at  $2 \text{ Ag}^{-1}$ . The following Table 7 is a list of new-generation lithium-ion batteries and their functioning characteristics.

**Table 7.** A list of lithium-ion batteries and their functioning characteristics [284].

Components	Nominal Voltage (V)	Cycle	Applications
Lithium–iron phosphate batteries ( $\text{FeLiO}_4\text{P}$ ); phosphate as a cathode	3.2	2000+	Electric vehicles, drones, power backup
Lithium cobaltate ( $\text{LiCoO}_2$ or $\text{CoLiO}_2$ ); a cobalt oxide cathode and a graphite carbon anode	3.6	1000+	Cameras, tablets, laptops, and smartphones
Lithium manganese oxide batteries ( $\text{LiMn}_2\text{O}_4$ ); lithium manganese oxide cathode	3.6	1000+	Medical equipment, portable power tools, and certain hybrid and electric cars
Lithium nickel manganese cobalt oxide batteries ( $\text{LiNiMnCoO}_2$ ); cathode made of nickel, manganese, and cobalt	3.7	500–1000	Power tools as well as e-bikes, scooters, and certain electric vehicle powertrains
Lithium nickel cobalt aluminum oxide (NCA) batteries ( $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$ )	3.6	2000+	Tesla favors NCA batteries for applications involving electrical power transmission and grid storage
Lithium titanate ( $\text{Li}_2\text{TiO}_3$ ) has a rapid recharge time as a result of its better nanotechnology	3.6	2000+	Electric cars and charging stations, constant power sources, energy storage for the sun and wind, solar-powered streetlights, telecommunications systems, and aerospace and defense technology

Another advancement is the fast charge or temperature working window, which may be adjusted thanks to the wide range of cell design options. Chen et al. [285] suggested that for adaptive online capacity prediction while receiving fast charging, a long short-term memory network-based transfer learning strategy may be created. In this research, a revolutionary voltage function is added that allows for a quick 10 minutes charge to reach an 80% state of charge. This attribute is extremely useful and is simple to test in all rapid charging scenarios.

The adoption of silicon as the anode material is another advancement since it is more prevalent, less expensive, and has a larger theoretical discharge capacity than graphite. In

order to increase the intercalation sites for Li atoms, Liangruksa et al. [286] employed SiO, which is highly adsorbed on divacancy graphene with an adsorption energy of  $-4.59$  eV. This increased Li's adsorption energy from  $-1.18$  eV to  $-4.15$  eV in comparison to pure graphene. Tang et al. [287] also used Si anodes, which were generated using a novel method of bonding between conductive agents and active components/binders. The Si anode demonstrates excellent electrochemical performance by producing the advantageous solid electrolyte interphase (SEI) composition.

How lithium-ion batteries should be disposed of after use is now the most important topic. When LIBs reach the end of their useful lives, they can affect the environment or human health because they are utilized in so many products, including electronics, toys, wireless headphones, handheld power tools, small and large appliances, electric cars, and electrical energy storage devices. When disposed of in domestic garbage and curbside recycling bins, these batteries and the devices they are in, though often safe when in use, might provide a fire hazard. When batteries are handled properly at end-of-life, they can be safely transported to recycling facilities, where the process of recycling will allow necessary resources to re-enter the supply chain.

### 5. Recycling Spent Li-Ion Batteries

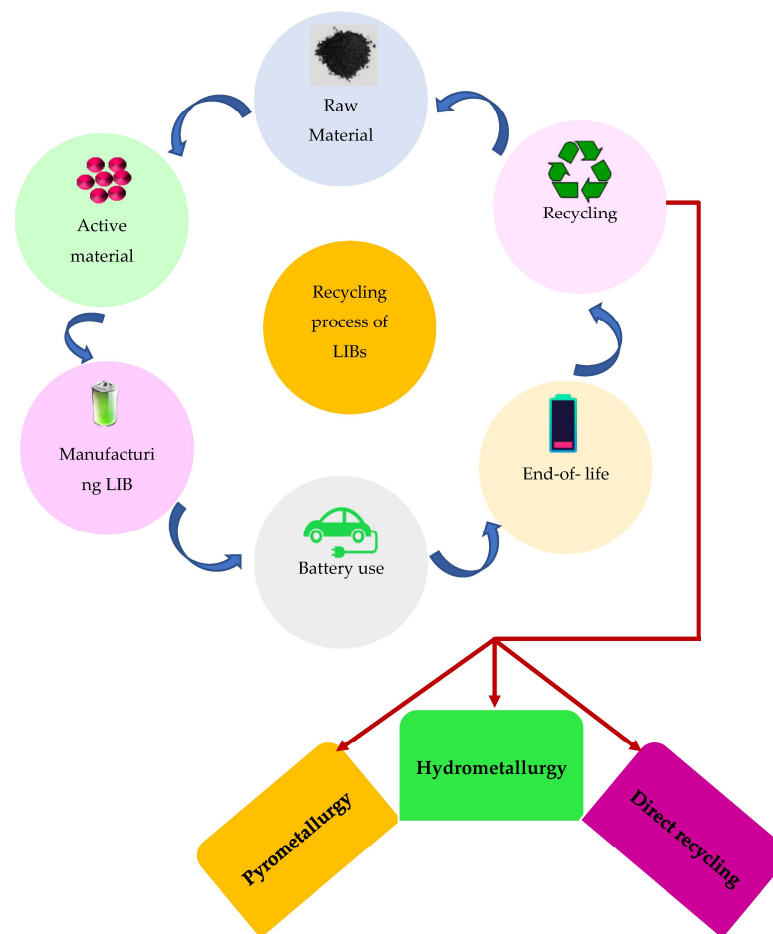
As technology progresses, the production process of these batteries becomes more advanced and proficient to meet the growing demand, which in turn causes the older models to eventually become obsolete. The older metal-ion batteries typically reach their end-of-life (EOL) and are then replaced and discarded, which unfortunately leaves behind the undesirable waste of precious minerals, including Li, Co, Ni, and other metals as scrap. These scrap metals require appropriate disposal to reduce harmful pollution [288]. Currently, to avoid wasting valuable metal resources, several avenues of research and development for recycling and repurposing LIBs are being conducted in order to limit waste generation and conserve the world's natural resources. Completely recycling a battery requires a number of phases, and enhancing the recyclability of new batteries entails a notable improvement in the sustainability and profitability of their recycling procedures as shown in Figure 10. Researchers are currently studying ways of recycling spent metal-ion batteries by re-harvesting the valuable metals left from the waste cathode materials to be remanufactured [289].

At present, research is being performed on recycling used and old LIBs, with the main goal of recycling the precious metals inside the waste cathode materials [40], which may include  $\text{LiCoO}_2$  (LCO),  $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$  (NCM),  $\text{LiFePO}_4$  (LFP), and  $\text{LiMn}_2\text{O}_4$  (LMO) [290,291]. Due to the high valence states of the transition metals and the robust chemical bonds of MO (M = Co, Ni, and Mn), the precious metals contained in waste cathode materials of LCO, LMO, and NCM have difficulty being directly recycled [292].

Recycling by remanufacturing LIBs with spent batteries is considered to be the most promising strategy for treating end-of-life LIBs without significant risk [288]. In addition, the detrimental environmental effects and contamination of the processes can be decreased with the aid of current advancements in research and industry related to pyrometallurgical, hydrometallurgical, and direct recycling [293]. Despite this, none of the battery recycling methods now in use are at their best; therefore, more obstacles must be overcome before the optimum process can be achieved [288]. If successful, effective recycling of the LIBs will allow for less demand for mining for the mineral deposits, which will help to slow down the depletion of the limited precious metals on Earth [294].

An important step in recovering spent LIBs is the pre-treatment of the batteries, such as discharging and dismantling the battery into its main components [295]. The materials for the cathode and anode are then separated, and they are crushed or ground into a fine powder. In particular, for hydrometallurgy and direct recycling operations, this method is essential for improved resource recovery efficiency and an energy-efficient recycling process [288]. The recovery of other less economically important elements or contaminants, such as Al, Fe, Cu, and C, as well as valuable metals including Ni, Co, and Li, makes this

phase crucial. According to the classifications given by various experts, the pre-treatment process is divided into a variety of phases and procedures [296].



**Figure 10.** Recycling of LIBs by using different recycling processes.

### 5.1. Discharging

The process of discharging spent LIBs is required in order to remove the potential of the cathode or anode short-circuiting, which is considered to be a possible fire hazard [289]. This process typically involves submerging the spent LIB into a saline solution (often NaCl) and discharging it over the course of 12 to 24 h. However, this process is not a simple task, and should only be conducted by someone more experienced and well-trained in handling LIBs who is familiar with the procedure. As a result, it is suggested that once the LIB has been entirely depleted, as a safety measure, one should not attempt to recharge it again. Even if the necessary gear is acquired, it is recommended to exercise caution when handling LIBs and it is advised to seek a local professional for help. It is important to keep in mind and be aware that LIBs are very poisonous and harmful, as they can cause wounds if used without supervision.

### 5.2. Dismantling

Lithium can only be separated at the end of the chain, which is a feature shared by all LIB recycling methods. Lithium carbonate contamination and high losses are related to this. In order to extract Li from a combination of spent LIB components (Co, Li), Pavón et al. [297] showed a four-stage counter-current extraction procedure. The authors demonstrated that not only Li as  $\text{Li}_2\text{CO}_3$ , but also Al, Mn, and Ni can be recovered from LIB black material formed from used LIBs with great selective power, considerably advancing the objectives of the circular economy. Yet a thorough economic, technological, and environmental assessment of the entire LIB recycling process is still required. The mechanisms involved in

grinding have not been well studied, despite the fact that there have been several studies on Co recovery in used LIB recycling.

## 6. Cutting-Edge LIB Recycling Techniques

As was already noted, the three main recycling methods now used are pyrometallurgy, hydrometallurgy, and direct recycling (Figure 11). Pyrometallurgy is a recycling method that includes high-temperature melting, material burning, and material separation. Hydrometallurgy is a recycling method that functions through the application of aqueous chemistry, via acid or alkali leaching, followed by concentration and purification of the materials. Lastly, direct recycling is a recycling technique that physically handles the spent batteries to recover the desired active materials within. Usually, this technique is carried out to preserve the original complex structure of the materials. Although pyrometallurgy and hydrometallurgy are already used on a large scale in industry, direct recycling is still only used in laboratory trials [298].

These recycling techniques were founded on the idea that transition metals in used cathode materials need to first be reduced from their high to their low valence states, and that in order to break the MO bonds during the recovery process, either a chemical reaction or physical force is required [299]. Several methods, such as carbothermic reduction [300], and decreasing leaching by  $\text{H}_2\text{O}_2$  [86],  $\text{NaHSO}_3$  [301],  $\text{Na}_2\text{S}_2\text{O}_5$  [302], or glucose [303,304], were described for the efficient leaching of precious metals from wasted LIBs. In contrast, there are two types of recycling processes currently used for used LFP cathode materials: direct rehabilitation and hydrometallurgical [305,306]. Despite the direct regeneration procedure being rather straightforward, contaminants in used cathode materials and the damaged structure have an effect on the electrochemical performance of newly manufactured materials [307,308].

### 6.1. Pyrometallurgy

The pyrometallurgical process, which typically entails two phases, is a high-temperature melting procedure [309]. In a smelter, where organic materials such as plastic and the separator are burnt away, compounds are broken down. LIBs are first burned; then, carbon reduction is used to create new alloys. The simplicity and maturity of this procedure are its key benefits. A mixture of LIBs and NiMH batteries can be recycled; sorting and size reduction are not required. The end product comprises fundamental, elemental building blocks that may be used in the synthesis of novel cathode materials with a variety of chemistries. The primary drawbacks are  $\text{CO}_2$  production and excessive energy use during the smelting process [310]. Alloy needs more processing, which raises the overall cost of recycling. Many LIB materials are not retrieved (e.g., plastics, graphite, and aluminum). The low Co content may make the business model for EV batteries less effective. Table 8 displays the developments in pyrometallurgical recycling techniques reported in the recent literature.

Various reduction methods are the subject of intense laboratory study and research interest [311–314]. While extracting compounds from LIBs, reduction roasting plays a crucial role. Xiao et al. [313] proposed that using a solid-to-liquid ratio of  $25 \text{ g L}^{-1}$  and a purity rate of 99.7% for  $\text{Li}_2\text{CO}_3$ , an ideal Li recovery rate of 81.90% is attained at 973 K after 30 min. To demonstrate the viability of in situ recycling  $\text{Li}_2\text{CO}_3$  from wasted LIBs under enclosed vacuum conditions, three different types of spent LIBs, including  $\text{LiMn}_2\text{O}_4$ ,  $\text{LiCoO}_2$ , and  $\text{LiCo}_x\text{Mn}_y\text{Ni}_z\text{O}_2$ , are treated. According to Tang et al., the vacuum pyrolysis technique may achieve recovery rates of over 93% for Li and 99% for Co by varying the C/ $\text{LiCoO}_2$  mass ratio and the pyrolysis temperature [314]. There are a number of advantages that the pyrometallurgical process can provide the recycling system, such as simpler scaling and the use of fewer chemicals, but there are also a number of disadvantages to this process, such as the inability to recycle materials in the electrolyte, the loss of valuable Li metal and graphite in the anode, the emission of harmful gases, and the high energy intake.

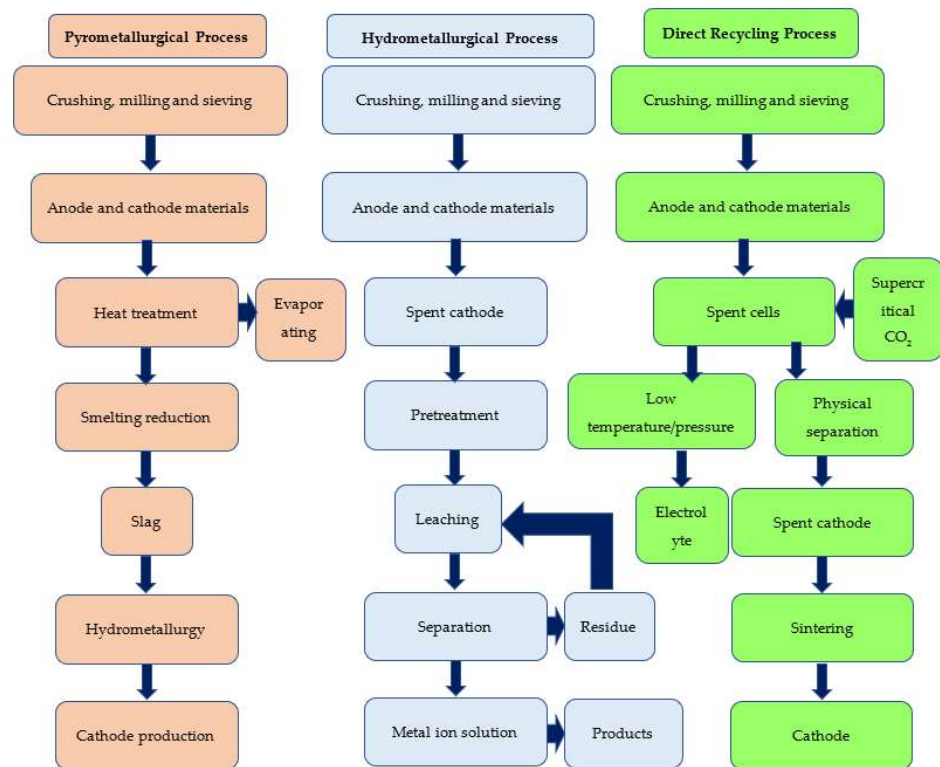


Figure 11. The main types of recycling processes for metal-ion batteries [288].

Table 8. Latest research on improvements in pyrometallurgical recycling [288,311–315].

Pyrometallurgical Procedure	Product and Productivity	Importance
Slag system: MnO-SiO <sub>2</sub> -Al <sub>2</sub> O <sub>3</sub> ; Smelting condition: 1475 °C, 30 min	Lithium-containing Mn-rich slag and the alloy Co-Ni-Cu-Fe; 99.79%, 99.30%, and 99.30% recovery efficiencies for Co, Ni, and Cu, respectively. 79.86% and 94.85% of Mn and Li were leached, respectively	Mn and Li are recovered preferentially using a new slag method
SiO <sub>2</sub> -CaO-Al <sub>2</sub> O <sub>3</sub> -Li <sub>2</sub> O make up the simulated slag; 1000 °C for 90 min of roasting 1000 °C for 30 min of oxygen-free roasting; wet magnetic separation 973 K for 30 min with a 1 kPa vacuum Roasting without oxygen: 1073 K, 45 min	LiCl; Li recovery rates from slag: 97.45%  Co, Li <sub>2</sub> CO <sub>3</sub> , and C and graphite recovery efficiency: 95.72%, 98.93%, and 91.05%  Li <sub>2</sub> CO <sub>3</sub> ; Li purity and recovery efficiency: 81.90% and 99.7%	Novel pyrometallurgical technology for recycling lithium from slag  Cobalt, Li <sub>2</sub> CO <sub>3</sub> , and graphite are recycled in-place in an LCO/C battery system.  Li <sub>2</sub> CO <sub>3</sub> recycling in situ from used LCO, LMO, and NMC
Reduction roasting: 650 °C, 3 h, 19.9% carbon dose; acid and carbonated water leaching  Roasting temperature 350 °C,	Li <sub>2</sub> CO <sub>3</sub> and Mn <sub>3</sub> O <sub>4</sub> ; Li recovery efficiency was 91.30%, while Mn purity was 95.11%. Li <sub>2</sub> CO <sub>3</sub> , NiSO <sub>4</sub> , CoSO <sub>4</sub> , and MnSO <sub>4</sub> ; efficiency of Li in water leaching: 84.7%; Ni, Mn, and Co acid leaching efficiency: >99% Li <sub>2</sub> CoO <sub>2</sub> -NH <sub>4</sub> Cl, 1 mol L <sup>-1</sup> oxalic acid, temperature 60 °C, time 1 h, and n(H <sub>2</sub> C <sub>2</sub> O <sub>4</sub> ): n(Co <sup>2+</sup> ) = 1.05:1	In situ recycling of Li <sub>2</sub> CO <sub>3</sub> and Mn <sub>3</sub> O <sub>4</sub> in LMO/C battery system An easy and effective method of recovering an NMC/C battery system is through reduction roasting. With water leaching at a solid-liquid ratio of 100 g/L, Li and Co had leaching efficiencies of 99.18% and 99.3%, respectively.
Vacuum pyrolysis, temperature range from 623 to 823 K	LiCoO <sub>2</sub> , solid-state reaction between Co <sub>3</sub> O <sub>4</sub> and Li <sub>2</sub> CO <sub>3</sub> at a Li/Co ratio of 1.05 at 1123 K for 13 h, and the ramping rate was 2 K/min	Vacuum pyrolysis, process performed at 673–723 K peel the electroactive materials off from the current collectors, leaving behind near-fresh Al and Cu foils, carbothermal reduction—the graphite to reduce and break down LiCoO <sub>2</sub> , producing Li <sub>2</sub> CO <sub>3</sub> and Co or CoO

## 6.2. Hydrometallurgy

In this procedure, material rescue is accomplished utilizing aqueous chemistry, by leaching in acids (or bases), followed by concentration and purification [316]. For LIBs, ions in solution are disconnected using a variety of methods (ion exchange, solvent extraction, chemical precipitation, electrolysis, etc.) and generated as diverse compounds. The primary benefits include the possibility to produce materials of high purity. The majority of LIB components can be restored while operating at low temperatures. There are also lower CO<sub>2</sub> discharges compared to the pyrometallurgical process. The primary drawbacks are the necessity of sorting, which necessitates more storage space and raises the cost and complexity of the operation [306]. Due to their comparable qualities, it can be difficult to separate some elements (Co, Ni, Mn, Fe, Cu, and Al) from the solution, which might increase expenses. The price of wastewater treatment and related expenses is high. Table 9 lists an overview of the hydrometallurgical techniques used to recover cathodes from used LIBs.

**Table 9.** An overview of hydrometallurgical techniques for recovering cathode from used LIBs.

Cathodes	Reductant	Leaching Agent	Leaching		Conditions	Ref.
			Li	Co		
LiCoO <sub>2</sub>	Ethanol	H <sub>2</sub> SO <sub>4</sub>	99%	99%	90 °C in 160 min, 3 mol/L H <sub>2</sub> SO <sub>4</sub> solution with 5 vol.% ethanol, at a S/L ratio of 20 g/L	[317]
LiCoO <sub>2</sub> , Li <sub>2</sub> CoMn <sub>3</sub> O <sub>8</sub> , (Li <sub>0.85</sub> Ni <sub>0.05</sub> )(NiO <sub>2</sub> )	1M H <sub>2</sub> SO <sub>4</sub> and 0.075 M NaHSO <sub>3</sub>	Diluted H <sub>2</sub> SO <sub>4</sub> (96%)	~96.7%	91.6%	Heating at 250–300 °C for 30 min	[318]
LiCoO <sub>2</sub>	H <sub>2</sub> O <sub>2</sub>	H <sub>3</sub> PO <sub>4</sub>	99%	99%	40 °C (T), 60 min (t), 4 vol.% H <sub>2</sub> O <sub>2</sub> , 20 mL·g <sup>-1</sup> (L/S) and 0.7 mol/L H <sub>3</sub> PO <sub>4</sub>	[319]
LiCoO <sub>2</sub>	H <sub>2</sub> O <sub>2</sub>	HNO <sub>3</sub>	75–85%	40–85%	75 °C; 0.5 h; 1 M; 20 g L <sup>-1</sup>	[320]
LiCoO <sub>2</sub>	H <sub>2</sub> O <sub>2</sub>	H <sub>2</sub> SO <sub>4</sub>	94%	93%	100 g L <sup>-1</sup> , 2 M H <sub>2</sub> SO <sub>4</sub> , 5 vol.% H <sub>2</sub> O <sub>2</sub> , with a leaching time 30 min and a temperature 75 °C	[321]
LiCoO <sub>2</sub>	H <sub>2</sub> O <sub>2</sub>	H <sub>2</sub> SO <sub>4</sub>	95%	80%	65 °C; 1 h; 6 %; 33 g L <sup>-1</sup>	[322]
LiCoO <sub>2</sub> , LiCo <sub>1/3</sub> Ni <sub>1/3</sub> Mn <sub>1/3</sub> O <sub>2</sub> , and LiMn <sub>2</sub> O <sub>4</sub>	H <sub>2</sub> O <sub>2</sub>	Citric acid (C <sub>6</sub> H <sub>8</sub> O <sub>7</sub> )	99.1%	99.8%	90 °C; 1 h; 0.5 M; 20 g L <sup>-1</sup>	[323]
LiCo <sub>1/3</sub> Ni <sub>1/3</sub> Mn <sub>1/3</sub> O <sub>2</sub>	H <sub>2</sub> O <sub>2</sub>	Citric acid (H <sub>3</sub> Cit) and H <sub>2</sub> O <sub>2</sub>	99%	95%	Citric acid concentration of 2 mol L <sup>-1</sup> , at a temperature of 80 °C 90 min, liquid–solid ratio of 30 mL g <sup>-1</sup> , and 2 vol. % H <sub>2</sub> O	[324]
LiCoO <sub>2</sub>	-	5 wt.% NaOH solution, 4 M H <sub>2</sub> SO <sub>4</sub> + 10% v/v H <sub>2</sub> O <sub>2</sub>	96%	95%	85 °C for 120 min	[325]
LiCoO <sub>2</sub>	5% H <sub>2</sub> O <sub>2</sub>	Diluted sulfuric acid	93.4%	66.2%	368 K and 50 g/L pulp density for 240 min	[326]



Table 9. Cont.

Cathodes	Reductant	Leaching Agent	Leaching		Conditions	Ref.
			Li	Co		
LiNi <sub>1/3</sub> Co <sub>1/3</sub> Mn <sub>1/3</sub> O <sub>2</sub>	Ascorbic acid and flavonoids	Citrus fruit juice (CJ)	~100%	94%	90 °C; 0.5 h; 50 g L <sup>-1</sup>	[327]
LiNi <sub>1/3</sub> Co <sub>1/3</sub> Mn <sub>1/3</sub> O <sub>2</sub>	H <sub>2</sub> O <sub>2</sub>	Acetic acid and metallic acid	98.39%	97.72%	H <sub>2</sub> O <sub>2</sub>	[328]

The valuable transition metals from cathodes, such as Li, Co, Mn, and Ni, can be recovered using a hydrometallurgical process [329]; however, in Table 9, we only concentrate on the recovery of the key components, such as Li and Co. Leaching, contaminant removal, metal recovery, and Li recovery are the four categories under which the procedure can be divided. The leaching process is the primary step in the hydrometallurgical process. The collected cathode AM will be leached with a variety of leaching agents, including hydrogen chloride (HCl), nitric acid (HNO<sub>3</sub>), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), phosphoric acid (H<sub>3</sub>PO<sub>4</sub>), acetic acid, citric acid, and also citric fruit juice. In order to dissolve the metals in the traditional hydrometallurgical process, powerful inorganic acids must be used as leaching agents, which may not be practical for large-scale applications.

### 6.2.1. Leaching

Leaching is a technique used to dissolve certain metals found in end-of-life (EOL) LIBs. This is followed by additional processing of the ensuing leachate to remove the metal ions from the combination and produce the desired product [330].

### 6.2.2. Alkali Leaching

Due to the selective leaching and ability to skip expensive separation or purification processes, alkali leaching has drawn interest. Since it can create stable ammonia complexes with metals such as Ni, Co, and Cu, an ammonia-based arrangement is employed. Sulfites were used as the reducing agent and ammonia or ammonium sulfate was the leaching solution, in studies by Zheng and Chen. Both experiments found good overall leaching efficiency for Ni, Co, and Li. Mn had distinct leaching characteristics. It was discovered that the concentration of (NH<sub>4</sub>)<sub>2</sub>SO<sub>3</sub> had a significant impact on the leaching efficiency of Mn, with 0.75M being the ideal concentration [331].

### 6.2.3. Acid Leaching

Acid leaching is equally common since it consistently has a relatively high efficiency. To recover high purity precious metals from processed battery waste, hydrometallurgical techniques are usually used. The cathode metals in a spilled ion were separated using an acid solution, and precious metals are enhanced either through removal and purging or by resynthesizing the cathode directly. Since all metal types may be leached and recovered with high transparency, competence, and yield for diverse cathode compositions, it is durable. Although organic leaching gives similar leaching productivity with biodegradable qualities, significant inorganic leaching might result in secondary contamination (excess acid solution and harmful gas release). Citric acid, ascorbic acid, oxalic acid, and formic acid are examples of organic leaching agents. Acid leaching agents contain HCl, H<sub>2</sub>SO<sub>4</sub>, and HNO<sub>3</sub> [332].

## 6.3. Direct Recycling

Direct recycling is a technique that has been developed to preserve the chemical composition and process value of battery materials. In order to prevent the chemical break of the active elements, which are the major recovery “target”, battery components are separated, generally via physical separation techniques, magnetic departure, and

moderate heat processing. The pre-treatment steps for the separation operations are stabilization, disassembly, and separation [333]. The advantages of the direct recycling process include fewer emissions of greenhouse gases and  $\text{SO}_x$ , lower energy consumption, and fewer processing steps required to produce new cells [334]. The key benefits of this technology include its relative simplicity, the ability to directly reuse active materials after regeneration, and its much reduced emissions and secondary contaminants compared to previous approaches. On the other hand, the principal drawbacks are the need for precision active material chemistry-based sorting and pre-processing, as well as the difficulty in ensuring consistent high purity and perfect crystal structure. The procedure is also quite sensitive to changes in the input stream. This procedure is a flexible but untested method that has only been used in lab settings up to this point [335].

Prior to direct recycling, it is best to identify the most effective ways to separate cathode and anode from single or mixed batches, as well as to eliminate Al/Cu-foils. It is unknown whether identical formulations might be separated from one another, even though magnetic separation is covered by a patent. Moreover, it may not be cost-effective. The possibility of direct recovery of high-value cathode products would be made possible by techniques for isolating cathodes from one another or for utilizing a mixed product [336]. Instead of traditional recycling, direct recycling technology has been suggested as a possible substitute [337]. However, it must overcome a number of obstacles before being used commercially. A uniform direct recycling procedure has not yet been developed, since direct recycling requires customized processes with particular rejuvenation conditions for different battery materials [338]. Separation and the revitalization and upcycling of active materials are the two key components of the direct recycling process. In order to improve the electrochemical performance of recycled materials, further actions are frequently required for repair, rejuvenation, and upcycling [339]. A typical direct recycling process flowsheet is shown in Figure 12.

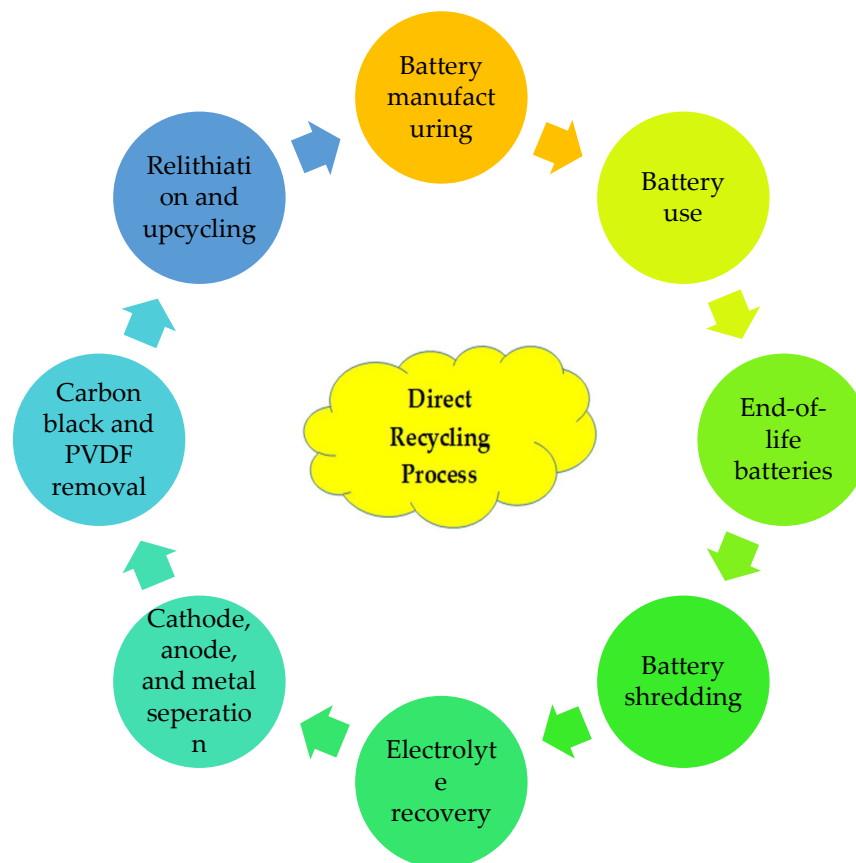
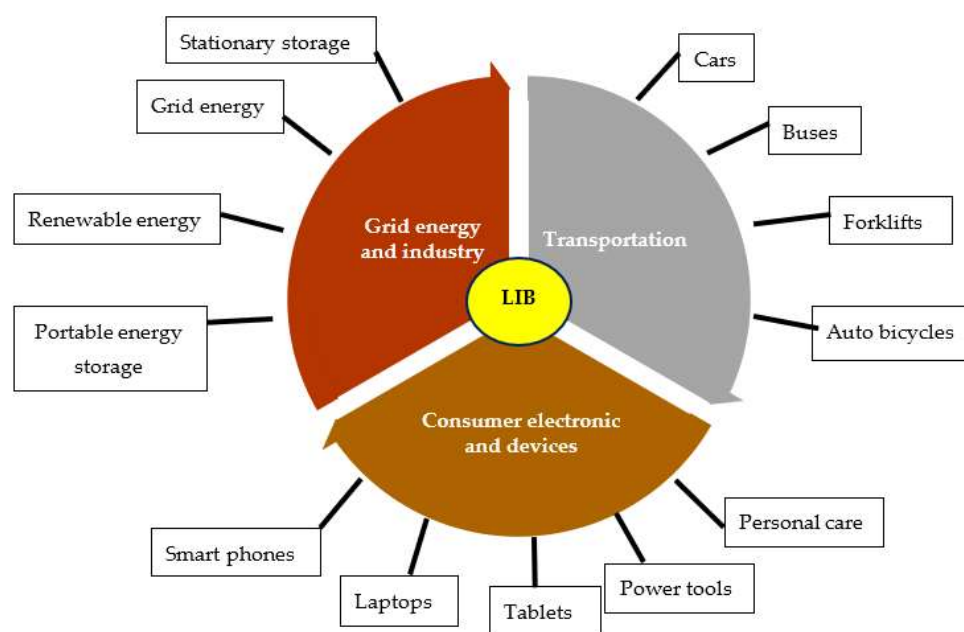


Figure 12. A conceptual illustration of the direct recycling process.

## 7. Applications of LIBs

The growing consumer market currently largely involves the use of LIBs for several forms of electronics, such as smartphones, smartwatches, computers, laptops, and power banks (Figure 13) [340]. Moreover, in the vehicle market, many electric automobiles including cars, trucks, scooters, and motorcycles also make use of LIBs [56]. After recognizing the significant benefits of LIBs, a large majority of countries are committing to the integration of electric vehicles (EVs) [341]. Meanwhile, in 2018, China's EVs have been reported to account for roughly 4.40% of all 28.1 million automobiles sold worldwide, which when compared to the USA and Europe, is much higher, with just over 2% and around 3%, respectively [342].

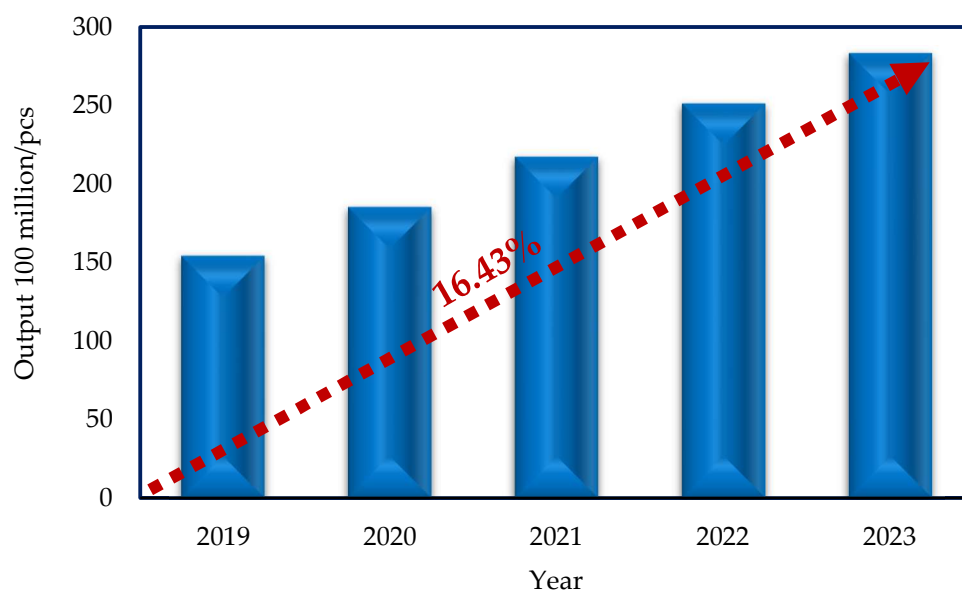


**Figure 13.** The applications of LIBs in different markets [343].

However, the rampant progress of electronic devices and electric vehicles poses a significant problem for the future, which is the rapid depletion of the Earth's natural resources, such as the precious metals that make up the electrodes and electrolytes of these LIBs [344]. Since the Earth has a finite amount of minerals, over time, the demand for lithium will climb higher and higher until the supply will become too insufficient or too expensive.

Nevertheless, the availability and a potential price spike of lithium remains continuously under constant debate. Experts have estimated that in the future, the rate of production will also soar, as illustrated in Figure 14 [345], and the demand for lithium may rise exponentially (or two to six times) above the current level of its mineral reserves [346].

With the least amount of toxic waste by-products, in an environmentally acceptable manner, and with little energy usage, the majority of battery components can be recycled by more than 90%. Manufactured precious metals and compounds should be highly pure in order to be used in new batteries. Injecting alternative Li, Co, and Ni compound sources back into the economy will give the limited resources another strategic route.



**Figure 14.** Chinese LIB production from 2019 to 2023 [345].

There are now various next-generation lithium-ion (Li-ion) batteries on the market that frequently have higher efficiency while having lower pricing. In comparison to conventional lithium-ion batteries, the newest generation of EVs and grid-scale storage batteries are more efficient, have a greater capacity, and last longer. Modern next-generation batteries have rapid charging and discharging speeds. Advanced battery types have emerged with a high capacity that lasts for more than 20 years. Lithium-ion batteries mixed with other metals or elements to create a new battery technology, such as Li-ion–cobalt oxide, are the most typical form of large-capacity, extensively utilized, and sophisticated batteries being produced today. Li-ion–cobalt oxide, lithium–iron phosphate, lithium-ion–silicon, lithium-ion–manganese oxide, and lithium-ion–sulfur is some of the advanced next-generation Li-ion battery technologies that are currently available, but they are less widely commercially available than Li-ion–cobalt varieties or even lithium–iron–phosphate batteries, which are currently a popular battery solution for some stationary battery applications. Lithium–iron–phosphate, which is often seen as being safer and more environmentally friendly than lithium ions, is what Tesla plans to use in its future next-generation batteries [347].

## 8. Conclusions

The rapid development and production of renewable energy electronics in this new era, such as communication devices and electric vehicles, are intended to combat environmental risks. However, the same application can result in devastating damage when disposed of and recycling practices are not followed. Li-ion batteries provide many benefits but also present a grave risk if they are inappropriately disposed of once they have extended to the end of their suitable lives. The development of Li-ion battery technology, the different widely used cathode and anode materials, and the benefits and drawbacks of each in relation to the most appropriate application were all thoroughly studied in this work. Furthermore, the electrochemical processes that underlie battery technologies were presented in detail and substantiated by current safety concerns regarding batteries. As a consequence of its high power and high energy density as compared to other types of batteries, lithium-ion batteries have become a trustworthy method of energy storage. To achieve high power and high energy density, cells can be constructed in a variety of ways, including cylinder, prismatic, and pouch designs. Additionally, thorough material research and design can increase the energy density of Li-ion batteries. There are several techniques available to enhance both modern and conventional electrolyte materials, such as chemical doping, innovation synthesis paths, and dense thin-film preparation. Furthermore, there are two technical difficulties, which are the development of highly scalable synthesis routes

and the tailoring of mechanical properties. However, further study is needed to create next-generation Li-ion batteries with significantly better cyclability, volumetric energy density, specific energy, and safety. This is especially true in light of the growing need for energy storage brought on by the more prevalent electric automobiles.

Since the introduction of the first generation of electric cars, the amount of used batteries that can be recycled has increased. To minimize negative effects on the environment and human health as well as the consumption of virgin raw materials, end-of-life battery management is crucial for electric vehicles (EVs). This review collected the reviewed recycling technologies and covered the three main LIB recycling technologies. Li, Co, and other metals, for instance, Ni and Mn, are the most valuable metals that can be recovered from old LIB cathodes since these metals are utilized to make the cathode materials for the LIBs. Pyrometallurgical and hydrometallurgical processes that have been developed are the most used cathode recovery techniques. Another low-cost method is direct recycling, which preserves the structure of the lithium-ion battery cathode. Together with reducing carbon emissions, the design process should also balance treatment costs. Thus, batteries can be reused for a further several years utilizing modern techniques, which is better for the environment. However, if they can be consistently scaled up, these techniques can become increasingly effective, and commercial battery recycling may be feasible.

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