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Invited Review

Reduction, reuse and recycle of spent Li-ion batteries for automobiles: A review

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Abstract: The demand for Li-ion batteries (LIBs) for vehicles is increasing. However, LIBs use valuable rare metals, such as Co and Li, as well as environmentally toxic reagents. LIBs are also necessary to utilize for a long period and to recycle useful materials. The reduction, reuse, and recycle (3R) of spent LIBs is an important consideration in constructing a circular economy. In this paper, a flowsheet of the 3R of LIBs is proposed and methods to reduce the utilization of valuable rare metals and the amount of spent LIBs by remanufacturing used parts and designing new batteries considering the concept of 3R are described. Next, several technological processes for the reuse and recycling of LIBs are introduced. These technologies include discharge, sorting, crushing, binder removal, physical separation, and pyrometallurgical and hydrometallurgical processing. Each process, as well as the related physical, chemical, and biological treatments, are discussed. Finally, the problem of developed technologies and future subjects for 3R of LIBs are described.

Keywords: Li-ion battery; reduction; reuse; recycle; processing; automobiles

1. Introduction

Efforts to decrease air pollution and save energy in the transportation industry have highlighted the need for vehicles with batteries bearing excellent characteristics. Li-ion batteries (LIBs) are high-quality batteries with a long lifetime. A forecast of the required production of various automobiles up to 2050 is shown in Fig. 1 [1]. Today, the production of passenger cars with internal combustion engines (ICEs) dominates the market, although the production of electric vehicles (EV), plug-in hybrid electric vehicles (plug-in hybrid vehicle) (PHEV (PHV)), and hybrid vehicles (HV) are expected to increase. The total production of passenger cars using batteries is estimated to exceed 0.1 billion units in 2050. Although Ni-metal hydride batteries are currently in use, LIB utilization is expected to increase in the near future.

LIBs are composed of four main parts, namely, the cathode material, the anode material, the separator, and the electrolyte. The shipping amounts as the value of these parts is shown in Fig. 2 [2]. Cathode materials are relatively expensive to ship, and costs related to this LIB component are estimated to equal approximately 15 billion USD in 2022. Several types of cathode materials have been developed over the years, as shown in Table 1. For example, $LiCoO_2$ (LCO) was developed in 1991, $LiMn_2O_4$ (LMO) in 1997, $LiNiCoMnO_2$ (NMC) in 2001, $LiFePO_4$ (LFP) in 2003, $Li_4Ti_5O_{12}$ (LTO) in 2007, and $LiNiCoAlO_2$ (NCA) in 2009, and LMO in 2018 [3]. LCO has a market share of 37wt%, and NMC has a market share of 29wt%. For the quality of cathode, the comparison of energy capacity, power, safety, performance, life span, and cost are described in Ref. [4]. The specific energies of LCO, NCA, and NMC are large, but their safety is low. By contrast, the specific energies of LFP and LTO are low, but their safety is high and life span are well established. Thus, each cathode presents unique advantages and disadvantages.

Rare metals used in cathode materials, such as Co, Li, Mn [5], and Ni, are in short supply because of the oligopoly of production. The top three countries produce 88wt% of Co, 79wt% of Li, and 56wt% of Ni, and the production situation of these metals was reported by Investing News in 2019 [6–8]. In 2019, approximately 60% of the Co available worldwide was produced in Congo, and the price of this metal fluctuated in the short term. The top producers of Ni in

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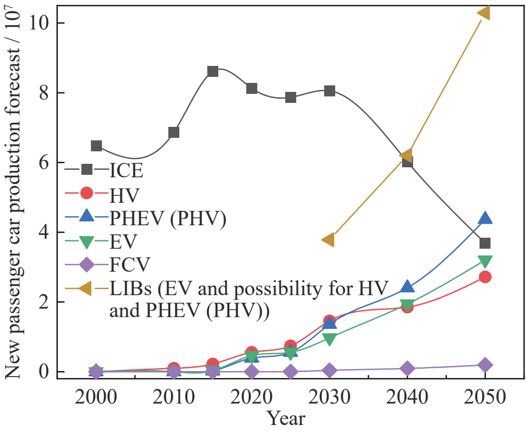


Fig. 1. Production forecast of various types of automobiles [1]. ICE—Internal-combustion engine; HV—Hybrid vehicle; PHEV (PHV)—Plug in hybrid electric vehicle (Plug-in hybrid vehicle); EV—Electric vehicle; FCV—Fuel cell vehicle.

2019 were Indonesia and the Philippines. The top three producers of Li in 2019 were Australia, Chile, and China in this order; 40wt% of the Li obtained was utilized to produce batteries, and 30% was utilized in the ceramics and glass industries [9]. A forecast of future demands for Ni, Co, Mn, and Li in 2020 and 2030 is provided in Table 2 [10−11]. The demand for Li in 2030 is forecasted to be eight times greater than that in 2020; in 2018, Li costs USD 16500 per ton. The demand for Co in 2030 is forecasted to be six times that in 2020; in 2018, Co costs USD 80000 per ton. The volumes of Ni and Mn required in 2030 are expected to increase by twofold compared with the corresponding demands in 2020. Considering the supply risk due to their limited production and the rapid increase in demand for these metals, the development of approaches to reduce the utilization of rare metals in cathode materials is necessary. Unfortunately, while the recent decrease in Co utilization in cathode materials mitigates the supply risk for this metal, the benefits of recycling Co also diminish.

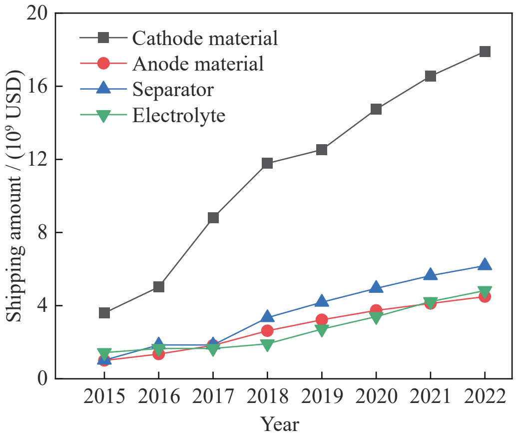


Fig. 2. Shipping amounts of the four main components of LIBs worldwide [2].

Researchers have well studied the reuse and recycling of LIBs because of environmental and resource considerations. The total number of published papers on this topic, including conference abstracts, over the last 20 years by using Web of Science and reference [12] is shown in Fig. 3. The total number of articles discussing the recycling of LIBs exceeds 1100, and the number of LIBS recycling vehicles is over 200 and about 20% in the publication related to LIBs recycling. Many papers have been published by Chinese researchers. The Swedish Energy Agency published a report on the reuse and recycling of LIBs after reviewing approximately 1000 articles [13]. The recovery of rare and precious metals from waste mobile phones has also been studied [14]. This article describes only the most important reports and reviews on LIB reuse and recycling on account of the sheer number of papers published on this matter.

Table 1. Development progress and estimated market share of various cathode materials [3]

Chemical formula	Cathode types	Li / wt%	Voltage / V	Cycle	Developed year	Estimated market share in 2018/%
$LiCoO_2$	LCO	7.09	3.8	500−1000	1991	37.2
$LiMn_2O_4$	LMO	3.84	3.3	300−700	1997	21.4
$LiNiCoMnO_2$	NCM (111, 523, 622, 811)	~7.20	3.7	1000−2000	2001	29.0
$LiFePO_4$	LFP	4.40			2003	5.2
$LiNiO_2$	LNO	7.11	3.8	300−700		7.2
$Li_4Ti_5O_{12}$	LTO (SCiB)				2007	
$LiNiCoAlO_2$	NCA	~7.20	3.6	500	2009	

Table 2. Comparison of future demands for Ni, Co, Mn, and Li in 2020 and 2030 [10−11]

Metal	Supply and demand 2030/2020 (times)	Supply and demand to product in 2020 / (10^6 t)	Price in 2018 / (USD $\cdot t^{-1}$)	Reference
Ni	2	2.20	8932	Wood Mackenzie
Co	6	0.13	80000	CDI
Mn	2	19.00	2060	BHP Billiton
Li	8	0.045	16500	TRU

Note: CDI—Community for data integration; TRU—TRU Group Inc.

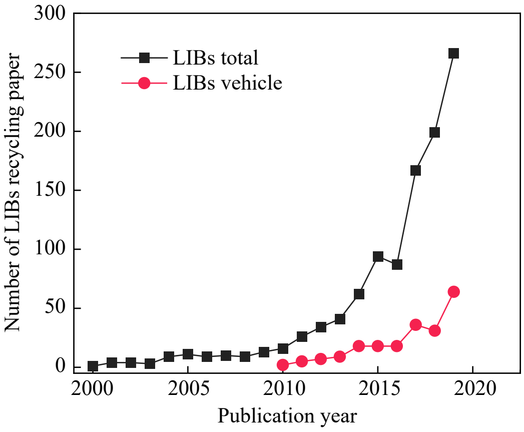


Fig. 3. Number of papers related to LIBs recycling published over the last 20 years (from Web of Science).

2. Composition of lithium-ion batteries and their recycling process

The cathode materials of LIBs mainly include Al foil coated with active materials containing Li (Table 1), conducting agents, organic solvents, and binders, such as polyvinylidene fluoride (PVDF). Anode materials usually include Cu foil with a surface coating, graphite, active substances, and organic binders. The separator is usually made of plastic, such as PP polypropylene (PP), polycarbonate (PC), and polyethylene (PE), with a membrane structure. Electrolytes are often made of Li salts, including $LiPF_6$, $LiClO_4$, $LiCF_3SO_3$, $Li(CF_3SO_4)_2N$, $LiBF_4$, $LiAsF_6$, PVDF, and *N*-methyl-2-pyrrolidone (NMP), in a solvent, such as dimethyl carbonate (DMC), diethyl carbonate, vinyl carbonate, methyl ethyl carbonate, and γ-butyrolactone (BL). The shell and cover for LIBs are mainly made of Al alloy and steel.

The characteristics of three types of LIBs for electric vehicles are listed in Table 3 [15−16]. Each battery pack weighs 200–500 kg and has a voltage of approximately 350 V. A pack is composed of 10–50 modules, and each module contains several to several hundreds of cells of 3.6–3.8 V. The chemical composition of several LIB cells is listed in Table 4 [17]. LCO and NMC are composed of 17.3wt% and 8.5wt% Co. However, considering the metal prices in Table 2, LFP and LMO are of little value. The Li content of LIBs is 1.2wt%–2.0wt%.

Table 3. Characteristics of three types of LIBs for electric vehicles [15−16]

Component		LIBs shape		
		Cylindrical	Prismatic	
			Hard case	Pouch
Pack		375 V, 530 kg	355 V, 235 kg	360 V, 294 kg
Module		25 kg, 16 modules per pack	25 kg, 8 modules per pack	3.8 kg, 48 modules per pack
Cell		444 cells per module	12 cells per module	4 cells per module
Composition of case		Steel (Fe−Ni alloy etc.)	Aluminum	Aluminum
Example	Height / mm	 70	 120	 260
	Width (diameter) / mm	20	170	230
	Thickness / mm		50	10

Table 4. Chemical composition of several LIBs [17] wt%

Material used in LIBs	LIBs			
	LCO	LFP	LMO	NCM
Al	5.2	6.5	21.7	22.7
Co	17.3	0	0	8.5
Cu	7.3	8.2	13.5	16.6
Fe/steel	16.5	43.2	0.1	8.8
Li	2.0	1.2	1.4	1.3
Mn	0	0	10.7	5.9
Ni	1.2	0	0	14.8
Binder	2.4	0.9	3.7	1.4
Electrolyte	1.4	14.9	11.8	11.7
Plastic	4.8	4.4	4.5	3.3
Total	70.1	79.3	67.4	95.0

Two companies recycled over 1000 t of LIBs in 2018 [17]. The Chinese company Green Eco-manufacture Hi-Tec Co. and the European company ERAMET processed 20000 t of LIBs in 2018. Other commercial recycling companies for LIBs are listed in Table 5 [17−18]. Most companies adopt three types of technological treatment, including the hydrometallurgical method, the pyrometallurgical method, and a combination of these methods. Green Eco-manufacture Hi-Tec Co., IPGNA, Sumitomo–Sony, Toxoco, Eurodieuze, and Zimaval utilize the pyrometallurgical method; Dowa, Batrec AG, Nippon Recycling Center, and INMETOCO use the hydrometallurgical method; and Umicore, Accurec GmbH, and Glencore PLC utilize both the hydrometallurgical and pyrometallurgical methods.

Life cycle assessment is used to estimate the environmental impact of recycling and reduction [19]. Circular economies must implement the reduction, reuse, and recycle (3R) strategy [20]. Several treatment processes to achieve the 3R of LIBs have been reported [15−17,21−22]. The flowsheet of a LIB treatment process considering 3R is shown in Fig. 4; here, each technology is assigned numbers and discussed in detail in Section 3.

Table 5. Commercial recycling companies for LIBs [17−18]

Process	Company	Raw material	Outcome
Pyrometallurgy	IPGNA Ent. (France)	All batteries	$LiCoO_2$
	Sumitomo-Sony (Japan)	LIBs	$Co(OH)_2$
	Toxco (Canada)	Li, Ni based battery	$LiCoO_2$
	Eurodieuze (France)	All batteries	Ni, Cd, Steel
	Zimaval (France)	Zn, Mn, Hg based Battery	Zn, Mn
Hydrometallurgy	Dowa (Japan)	All batteries	Co, Ni
	Batrec AG (Switzerland)	Li, Hg based battery	
	JX Nippon Mining & Metals (Japan)	LIBs	Li, Co, Ni, Mn
	INMETCO (USA)	Ni−Cd	Ni, Cd, Zn
Pyrometallurgy + hydrometallurgy	UMICORE (Belgium)	LIBs, Ni−MH	Co, Ni
	Accurec GmbH (Germany)	All batteries	Ni, Cd, Fe, Li_2CO_3
	Glencore PLC (Switzerland)	LIBs, EV	Cu, Ni, Zn, Li

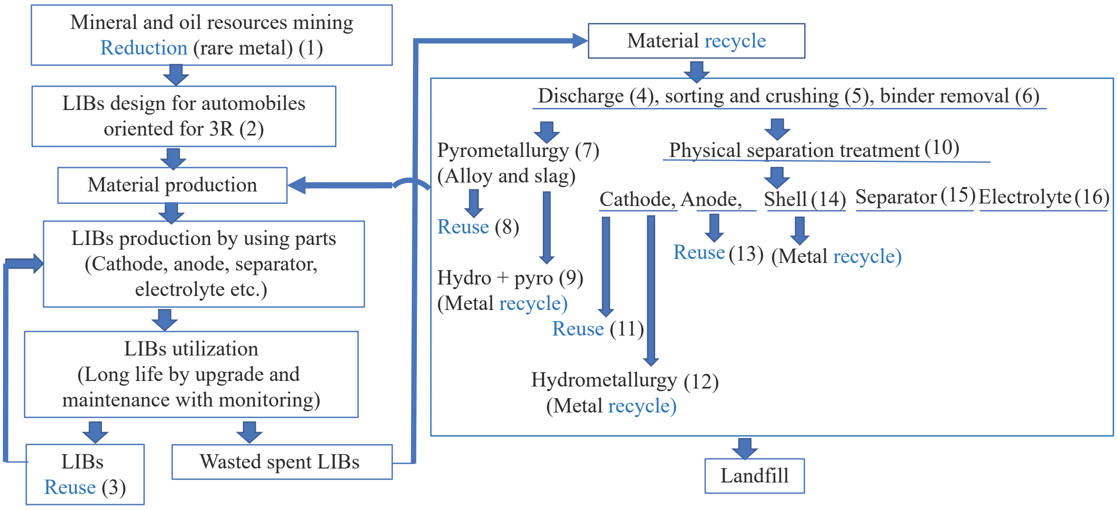


Fig. 4. Flowsheet of a LIB treatment process considering the concept of 3R.

3. Reduction, reuse and recycle technologies for LIBs

There is generally an order to construct the 3R of LIBs. To considering the ranking of 3R, the first priority is the reduction, the second is reuse, and the third is recycle. In this section, the technological contents are mainly described in the order from (1) to (16) in Fig. 4.

3.1. Reduction (1)

Reduction [23] refers to a decrease in the utilization of valuable rare metals and the decrease of landfill amount by remanufacturing process of used parts including 3R model [24] and the small size and lightweight LIBs production [25]. Reduction of the use of rare mineral resources is necessary to meet future demands (Table 2). The Co necessary to produce LIBs could be substituted with other elements to minimize the supply risk and enhance the quality of LIBs. The composition of cathode materials has seen great improvements over the last 20 years. Reuse and recycling methods that could accommodate changes in the cathode must be considered. However, while reduction of the use of rare metals in LIBs may help meet future demands for mineral resources, economical technologies, and systems for LIB recovery must also be considered.

3.2. Design (2)

The inverse manufacturing design of LIBs is necessary to achieve easy assembly and disassembly, reuse parts, and recycle spent materials. There are some ideas and methods for the design of LIBs by considering reuse and recycle: An automated disassembly system of used LIBs enables by using toolsets at material handling. Cathode sheets, anode sheets, separators, and Al film housing can be automatically separated [26]. A microscale architecture at the interface between the collector and composite film has been designed. During recycling, the composite film can be peeled off from the collector in a certain direction [27]. The recycling methods related to the automatic dismantling of EV battery packs into modules and cells have been studied included automatic and intelligent recovery systems [17].

Circular-economy companies recycling LIBs have worked closely with battery manufacturers to adopt designs that would enable the easy dismantling of a product and have been developed for the green recovery of all valuable battery components. Companies considering a circular economy for LIB recycling would be possible to easily dismantle the product and uptake new green chemistry processes using H_2O_2 [21]. Cooperation between design teams and recycling engineers is necessary to establish extended producer responsibility.

3.3. Reuse (3)

Battery manufacturers and vehicle production companies using these batteries must consider ways to reuse LIBs. Energy storage in LIBs is essential to produce clean and renewable electricity [21]. Commercially available LIBs must be reused many times. The reuse means a long-life utilization for material and energy saving. The upgrade and maintenance by monitoring the use of LIBs equal to a long-life utilization. Spent LIBs can also be used in smart grid systems. An energy management system is utilized to achieve smart-grid energy balance in electric vehicles featuring scrapped LIBs [28]. The charge percentage is an important consideration in a battery's lifetime. For example, the number of cycles from a typical Li-ion battery (NCM) is 6000 to 15000 discharge cycles in 10% depth of discharge, while 300 to 600 discharge cycles in 100% of the depth of discharge. Some manufacturers have added software controls to their battery management systems to help maintain battery life. An efficient electrode design is needed to reduce heat and improve battery life and capacity [4]. LIBs may be reused as energy storage systems in smart grids.

3.4. Discharge of used LIBs for recycling treatment (4)

In the pyrometallurgical process, high-temperature incineration can automatically discharge LIBs. Heating and thermal runaway produce HF and other hazardous gases. LIB discharge may also be carried out by soaking LIBs in 5wt% NaCl aqueous solution before pyrolysis [29].

In the hydrometallurgical process, discharging is useful to prevent short-circuiting and self-ignition. LIBs can be discharged in salt aqueous solutions, such as 0.8 mol/L NaCl, 0.8 mol/L $FeSO_4$, or 0.4 mol/L $MnSO_4$, to decrease their voltage. In two large-scale discharge models, the quick model reduced the residual voltage to 1.0 V from approximately 3.6 V within 2 h and the full model reduced the residual voltage to 0.5 V within 3 h [30]. LIBs may be soaked in Na_2SO_4 and other saturated salt solutions to discharge. For example, the electrodes of spent LIBs are soaked in brine water and discharged via the following reactions:

$LiPF_6 + H_2O \rightarrow LiF + POF_3 + 2HF\uparrow$;

$LiFePO_4 + 2HF \rightarrow LiH_2PO_4 + FeF_2$;

$(CH_2OCO_2Li)_2 \rightarrow Li_2CO_3 + C_2H_4 + CO_2 + 0.5O_2$.

Supercritical and subcritical conditions can reduce gas generation [31]. If LIBs are discharged, the subsequent treatment and reuse become easy. An economical rapid discharge technology for LIBs must be developed.

3.5. Crushing (5)

Several crushing methods may be employed to separate the materials of LIBs. A hammermill is widely used in various crushing atmospheres to crush LIBs; however, this method may produce toxic gases, such as HF, PF_5, POF, and –CH–CF. Crushing may be carried out in a closed space, such as a glove box, in an inert atmosphere, such as CO_2 or Ar [30], and in water. In the Batrec process, mechanical processing is performed in a CO_2 atmosphere within a tightly closed chamber [32]. Cryogenic crushing with liquid nitrogen at approximately −200°C [33] or liquid CO_2 at −56 to −20°C is carried out to prevent HF gas generation [31]. Cutter mills are useful to separate plastics, and abrasion mills can effectively remove fine particles from metal foil. Ball mills are popular for grinding. The planetary ball mill, for example, is used extensively for mechanochemical grinding with polyvinyl chloride and co-grinded $LiCoO_2$ with ethylenediaminetetraacetic acid. Also, the crushing underwater explosion has prevented producing toxic HF gas with calcium salts [34]. Fire and toxic gas emissions must be prevented during the crushing process.

3.6. Binder removal (6)

Infrared spectrometry has revealed that the cathode and anode binders are composed of PVDF and carboxymethylcellulose, respectively [35]. Cathode powders containing Li transition metal oxides are fixed on Al foil, while anode powders, such as graphite, are coated on Cu foil. Removal of the binder is important to recycle the particles on the foils. Various binder removal methods are listed in Table 6. Thermal treatment at the optimum temperature [29,36], microwave roasting [37], solvent utilization [35,38], the mechanochemical process, the supercritical CO_2 method [18], and the high-voltage pulse method [39] have been proposed to remove binders from foils. Control of the atmosphere during the thermal treatment of LIBs is important because thermal treatment with 20vol% oxygen produces toxic gas, as determined by thermogravimetry [35]. This gas mainly contains light alkenes and pyrolytic tar containing aromatic long-chain alkenes and light alcohols, and the PVDF binder is decomposed to H_2O, CO_2, and fluoride-containing gases with a maximum emission peak at 522°C. Thermal treatment is performed in a vacuum or inert atmosphere, such as N_2 gas at 550°C for 2 h to prevent toxic gas emissions [29]. In the utilization of solvent to remove the binder from powders, aqueous exfoliating and extracting solution (AEES) methods were reported [35]. Chemical reagents, such as NMP, BL, dimethylformamide, and dimethyl sulfoxide, are also employed, although they are toxic solvents [36]. Citrus fruit juice can remove the binder and leach the metal with removal efficiencies of 94%–100%; this method is considered extremely environment friendly. The combination of thermal and solvent methods may be necessary to remove the binder from cathode and anode materials in an economical manner.

Table 6. Various binder removal methods

Method	Condition
Thermal treatment	·Air atmosphere at 150–500°C for 1 h to burn binder with emission of harmful gas ·Nitrogen atmosphere at 550°C for 3 h to evaporate or decompose binder ·Vacuum atmosphere at 600°C for 0.5 h to evaporate or decompose binder
Microwave roasting	Cathode particles with carbon mixing by ball milling and then microwave treatment
Solvent utilization	·*N*-methyl-2-pyrrolidone (NMP) at 100°C to dissolve PVDF ·*N*,*N*-dimethylfolmamide (DMF), dimethylacetamide (DMAC), and dimethyl sulfoxide (DMSO) to dissolve binder (PVDF and PTFE) ·NMP + NaOH aqueous solution and thermal treatment combination ·Citrus fruit juice to dissolve PVDF
Mechanochemical method	Mechanical abrasion and Fenton reaction assistance
High voltage pulse	Separation of PVDF from cathode particle ($LiCoO_2$) in aqueous phase

3.7. Pyrometallurgy (7)

The pyrometallurgical process of feeding LIBs into the furnace does not emit toxic gas because the exhaust gas is treated appropriately. The pyrometallurgical process produces alloys from cathode materials and the slag. The furnace, pyrolysis residue, disposal devices for pyrolytic tar, and gas are described, and the chemical and structural formula of gas generated in each process is explained [29].

Flowsheets of Umicore's process for the production of Ni, Co, Cu, and Fe alloy, Inmetco's process, and Glencore process for the production of Fe-based alloys are described [31]. A Li and Al concentrate into slag and organic materials and carbons are used as an energy source and reducing agent. Li_2CO_3 and MnO are produced at 800°C for 45 min under vacuum conditions, and $LiCoO_2$ is produced at 850–950°C for 12 h. After roasting at 1000°C for 30 min, Li_2CO_3 particles are separated from graphite particles by wet magnetic separation.

In Accurec's process, the cells are heated to 250°C after disassembly, and the electrolyte is evaporated and collected in a condenser. Next, the active electrode materials are subjected to vacuum pyrolysis to recover Li. Li is recovered as a pure metal by direct evaporation and distillation, and Li oxide may be recovered by selective gas evaporation [31].

A molten salt-based electrolytic method to recover Li and Co from spent $LiCoO_2$ (LCO)-based LIBs have been developed [40]. After battery discharge and binder removal by heating in a vacuum at 450°C and calcination at 800°C, electrolysis is performed in a system with an LCO pellet cathode and graphite rod anode in a molten salt of $NaCO_3$ and K_2CO_3 at 750°C. LCO could be converted to CoO or CO and Li_2O using this process with average Li and Co recovery rates of 85% and 99%, respectively. The regeneration of LCO via the solid-state reaction is described in the next section. The pyrometallurgical process has the advantages of easy operation and no requirement of passivation; however, the process also features high energy consumption, hazardous gaseous emissions, and high capital cost [41]. While this process is useful for the recycling of large amounts of Co from LIBs, the merits of the pyrometallurgical process are low for LMO and LFP, for example, which do not contain valuable Co. In the metal alloy and slag production by pyrometallurgy, a large amount of similar composition of LIBs treatment as feed material is convenient.

3.8. Reuse via the pyrometallurgical method (8)

If the produced alloy contains Ni and Co, it can be reused as a hydrogen-absorbing alloy [37]. Honda Motor Co., Ltd. in Japan collected used LIBs, and Matsuda Sangyo Co., Ltd. in Japan studied the discharge of LIBs and recovery of cathode materials by physical separation. Japan Metals and Chemicals Co., Ltd. in Japan produced a hydrogen-absorbing alloy with Ni and Co to separate alumina slag from cathode particles via a pyrometallurgical method [42]

Some reports have described the process of cathode regeneration from LIBs. The relevant regeneration methods include solid-state reactions, the hydrothermal process, the sol−gel method, co-precipitation, and carbon-thermal reduction. Each method presents advantages and disadvantages [43]. In this section, methods using the thermal process as a pretreatment are described. The hydrothermal process, the sol−gel method, and the coprecipitation method are introduced in Sections 3.11 and 3.12.

The use of the solid-state reaction to regenerate LCO has been studied, as described in the previous section [40]. The regeneration process for spent LFP powders via the carbon-thermal reduction method could be achieved by planetary ball milling of a mixture of the carbon source and treated powder mixture followed by calcination at 650°C for 10 h in a H_2/Ar atmosphere [44].

The direct recovery method of Li from the cathode of LIBs mixed 18wt% carbon powder could be achieved by microwave reduction roasting. The microwave can increase the temperature of cathode material to 900°C by using 1500 W [37].

For the reuse of anode, the wasted graphite of anode is possible to reuse at 1300°C calcination by considering intercalation [45]. In the pyrometallurgical process, metal recovery from the alloy and the production of different alloys are possible. The regeneration of cathode and anode particles by thermal treatment is a feasible approach when considering an economical treatment method.

3.9. Recycling of slag and dust after pyrometallurgical treatment (9)

Chlorination roasting to recover Li from the slag after pyro-treatment of spent LIBs has been studied. The prepared slag is mainly composed of $CaSiO_3$ and $LiAl(SiO_3)_2$. A $CaCl_2$ among NaCl, $AlCl_3$, and $CaCl_2$ is the suitable chlorination reagent. When $CaCl_2$ and slag are mixed at a Cl:Li molar ratio of 1.8:1 and calcined at 1000°C for 90 min, 97wt% Li evaporation as LiCl may be achieved [46].

Pyrometallurgical treatment of LIBs is carried out in an electric arc furnace, and Li_2O can be accumulated in the flue dust. Li oxide is easily dissolved and recovered by the hydrometallurgical process [47]. However, a technology to reduce the cost of Li recovery cost by minimizing the energy consumed during the thermal treatment of slag must be developed.

3.10. Dry and wet physical separation (10)

The crushed parts are separated by heat treatment and physical separation. Various physical separation methods of liberated particles are listed in Table 7. First, the crushed LIB particles are classified by sieving. Binder removal then facilitates the separation of cathode particles on the Al foil and graphite particles as an anode on the copper foil.

Table 7. Physical separation methods of liberated particles

Methods	Condition
Sieving	Al-enriched fraction (>2 mm), Cu and Al-enriched fraction (0.25–2 mm), and Co and graphite-enriched fraction (<0.25 mm)
Pneumatic separation	Separation of Cu particles from carbon particles
Flotation	Separation as float of graphite particles from cathode particles
Magnetic separation	After heat treatment, cathode particles are magnetically captured
Eddy current separation	Al foil and Cu foil separation in the range of 2 to 20 mm
Electrostatic separation	Al and Cu capture from other nonconductive particles

Al, Cu, and plastics, such as PE and PP, could be recovered by an eddy current separator [48]. Foils such as Cu could be separated from carbon powder by pneumatic separation. Cathode particles, including Fe, Co, and Ni, could be separated from nonmagnetic materials by magnetic separation. After drying, conductive particles could be separated from nonconductive ones by electrostatic separation.

Froth flotation may be used to separate fine cathode particles from a mixture of cathode- and anode-like graphite particles [49]. In this process, kerosene is used as a collector and methyl isobutyl carbinol is used as a frother. The resulting graphite powder can then be floated and separated from nonfloated cathode powders. Wastewater treatment and water recycling are necessary for wet processes. Physical separation is necessary before hydrometallurgical treatment. Several mineral processing techniques can be combined with thermal pretreatment.

3.11. Reuse of the cathode powder (11)

The cathode powder obtained after physical separation can be reused following chemical treatment. Here, some examples of the reuse of cathode powders are described. The quality of recycled precursor particle in four differing cathode compositions of mainly NMC and another cathode mixture have been investigated. The cathode material of LIBs could be crushed, sieved, and then leached to remove impurities. The powders are then coprecipitated and calcined. NMC111 made of recovered precursors show higher rate capability than a commercial NMC111 prepared from virgin materials [50].

Repaired LFPs could be reused as a novel anode material for alkaline Ni/Fe batteries. The repaired LFP showed high specific capacity, suitable performance, and good cycling life in alkaline Ni/Fe batteries. A unique alkaline $Ni(OH)_2$/$LiFePO_4$ battery system was successfully developed by adopting recovered LFP materials and $Ni(OH)_2$ cathodes [51]. $LiFe_{0.6}Mn_{0.4}PO_4$ could be prepared from the whole LMO cathode of spent LIBs by dissolution with acid, the addition of the necessary reagents, and calcination under an Ar atmosphere [52].

Transition metal nanowire anodes have been prepared from spent LIBs. The relevant process relies on electrodeposition from solution after leaching of the batteries' electronic powder. The quality of the prepared nanowires was virtually similar to that of nanowire anodes produced via the same electrodeposition method with synthetic Co solution

[53]. On the other hand, the used cathode powder can be reused in the battery as follows. It was provided a route of sphagnum uptake of spent LIBs cathode toward high-performance Li−sulfur (Li–S) battery [54]. Reconditioning of LCO could be achieved by aqueous pulsed discharge plasma after the removal of the PVDF binder from LCO particles [39]. LMO obtained from spent LIBs could be directly reused as a cathode material for Na-ion batteries [55]. Several types of cathode powders could be regenerated as cathode powders and used as anode nanowires. Regeneration and reuse are also described in Session 3.8 and 3.12.

3.12. Metal recovery from the cathode by hydrometallurgical processes (12)

Many reagents can dissolve cathode powders. The leaching reagents used for various cathode materials are listed in Table 8. These leaching reagents include inorganic acids (HCl, HNO_3, H_2SO_4, H_3PO_4), organic acids with H_2O_2, and ammonium sulfate with ammonia and sodium sulfite. Bacterial leaching with *Acidithiobacillus thiooxidans* has been investigated. Inorganic acids leach all types of metals; however, organic acids can leach metals with high selectivity. The advantages of leaching spent LIBs include high selectivity, efficiency, solid/liquid ratio, and industrial viability [56]. Bioleaching has been investigated to recover metals from LIBs. *Aspergillus niger*, for example, produces different types of organic acids and gluconic acid. Approximately 100% Li, 94% Cu, 72% Mn, 62% Al, 45% Ni, and 38% Co could be successfully leached from spent LIBs by using this bacterium [57].

After leaching, extraction and precipitation are utilized. The separation methods of leached metal ions are listed in Table 9 [56,58]. Various metals could be separated by solvent extraction, chemical precipitation, electrochemical deposition, and the salting-out method. Li is precipitated as Li_2CO_3 and Li_3PO_4.

After the filtration of undissolved metal oxides and graphite, Li salts (e.g., LiCl, Li_2SO_4, and Li_2CO_3) in the filtrate are precipitated by exceeding the solubility concentration and Li_2CO_3 is finally recovered. The leaching conditions of spent LIBs have been investigated using electrochemical methods [59].

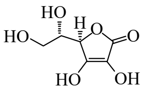
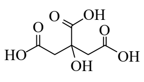
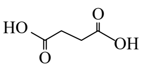
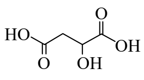
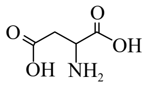
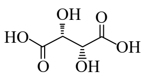
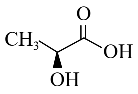
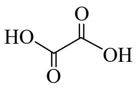
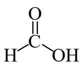
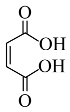
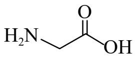
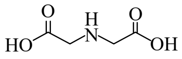
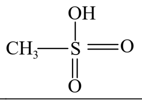
A process utilizing cation exchange membranes to separate and recover Li, Ni, Co, and Mn for the hydrometallurgical recycling of LIBs cathodes has been proposed. The cation exchange membrane separates a feed solution containing one group of electrolytes and a stripping solution containing another group of electrolytes [60]. The regeneration processes that do not need thermal pretreatment are the hydrothermal process, the sol−gel process, and the co-precipitation method, as described in Sections 3.8 and 3.11.

LCO is regenerated by the hydrothermal process and thermal treatment [61]. After leaching of mixed cathode materials (e.g., LCO, NMC, and LMO) by citric acid and H_2O_2, the resulting leachate is used directly in a sol−gel process to resynthesize NMC [62]. In the co-precipitation process, NMC is prepared from the leachate of spent LIBs via co-extraction followed by precipitation and sintering [63].

A natural biopolymer, κ-carrageenan, could form a composite membrane for the selective separation of Li^+ from Co^{2+} and increase L^{i+} ionic conductivity [64]. Li is recovered by solvent extraction, ion-sieve adsorption, electrochemical approaches, and membrane technology [9,65].

The hybrid system of citric acid and *Lysinibacillus* sp.

Table 8. Leaching reagents of cathodic materials

Category	Reagent					
Inorganic	Acid	HCl, HNO_3, H_2SO_4, H_3PO_4, sometimes with H_2O_2 ($Co^{3+}\rightarrow Co^{2+}$, $Mn^{4+}\rightarrow Mn^{2+}$) $Na_2S_2O_2$ and Na_2CO_3, CH_3COOH and H_2O_2				
	Alkaline	$(NH_4)_2SO_4$ (ammonium sulphate) with NH_3(ammonia) and Na_2SO_4 (sodium sulfite)				
Organic*	Ascorbic acid 	Citric acid 	Succinic acid 	Malic acid 	Aspartic acid 	Tartaric acid 
	Lactic acid 	Oxalic acid 	Formic acid 	Maleic acid 	Glycine 	Iminodiacetic acid 
	Methanesulfonic acid (CH_3SO_3H) 					
Biological	Acid thiobacillus ferroxidase and acid thiobacillus trioxidanes with H_2SO_4, Aspergillus Niger, Lysin bacillus–citric acid hybrid combination, etc.					

Note: *H_2O_2 is used as mixture of organic reagent.

Table 9. Separation methods used to recover metal ions from the leaching solution [22,56,58]

Methods	Condition	Metal to recover
Solvent extraction	Cyanex 272, (Cyanex 301)	Co, Ni
	D2EHPA	Mn
	PC-88A and Cyanex 272	Co, Ni, Mn
	Saponified P507	Co
	LIX841	Ni
	Cyanex 272 with TOA or TBP, (Alamine 336)	Co, Ni
Chemical precipitation	Addition of alkaline aqueous solution [$Co(OH)_2$]	Co, Ni
	Sulfidation with Na_2S [CoS]	Co
	Addition of saturated Na_2CO_3 solution or CO_2 purging [Li_2CO_3]	Li, Co, Ni, Mn
	Addition of H_3PO_4 or Na_3PO_4 to precipitate Li [Li_3PO_4]	Li
	Addition of $H_2C_2O_4$ ion to precipitate [CoC_2O_4]	Co, Mn
	Addition of NaClO [MnO_2]	Mn
	Addition of dimethylglyoxime [$Ni(C_4H_7N_2O_2)_2$]	Ni
	Addition of phytic acid [M_x phytate]	Fe, Co, Ni, Mn
Electrochemical deposition	Cobalt(III) is reduced to cobalt(II), highest purity of $Co(OH)_2$	Co
	γ-MnO_2 (cathode) and Pt (anode), $FePO_4$ (cathode) and Ag (anode), $Li_{1-x}Mn_2O_4$ (LMO) (cathode) and polypyrene (anode)	
Salting out	Addition of saturated ammonia sulfate solution and absolute ethyl alcohol into the leachate of $LiCoO_2$ cathode, and then salting out of Co^{2+} in the leachate	Co
Membrane	Utilization of cation exchange membranes	Li, Co, Ni, Mn
Biosorption	Biopolymer K-Carrageenan	Li, Co, Ni, Mn
	Chemical-biological hybrid system	

could enhance hydroxyl sites for Li leaching and improve C–OH and CO–O sites for Co biosorption. This hybrid treatment promotes Li leaching and Co biosorption [66]. Though there are advantages and disadvantages between pyrometallurgy and hydrometallurgy methods, the merits of hydrometallurgy of high material recovery and purity, low waste emissions, high prevalence of chemical reuse are described [67].

A hydrometallurgical process with phytic acid could be used for metal precipitation. The product recovered from spent LMO batteries is used as a solid polymer electrolyte for high-performance Li metal batteries [68]. The hydrometallurgical process shows the merits of high recovery efficiency of LIBs, mild reaction conditions, moderate energy consumption, and absence of gaseous emissions; however, the process requires wastewater treatment and complex procedures, including pretreatment [41]. Several types of leaching reagents, including inorganic, organic, and biological reagents, may be used, and the leached metals are collected by various methods. Cathode regeneration is possible as an application, however, the economical estimation is necessary for the hydrometallurgical process.

3.13. Reuse of the anode graphite (13)

The graphite powder of the separated anode could be reused as a toner in low-grade reuse. The spent LIB anodes to graphene and graphene oxide are proposed here. Shear mixing enhances graphene productivity by fourfold. High-quality graphene and its derivatives have been produced in this manner [69–70].

The ultrasonic leaching method can be used to recover anode particles. Graphene can be prepared with vitamin C as a reducing agent [63]. Graphite anode materials could be obtained from spent LIBs by a hydrometallurgical process without separation. High-quality graphite powder has been obtained by leaching and the removal of all impurities [71]. The graphite particle in the anode can be reused after separation and particle treatment.

3.14. Recycling of Al and Cu (14)

The Al flame of LIBs and Al and Cu foils in the electrode can be separated by an eddy current separator and recovered as recycled materials. Approximately 17% of the Cu and Al in foils in LIBs could be recovered by eddy current separation. The simulation of particle movement in the separation is studied by MATLAB [48]. Melting the alloy in an inert atmosphere is necessary to protect the surface of the liquid metal from oxidation and completely recycle Al–Li-type alloys [72–73]. If the Cu foil on the anode and the Al foil on the cathode can be separated after removing the powders, they may be recycled. The Al in LIBs is relatively easy to separate for recycling.

3.15. Recovery of the polymer separator (15)

While separators are not active components of LIBs, they play a key role in the safety of LIBs and help maintain the mechanical strength, thermal stability, wettability, and pore size. Many separators are obtained from the membranes of polyolefins, such as PE and PP. However, polyolefins suffer from poor thermal stability and low melting points. To address these issues, hybrid polyolefin separators with inorganic nanoparticles have been developed. Recent advances in separator membranes for LIBs are listed in Table 10 [74]. Separators are usually incinerated and buried, which could cause environmental pollution. Plastics separated from battery separators, such as PE and PP, maybe recycled after removing their impurities. New porous separators incorporated with attapulgite nanofibers (ATP), a natural mineral, sodium alginate (SA), and biodegradable polysaccharides extracted from brown algae have also been developed.

SA/ATP separators have high thermal and chemical stability. These ecofriendly separators could be degraded in soil without inducing any contamination [75].

Reports on the recycling of membrane polymers other than hybrid membranes are scarce [76]. In general, the reuse of membranes in LIBs is challenging, although recycling methods for PP and PE plates have been published. The future trend of polymer products for the next generation of solid-state batteries involves the utilization of natural polymers with controlled microstructures and excellent mechanical and thermal stability [77]. Polymer reuse and recycling are important subjects for future research.

3.16. Recycling of electrolytes (16)

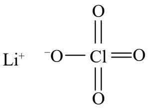
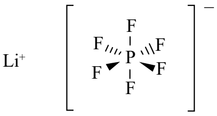
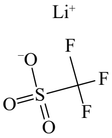
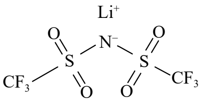
The main properties of polymer electrolytes include excellent electrochemical stability, high thermal stability, low vapor pressure, incombustibility, low viscosity, and high ionic conductivity. Liquid-based electrolytes generally consist of organic carbons, including PC, ethylene carbonate (EC), and DMC [74]. Different Li salts and solvents used in electrolyte solutions are listed in Table 11 [74,78]. Electrolytes allow Li ions to pass back and forth between the cathode and anode.

Table 10. Separator membranes for LIBs [74]

Separator	Materials (for example)									
Microporous membranes	PP	PVDF	PVDF−HFP	PVA	Cellulose	PI	PBI	PBO	PEEK	PMIA
Nonwoven membranes	PET	Ester/ Al_2O_3/PET	PVDF−HFP	Bacterial	PET/cellulose	PI	PP/SiO_2	Alginate	PVDF−HFP/SiO_2	PVDF/SiO_2
Electrospun membranes	PAN	PI/PVDF/PI	PVDF/PAN	PVDF	PVDF/PAN	PI	PAN/ SiO_2	PVDF/Al_2O_3	PAN/Lignin	PMIA
Membranes with external surface modification	PAN	PVP/PAN	PVDF/HFP	PVDF	Cellulose	PI	PET	PP/PE/PP	PE	PP
Composite membranes	PAN	PS	PVDF/HFP	PVDF	Cellulose	PP	PS	PVDF/TrFE	PE	PMMA
Polymer blends	PS/ PEO	PEO/ PVP	PEG/ LC	PEO/ PVC	PVDF/ PI	PVDF/ PAN	PVDF/ PEO	PVDF− TrFE/PEO	PVDF/ cellulose	PVA/ cellulose

Note: PAN—Polyacrylonitrile; PBI—Polybenzimidazole; PBO—Polypenzoxazole; PE—Polyethylene; PEEK— Poly ether-ether-ketone; PEG—Polyethylene glycol; PEO—Polyethylene oxide; PET—Polyethylene terephthalate; PI—Polyimide; PMIA—Poly m-phenylene iothalamate; PMMA—Pym ethylene oxide; PP—Polypropylene; PS—Polystyrene; PVA—Polyvinyl alcohol; PVDF—Polyvinylidene fluoride; PVDF−HFP—Polyvinylidene fluoride-co-hexafluoropropene; PVDF-TrEF—Polyvinylidene fluoride-co-trifuroethylene; LC—Lignocellulose.

Table 11. Different Li salts and solvents used in electrolyte solution [74,78]

Lithium salts	
Lithium perchlorate $LiClO_4$ 	Lithium hexafluorophosphate $LiPF_6$ 
Lithium trifluoromethanesulfoate $LiCF_3SO_3$ 	Lithium bis trifluoromethanesulfonimide $Li(CF_3SO_2)_2N$ [LiTFSI], etc. 
Solvents	
Dimethyl carbonate (DMC), diethyl carbonate (DEC), propylene carbonate (PC), ethylene carbonate (EC), γ-butyrolactone (BL), etc.	

Most organic electrolytes in spent LIBs are recovered by low-temperature volatilization. Electrolytes such as EC, PC, and DMC could be recycled by distillation; however, this process requires energy. After binder removal using an effective peeling agent, EC and PC could be extracted from electrodes and recovered by distillation. $LiPF_6$ could be precipitated from EC and PC and recovered by filtration [36].

Solid-state electrolytes have enabled LIBs to become smaller and more compact while maintaining the same storage capacity and power output [66]. The future development of spent LIBs has been reported, and the disposal of electrolytes must pay more attention to health and safety considerations [22]. Distillation is a useful method for recycling electrolytes. If a solid electrolyte is developed in the future, new reuse and recycling methods must be considered.

4. Trends for the 3R of LIBs

Over 1000 papers related to the recycling technology of LIBs have been published. However, reviews on the total recycling of these batteries are scarce. In this section, total recycling systems and technologies are reviewed from the viewpoint of 3R, which is closely related to a circular economy. The number of vehicles using LIBs is estimated to increase rapidly over the next 30 years. Thus, the 3R concept is important to prevent the depletion of valuable resources and achieve a circular economy. Demands for Co and Li are expected to increase in the near future.

The most valuable component of LIBs is the cathode material, especially if it contains a large amount of Co. The pyrometallurgical company prefers to collect valuable metals; therefore, the Co content similar to LCO was important before. However, the composition of the cathode materials of LIBs may be modified annually to achieve high quality and safe utilization. Amassing a large amount of spent LIBs with cathode materials of the same compositions is convenient for recycling operations but ensuring that cathode materials always have the same composition is difficult. Implementing a sorting system involving the nondestructive inspection of spent LIBs is important in efforts to collect LIBs of the same quality. If the same quality of LIBs is corrected, the reuse and regeneration of cathode in LIBs become easier. The regeneration methods of LCO, LMO, LFP, NMC, and other cathode materials have been previously investigated.

The valuable and oligopoly Co content using in the cathode is decreasing nowadays. Avoiding the use of rare metals can reduce the supply risk, but the use of less-valuable metals decreases the economic benefit of recycling LIBs. Moreover, besides metals, LIBs contain toxic organic materials. Recycling is important to maintain a good environmental condition.

The availability of Li is limited by the country producing it, and the demand for this metal is expected to surpass that for Co in the future. Li is used not only in electrodes but also in electrolytes. Li recycling is not especially popular because of the lack of an economical recycling process for this metal. Extensive recovery of the Li in spent LIBs is necessary to promote further technological development and new designs of these batteries.

The flowsheet of LIBs treatment process for 3R has been shown in this paper. A battery management system helps control the depth of discharge during the utilization of LIBs. Thus, when using LIBs for automobiles, the development of quick-charge technologies and rapid exchange systems in the charging stand must be considered. Designs aiming to upgrade the battery module and the long-life utilization by checking LIBs quality with monitoring are necessary. Many technologies must be developed to achieve the recycling and reuse of LIBs; therefore, designs oriented toward the 3R concept are important in the manufacturing process.

Finally, LIBs must be processed by recycling plants. The following recycling technologies have been well studied and developed: LIB production with low rare metal utilization; binder removal from the cathode and anode to separate particles from the metal foils; dry and wet separation after crushing as a pretreatment for recycling; metal recycling and reuse by pyrometallurgical treatment; metal recycling and reuse by hydrometallurgical treatment of the cathode and anode by using various leaching reagents.

The following recycling technologies remain insufficient and must be further investigated and developed: rapid discharge methods to recycle spent LIBs; crushing methods to liberate LIB parts without toxic gas emission; Environment-friendly binder production; polymer recovery and separator reuse; Li electrolyte recycling and environment-friendly electrolyte production.

Also, the 3R (reduce, reuse, and recycle) of new solid-state LIBs has to be considered in the future with social system and regulation.

5. Conclusions

The rapid growth in the number of publications on LIBs recycling is beneficial for future electric vehicle utilization. In this report, the complete 3R of LIBs is discussed. Many types of recycling processes for these batteries have been developed. Designs considering reuse and recycling are also necessary for the manufacturing of LIBs. As the composition of cathode materials improves, various optimal recycling systems must be prepared. Reduction of the use of valuable Co in LIBs reduces the benefit by recycling treatment. Therefore, the reuse and regeneration of cathode powders is an important undertaking. A technology that could enable the high recovery of valuable Li is also necessary. Safe treatment methods and implementation of the 3R concept for the separator and electrolyte in LIBs must be considered to recycle the whole used battery. Because energy is necessary for recycling, the place of low electricity costs can perform the recycling of LIBs to reduce the energy cost for recycling. New LIBs, such as solid electrolytes, may be developed in the future. Therefore, research on reuse and recycling technologies must be continued to address issues related to cost and environmental pollution with social system and regulation.

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