

Recent Advances in Lithium-Ion Battery Recycling and Regeneration Technologies

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Abstract

With the ever-growing need for lithium-ion batteries, particularly from the electric transportation industry, a large amount of lithium-ion batteries is bound to retire in the near future, thereby leading to serious disposal problems and detrimental impacts on environment and energy conservation. Currently, commercial lithium-ion batteries are composed of transition metal oxides or phosphates, aluminum, copper, graphite, organic electrolytes with harmful lithium salts, polymer separators, and plastic or metallic cases. The lack of proper disposal of spent lithium-ion batteries probably results in grave consequences, such as environmental pollution and waste of resources. Thus, recycling of spent lithium-ion batteries starts to receive attentions in recent years. However, owing to the pursuit of lithium-ion batteries with higher energy density, higher safety and more affordable price, the materials used in lithium-ion batteries are of a wide diversity and ever-evolving, consequently bringing difficulties to the recycling of spent lithium-ion batteries. To address this issue, both technological innovations and the participation of governments are required. This article provides a review of recent advances in recycling technologies of spent lithium-ion batteries, including the development of recycling processes, the products obtained from recycling, and the effects of recycling on environmental burdens. In addition, the remaining challenges and future perspectives are also highlighted.

Keywords: Lithium-ion batteries, battery recycling, regeneration, hydrometallurgy, pyrometallurgy, direct recycling, resource recovery.

Introduction

Owing to the high energy density, long lifespan and low self-discharge, lithium-ion batteries (LIBs) are higher portable than other commercial energy storage devices and have become the most leading power source for consumer electronics (CEs) since they were successfully commercialized by SONY in 1990s [[1], [2], [3], . Nowadays, with the rapid advances in microprocessor technology and constant offers of upgrades, the period of product updating for CEs has been largely shortened, resulting in rising production of LIBs, as well as rapid generation of spent LIBs from waste CEs [4],[5]. In the past several years, with the development of new electrode materials possessing higher energy density and power output, the application targets of LIBs have spread into the electric vehicles (EVs)[6], [[7], [8], [9]]. In the meanwhile, with the maturity of high energy-density LIBs and policy orientations, the EV market is growing rapidly. Fig. 1 shows the development of EVs in the world [10]. It can be seen that both the global EV stock and EV registrations have increased fast since 2012,

particularly in China and US. Considering that the EV market is just beginning to boom, and the lifetime of EV batteries is designed to be longer than that of the ones used in CEs, hence, we may not face serious disposal problems of spent LIBs from EVs in a few years [11,12]. However, we should foresee the influences of numerous retired LIBs from EVs on resource conservation and environmental protection in the near future, and be aware what actions should be taken now to deal with the spent LIBs.

It can be predicted that the lack of proper disposal and post-treatment of spent LIBs will result in grave consequences, such as environmental pollution and waste of resources. Generally, a LIB is composed of a cathode, an anode, a separator, electrolyte, an outer case and sealing parts. Commercial LIBs are currently using various types of Li-containing oxides and phosphides as cathode materials, such as LiCoO_2 , LiMn_2O_4 , LiFePO_4 , $\text{LiNi}_{1-x-y}\text{Co}_x\text{Mn}_y\text{O}_2$ (NCM), etc. It is shown from a survey in 2012 that more than half of the cathode material market was still occupied by LiCoO_2 and $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$, which contain hazardous heavy metal, i.e., cobalt [13]. In addition, organic electrolytes also contain harmful substances such as flammable organic solvents and fluorine-containing lithium salts [14]. Thus, directly dumping LIBs in the trash will cause serious environmental pollution. Moreover, LIBs contain high-value metals, such as Li, Co, Ni, Cu, Al, etc. Among them, Co and Li are rare and relatively more expensive than other metals, and their contents in LIBs are 5–15 wt. % and 2–7 wt. %, respectively, which are higher than those in natural ores [15].

Based on the above-mentioned point of view, recycling of spent LIBs is highly desirable and the corresponding recycling technologies and regulations should be development for establishing a complete recycling system for spent LIBs. In fact, there is already well-operated lead-acid battery recycling system, which may provide experience and guidance. For example, in the United States, ~99% of lead-acid batteries are recycled, even more than any other common daily wastes (e.g. tires, paper, aluminum cans, glass, etc.) [16]. However, it is worth mentioning that lead-acid batteries and LIBs are quite different in terms of the chemical composition of their components. For the former, ~60% of the battery mass is lead, which can be easily separated from other components after disassembling the battery [17]. For the latter, by contrast, the diversity of the materials used in LIBs makes the recycling process more complicated. As a comparison, the materials used in the components of lead-acid batteries and LIBs are listed in Table 1. Moreover, the shapes and sizes of a lithium-ion cell vary greatly, and the amount of individual cells in battery packs or modules may vary from tens to thousands (e.g. the battery module in Tesla electric car) depending on the applications, causing big problems for the recycling. The diversity of the materials and the variety of the shapes and sizes leave the recycling system for LIBs far behind that for lead-acid batteries. In spite of this, opportunities always coexist with challenges; hence recycling of spent LIBs is attracting ever-increasing attentions and many studies related to this topic have been reported [18]. This article provides a review of recent advances in recycling technologies of spent LIBs, including the development of recycling processes, the products obtained from recycling, and the effects of recycling on environmental burdens. In addition, remaining challenges and perspectives are highlighted as well.



Approaches for recycling lithium-ion batteries

A complete recycling process for LIBs generally needs two typical processes: physical processes and chemical processes due to the complicated assembly of LIBs or battery packs, as well as the diversity of electrode materials. It is noteworthy that spent LIBs still have residual energy, which may lead to fire and explosion during recycling. For this reason, spent LIBs usually need to be discharged before entering recycling process. Physical processes include pretreatments, such as

Products from recycling spent LIBs

At the last stage of a battery recycling process, the metal values from spent LIBs are transferred into other substances, such as alloys, slags, solutions and precipitates. For example, after a pyrometallurgical process, the lithium, aluminum and some iron will go into the slag, which can be used as a beneficial aggregate in concrete; an alloy with copper, cobalt, nickel and some iron will be further separated and refined into pure metals for reuse. As for hydrometallurgical process, a solution

Effect of recycling spent LIBs on environmental burdens

Generally, EVs are considered as “zero emission” vehicles compared with those equipped with internal-combustion engines. However, although EVs have no gas emission in operation, many factors related with LIBs, which are the power sources of EVs, indeed have significant impact on environment. With the market expansion of EVs, LIBs manufacture and

the upstream industries have increased substantially, consequently creating environmental burdens, such as resources consumption, energy generation and

Conclusion and perspectives

The substantial growth in the production and adoption of electric vehicles (EVs) is expected to generate a significant quantity of spent lithium-ion batteries (LIBs) in the coming years. As EV batteries reach the end of their useful service life, the increasing accumulation of spent LIBs is likely to become an important environmental and resource-management challenge. Appropriate recycling and recovery of these batteries are therefore essential because they contain considerable amounts of valuable metals, including lithium, cobalt, nickel, manganese, copper, and aluminum, as well as potentially hazardous organic electrolytes and other chemically reactive components. If spent LIBs are improperly discarded or treated, these constituents may pose risks to the environment and human health and may also result in the loss of valuable secondary resources. An ideal LIB recycling process should therefore combine high recovery efficiency with low energy consumption, minimum chemical usage, reduced waste generation, and limited environmental impact. Such a process should preferably recover not only the major valuable metals but also other useful battery components while maintaining the quality and economic value of the recovered materials. In addition, an effective recycling system should incorporate safe handling, efficient separation, environmentally responsible treatment, and the possibility of recovering materials suitable for reuse in new battery production. Developing such sustainable and economically viable recycling technologies will be essential for establishing a circular battery economy, reducing dependence on primary mineral resources, minimizing environmental pollution, and supporting the long-term expansion of electric mobility.

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