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A Brief Review on the Recycling of Decommissioned Power Battery Resources from EVs

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Abstract

With the development of electric vehicles, the industry of lithium ion battery has greatly promoted. It is predicted that the market of global lithium ion battery is expected to reach $99.98 billion by 2025, with its absolute dominance in consumer electronics and electric vehicles. The rapid and massive introduction of lithium ion battery in vehicles will produce a large number of spent batteries in 10 years. It is important to recycle spent lithium ion batteries for sustainable production. This paper summarizes the latest development of pyrometallurgical process, hydrometallurgical process and direct recycling process in industry. Currently, none of the technological process are ideal, and there are many challenges should be solved. This paper gives some suggestions for the challenges of recycling of batteries, hoping the efforts of academia, industry and government, the industry of recycling spent lithium ion battery can be further improved.

Keywords: Spent lithium ion battery; Pyrometallurgical process; Hydrometallurgical process; Regenerate

Introduction

Company	Battery types	Process	Location
Toxco	Ni, Li-based	Cyromilling (Li), pyrometallurgy (Ni)	Trail, BC, Canada Baltimore, OH, USA
Salesco Systems	All type battery	Pyrometallurgy	Phoenix, AZ, USA
OnTo Technology	Li-based	Liquid-liquid extraction	Bend, OR, USA
AERC	All type battery	Pyrometallurgy	Allentown, PA, USA Hayward, CA, USA West Melbourne, FL, USA
Dowa	All type battery	Pyrometallurgy	Japan
Japan Recycle	All type battery	Pyrometallurgy	Osaka, Japan
Sony Corp. & Sumitomo Metals and Mining Co.	All type battery	Pyrometallurgy	Japan
XStrata	All type battery	Pyrometallurgy + electrowinning	Horne Que, Nikkelverk Nor, Sudbury Ont, Canada
Accurec	All type battery	Pyrometallurgy	Mulhiem Grenada
DK	All type battery	Pyrometallurgy	Duisburg, Greece
AEA Technology	Li-based		Sutherland, Scotland
Batrec AG	Li-based, Hg	Pyrometallurgy	Wimmis, CH, Switzerland
AFE Group (Valdi)	All type battery	Pyrometallurgy	Zurich CH, Switzerland Rogerville, France
Citron	All type battery	Pyrometallurgy	Zurich CH, Switzerland Rogerville, France
Euro Dieuze/SARP	All type battery	Hydrometallurgy	Lorraine, France
SNAM	Cd, Ni, MH, Li	Pyrometallurgy	Saint Quentin Fallavier, France
IPGNA Ent. (Recupyl)	All type battery	Hydrometallurgy	Grenoble, France
Umicore	All type battery	Pyrometallurgy + electrowinning	Hooboken, Belgium

Figure 1: Summary of battery recycling process by various company all over the world.

Vehicle power battery requires frequent charging and discharging, which greatly affects the capacity of the battery. However, the capacity decreases below 80% of the initial capacity needs to be replaced. The life of lithium ion batteries in electric car is 3-6 years. The rapid development of new energy vehicles will produce a large number of spent lithium ion batteries. China's new energy vehicles entered the explosive growth stage in 2014. China will produce about 800,000 tons of spent lithium-ion batteries (134.39GWh) by 2025. Even more, the flammable and toxic waste generated from the disposal of spent batteries can cause severe environmental pollution if not carefully treated. Therefore, it is urgent to develop technologies to recycle and reuse LIBs for the benefit of both recapturing valuable materials and mitigating environmental pollution [1]. Governments and researchers all over the world are looking for an effective solution to the utilization of spent batteries. The energy department of USA has announced laboratory battery recovery and development center. China, Germany, Japan and other major electric vehicle countries have also formulated guidelines on power battery recycling (Figure 1). Thus, it is quite necessary for us to pay close attention to the recycling of the spent Li-ion batteries [2].

Traditional Process

Since cathode materials account for about 40% of the material value in typical LIBs, recycling the cathode materials is especially important for optimal economics. Novel approaches are the subject of extensive development in industry and academia. There are three different battery recycling technologies are shown in Figure 2:

a) hydrometallurgical processes,

b) pyrometallurgical processes,

c) direct recycling processes. Hydrometallurgical processes and pyrometallurgical processes are starting to operate at industrial scales, and the third is presently at the lab and pilot scale (Figure 2).

Figure 2: Global distribution of lithium ion battery recyclers.

Hydrometallurgical processes

At present, Hydrometallurgical processes consist of several chemical procedures, leaching, chemical precipitation, extraction, et al. metal values can be leached with high leaching rate. Typically, nickel, manganese and cobalt can be recovered. Some recycling processes (such as extraction process, chemical precipitation process, electrolysis process and co-precipitation process) have been developed to recycle valuable materials in the form of Li_2CO_3, $LiOH$, $NiSO_4$, $CoSO_4$, $CoCl_2$, CoC_2O_4, $Ni_xCo_yMn_{1-x-y}(OH)_2$ and $Ni_xCo_yAl_{1-x-y}(OH)_2$ from spent Li-ion batteries. Finally, for the treatment of the leachant, the solvent extraction is always adopted to obtain the Co, Li, Cu, and Al raw materials, which are further applied for fabrication of the renovated cathodes. Inorganic acids are used in this process, such as HCl, H_2SO_4, HNO_3, but the widely used of acids would cause secondary pollution, which will bring acid solution streams, resulting in wastewater pollution [3].

Pyrometallurgical processes

Pyrometallurgical processes are common in industry. However, it requires extreme temperatures (above 1400°C), high energy consumption, high costs, and the release of harmful fumes, requiring stringent safety and environmental precautions. Furthermore, this method alone cannot completely recycle all metals [4].

Direct recycling processes

Direct recycling processes has also been used in which cathode harvested from spent LIBs is sintered with a pre-determined amount of Li salt. The synthesis approach is relatively easy; however, the Li/TM (transition metal) ratio must be accurately measured before the dosage of Li_2CO_3 is investigated. The limitation of this approach is that the regeneration conditions are different from each individual battery because the Li/TM ratio changes with the cycling performance [5].

Technology prospect

Yang Shi demonstrates a simple yet efficient approach combines hydrothermal. Treatment and short annealing to regenerate degraded NCM cathode particles (Figure 3). Finally, nearly ideal stoichiometry, low cation mixing, and high phase purity were achieved in the regenerated NCM particles, which

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displays high specific capacity stable cycling stability, and high rate capability. The process with obvious advantages over traditional hydrometallurgical methods and builds an important foundation for the sustainable manufacturing of energy materials [6,7].

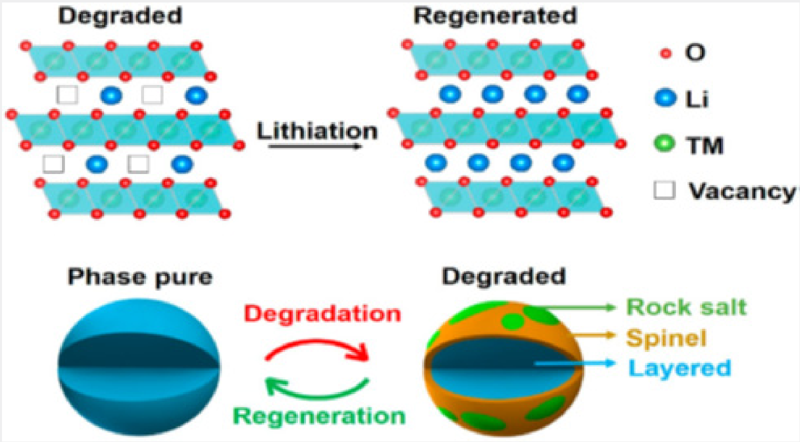

Figure 3: Illustration of the hydrothermal lithiation process.

Cao Yuan-Cheng successfully offer an innovative method to regenerate degraded $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ cathode particles to obtain new active particles. The results show that the regeneration materials display the discharge capacities of 162.0mAh/g at 0.1C. 128.6mAh/g can be obtained at 1C after 100 cycles with capacity retention of 91.9%, which is comparable with commercial cathodes [8].

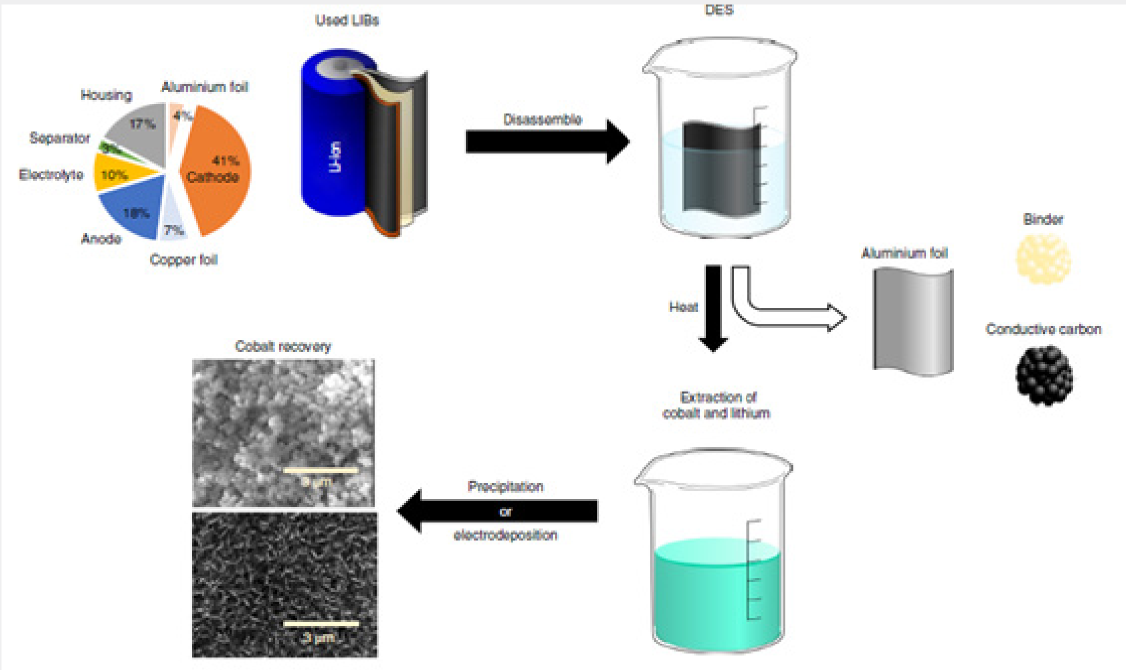

Figure 4: Battery recycling schematic.

Pulickel m. Ajayan and the Ganguli Babu team at Rice university in the United States exhibit a method to recycle LIBs using deep eutectic solvents to extract transition metals, including lithium cobalt, oxide and lithium nickel manganese cobalt oxide. For the metal extraction from lithium cobalt oxide, leaching efficiencies of ≥90% were obtained for both cobalt and lithium (Figure 4). Deep eutectic solvents could provide a green alternative compare with conventional methods of LIB recycling, which remains crucial to meet the demand of the exponentially increasing LIB production [9,10].

Conclusion

Need for development of efficient and suitable technology for valuable metal recovery from spent lithium ion batteries is

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important. The urgency for the alternative recycling technologies/ processes to recover the lithium in particular from LIB also needs an attention to avert the projected crisis in the near future. Direct recycling processes through recycling from LIB can be an alternative feasible option to meet future demand, sustainability of energy, environment, and circular economy.

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