RESEARCH IN LOGISTICS & PRODUCTION

LI-S BATTERY RECYCLING

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Abstract Lithium-ion batteries are not situable for electric vehicles with high millage, military power supplies and fixed power networks. Therefore, the Li–S batteries have been intensively investigated, due to the high capacity, low cost, widespread source, and nontoxicity. The development of Li-S batteries causes increasing need to find the methods for their recycling. Some of them are discussed in the paper. The recycling of Li-S cell relates to its anode, cathode, electrolyte, binder and separator. The Li-S battery should be fully charged before recycling. There are potential methods for recycling of lithium from anodes, especially by re-melting. It is also possible to recycle some materials from the cathodes, especially sulfur by re-melting and graphite by dry crushing, Eco-bat Technologies method or the method investigated by Xiang et al. There is no effective recycling methods for electrolytes, binders and separators. It is necessary to carry out further studies on them.

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1. INTRODUCTION

The global demand for energy needs the new electrical energy storage systems. Lithium-ion batteries are not suitable for electric vehicles with high millage, military power supplies and fixed power networks (Ellis, Lee & Nazar, 2010; Goodenough & Kim, 2010; Etacheri & et al., 2011)

The LIBs based on intercalation compounds are already facing their energy density limit, so many researches were aimed to develop new energy-storage systems with high-energy density (Bruce & et al. 2012). The Li–S batteries have been intensively investigated, due to the high capacity (1675 mAh g−1), low cost, widespread source, and nontoxicity (Manthi-ram & et al.2014; Son & et al., 2015; Pang & et al., 2016).

With the development of the optimization of design, the methods of load balancing of component cells, the methods of manufacturing and the conditions of battery operation, the problem of managing used batteries has appeared.

The problem was partially recognized for LIBs i.e. (Kushnir, 2015; Kwade & Diekmann, 2018) however, for the Li-S cells, there are very little papers regarding their recycling (Yu, Jung, Park & Goodenough, 2017; Ma & et al. 2015). This paper discussed some possible methods of Li-S cell recycling.

2. LI-S BATTERY CONFIGURATION AND COMPONENTS

2.1. Battery configuration

Typical Li-S cell uses the C-S cathode, metallic Li anode and a liquid organic electrolyte (Mikhaylik & et al., 2010). The cell can contain the binders maintaining the structural integrity of the electrodes, and the separator conducting the ions, blocking the electrons to perform reversible charging/discharging and preventing direct contact between cathode and anode.

Many efforts have been devoted to the new sulfur cathodes in Li-S cells (Barghamadi, Kapoor & Wen, 2013; Bresser, Passerini & Scrosati, 2013; Evers & Nazar, 2013; Manthiram & et al., 2014; Song, Cairns & Zhang, 2013; Chen & Shaw, 2014; Xu & et al., 2014).

The liquid electrolytes can be replaced by polymer or gel electrolytes (Park, Yeo, Park & Lee, 2010; Ahn, Kim, Ahn & Cheruvally, 2010). All-solid Li-S cell can be based on glass ceramic electrolytes (Zhu, Li & Liu, 2017; Lopez-Aranguren & et al., 2017).

The casings of Li-S cells can be made of steel in case of cylinder cell or of aluminium in case of pouch cell. They can be recycled from Li-S cells mechanically using magnetic separation or sieving (Pan & Weyhe, 2016).

The recycling of Li-S cell relates also to its anode, cathode, electrolyte, binder and separator. The recycling of the Li-S battery differs from one of the LIB.

The Li-S battery should be fully charged before recycling in contrast to the LIB battery, which needs be fully discharged before disassembly of its components.

In case of full charging the anode contains maximum volume of metallic lithium therein. Also, the cathode contains the highest amount of sulfur.

2.2. Cathode materials

A lot of strategies were used for Li-S battery cathodes, namely: using sulfur host materials (Wang, Wang, Zhang & Jin, 2013; Pang, Kundu, Cuisinier & Nazar, 2014), protective coating layers (Wei She & et al., 2013; Zhou & et al., 2013; Chen & et al., 2015; Seh & et al., 2014), and interlayer between cathode and separator (Zhou & et al., 2014).

Cathodes in Li-S battery are connected with current collectors made of Al foil or Carbon nanofiber. The former can be mechanically recycled via sieving (Pan & Weyhe, 2016).

Sulfur from the cathode can be re-melted under argon atmosphere preventing reaction of sulfur with oxygen. Sulfur can be also removed under the hot steam (Ma & et al., 2015) described the method for recycling of the S-doped graphene from cycled Li–S batteries and employing it as a metal-free electrocatalyst for the oxygen reduction reaction. The recycled SG electrocatalyst can provide the method to reuse graphene-based electrodes in Li–S batteries.

After removal of sulfur from the cathode, the graphite can be recycled using the Eco-bat method (Gaines, Sullivan, Burnham & Belharouak, 2011). This patented process contains no high-temperature processing. Its first process step involves violating the cell packaging just enough to allow fluids to be exchanged. The electrolyte is extracted using supercritical carbon dioxide. It carries the salts with it and can be reused. The CO_2 could be recovered from combustion waste. The remaining structure can then safely be chopped into small pieces that are amenable to a series of separation processes based on surface properties and solubility.

Graphite from the cathodes material can be recycled using the dry crushing method described in (Zhang & et al., 2013) or the method investigated by (Xiang & et al., 2012).

The dry crushing is carried out by a joint two-stage way. Firstly, the spent batteries are chopped in to pieces by shear crusher and then the products are crushed in the impact crusher for 20 s.

The second method involves the five steps:

- Disassembling the batteries and taking out the graphite electrode plates followed by washing them with an organic solvent (e.g., DMC) to remove collected residue electrolyte from the surface of the electrode.
- Drying of the plates in 85–100 °C for evaporation of the solvent.

- Soaking the dried graphite electrode into HCl acid solution under ultrasonic vibration to separate the graphite film from the copper foil and membrane completely. Moreover, the acidic solution step purifies graphite from the by-products of charge–discharge cycles, solid electrolyte interface layer, and carboxy methyl cellulose (CMC) thickener.
- Separating the graphite powder from the acidic solution by centrifuging, rinsing, and drying.
- Sieving, polishing, and preparing an electrode material from the dried powder to be inserted in a new battery.

2.3. Binders

Several materials, such as polyethylene oxide (PEO), Gelatin, polyvinyl pyrrolidone (PVP), Na-alginate, Gum Arabic (GA) can be alternatives to polyvinylidene difluoride (PVDF) common binder (Duan, Han, Li & Chen, 2014; Lacey, Jeschull, Edström & Brandell, 2013; Pan & et al., 2015; Chen & et al., 2015; Li & et al., 2015; Li, Cai, Liu & Li, 2015). According to (Frischmann, Hwa, Cairns & Helms, 2016), also PVP blends with Nafion, PAMAM dendrimers, polycationic-cyclodextrins, poly (acrylic acid), poly- (ethylene oxide), and carboxymethyl-cellulose: styrenebutadiene-rubber (CMC:SBR) can be binders. Binders can be recycled by the Eco-bat method (Gaines, Sullivan, Burnham & Belharouak, 2011).

2.4. Electrolytes

Electrolyte in Li-S cells can be liquid or solid, such as gel and ceramic one. Liquid electrolytes were reported in (Zhang, 2013; Kim & Jeong, 2011; Xu & et al., 2014; Barchasz, Lepretre, Patoux & Alloin, 2013).

Some ionic liquid electrolytes was presented in (Park & et al., 2013, Wang & Byon, 2013). Solid electrolytes were discussed in (Idris, Rahman, Wang & Liu, 2012; Zhao & et al., 2012).

Gel polymer electrolytes (GPE) were reported in (Zhang & Tran, 2013). Solid non-polymer electrolytes were reported in (Yamada & et al., 2014; Yamada & et al., 2015; Nagata & Chikusa, 2014; Han & et al., 2016; Wang & et al., 2016).

The electrolyte in Li-S cell also can contains additives sauch as LiNO3, poly-sulfides or phosphorus pentasulfide (P2S5) (Azimi & et al., 2015; Azimi & et al., 2015; Zhang, 2013)

Solid electrolytes and lithium salts can be recycled using the Eco-bat method (Gaines, Sullivan, Burnham & Belharouak, 2011). Liquid electrolytes can be extracted from cells, but substances used for extraction are unrecognized and require further investigations.

2.5. Anode materials

The Li-S cell often has metallic lithium anode (Liu & et al., 2017). Cheng, Huang and Zhang (2018) reported several hosts for Li anode, including carbon nanotubes (CNTs), graphene, graphene−CNT hybrid, carbon nanofiber, porous carbon, graphite particles, graphite microtubes, TiC-carbon hybrid, 3D Cu, Cu-Ni hybrid, metal foam, AlF3 framework, Li alloys, and glass fibers (Cheng & et al., 2014) presented a 3D fibrous Li7B6 host.

The composite Li metal anodes can be obtained by a melting strategy (Liu & et al., 2016; Liang & et al., 2016; Lin & et al., 2016; Jin & et al., 2017).

Using LiF surface passivation on 3D layered Li-rGO electrode, was achieved a composite Li metal anode (Lin & et al., 2017). A thin protective layer can be created on the anode surface (Cheng, Huang & Zhang, 2018; Cha & et al., 2018).

The lithium from the anode can be recycled using melting process under argon atmosphere preventing reaction of lithium with oxygen. The anodes can be frozen and then milled.

If anodes contein lithium and other metals, they can be recycled by the Eco-bat method (Gaines, Sullivan, Burnham & Belharouak, 2011).

According to (Weyhe & Pan, 2016), in the case of Li-S batteries, due possible existence of metallic lithium, safety risk should be careffuly controlled. The pre-tritment steps such as cryogenic or thermal one are recommended. The Li-S batteries contain no high-volume metals such as Ni and Co, so the recycling via the pirometalurgy are not recommended for them. The pirometalurgy is economical for recovery of Ni, Co and Cu, but is too expensive in case of Li and Al.

2.6. Separators

The separator can be made of polypropylene (PP), pure or coated by multi-wall carbon nanotubes or a Super P, or be the microporous polyolefin membrane (Yao & et al., 2014). Huang, Zhang, Peng, Liu, Qian and Wei (2014) reported a PP/PE/PP separator with a Nafion layer.

Liu, Qin, He, Li and Kang (2017) reported coating the separator by a thin layer of functional materials (carbon, polymer and oxide). Some materials, like Super-P, graphene, mesoporous carbon, multi walled carbon nanotubes (MWCNT)@PEG, Nafion, polydopamine, glassy fiber paper, black phosphorus, sulfonated acetylene black, and carboxyl functional groups, were introduced on commercial separators.

The modern separators include: graphene-protected one (Zhou & et al., 2014), PP with carboxyl groups (Yu, Joseph & Manthiram, 2016), one with graphene layers and Li4Ti5O12 nanoparticles (Zhao & et al., 2016), one with a macroporous PP matrix layer, GO barrier, and Nafion retarding layer (Zhuang & et al., 2016), porous PAN/GO nanofiber membrane (Zhu & et al., 2016), a metal–organic framework (MOF)/GO composite film (Bai & et al., 2016), single-wall carbon nanotube (SWCNT)-modulated one (Chang, Chung & Manthiram, 2016).

Separators of LIBS often are not recycled (Moradi & Botte, 2016)]. In case of carbon separators the Eco-bat method (Gaines, Sullivan, Burnham & Belharouak, 2011) can be used. Obtained materials can be reused for new separators.

3. RECYCLING PROCESS

The Li-S battery recycling process designed by Accurec was described in (Pan & Weyhe, 2016) and its flow chart is shown in the Fig. 1.

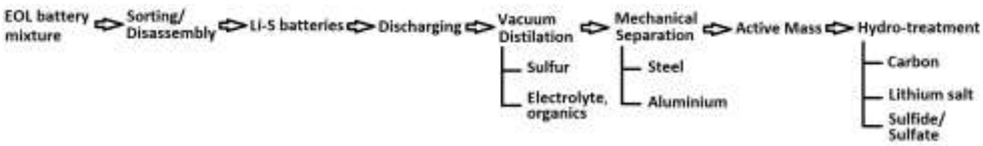

Fig. 1 The Li-S battery recycling process designed by Accurec (Pan & Weyhe, 2016)

The designed recycling process chain includes sorting/disassembly, discharging, vacuum distillation, mechanical separation and hydrotreatment. The expected recovered materials include steel, aluminium, lithium, sulfur, electrolytes and carbon. The designed process chain seems to be not verified enough. In particular, the planned discharging process of Li-S batteries raises serious doubts.

4. CONCLUSION

The recycling of the Li-S cell relates mainly to its anode and cathode. The Li-S battery should be fully charged before recycling in contrast to the LIB. There are potential methods for recycling of lithium from anodes of the Li-S batteries, especially by re-melting.

There are potential methods for recycling of some materials from the cathodes of the Li-S batteries, especially sulfur by re-melting and graphite by dry crushing, Eco-bat Technologies method or the method investigated by Xiang et al.

There are no effective recycling methods for electrolytes, binders and separators. It is necessary to carry out further studies on them.

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BIOGRAPHICAL NOTES

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