30th International Conference Ecological Truth & Environmental Research 2023

Proceedings

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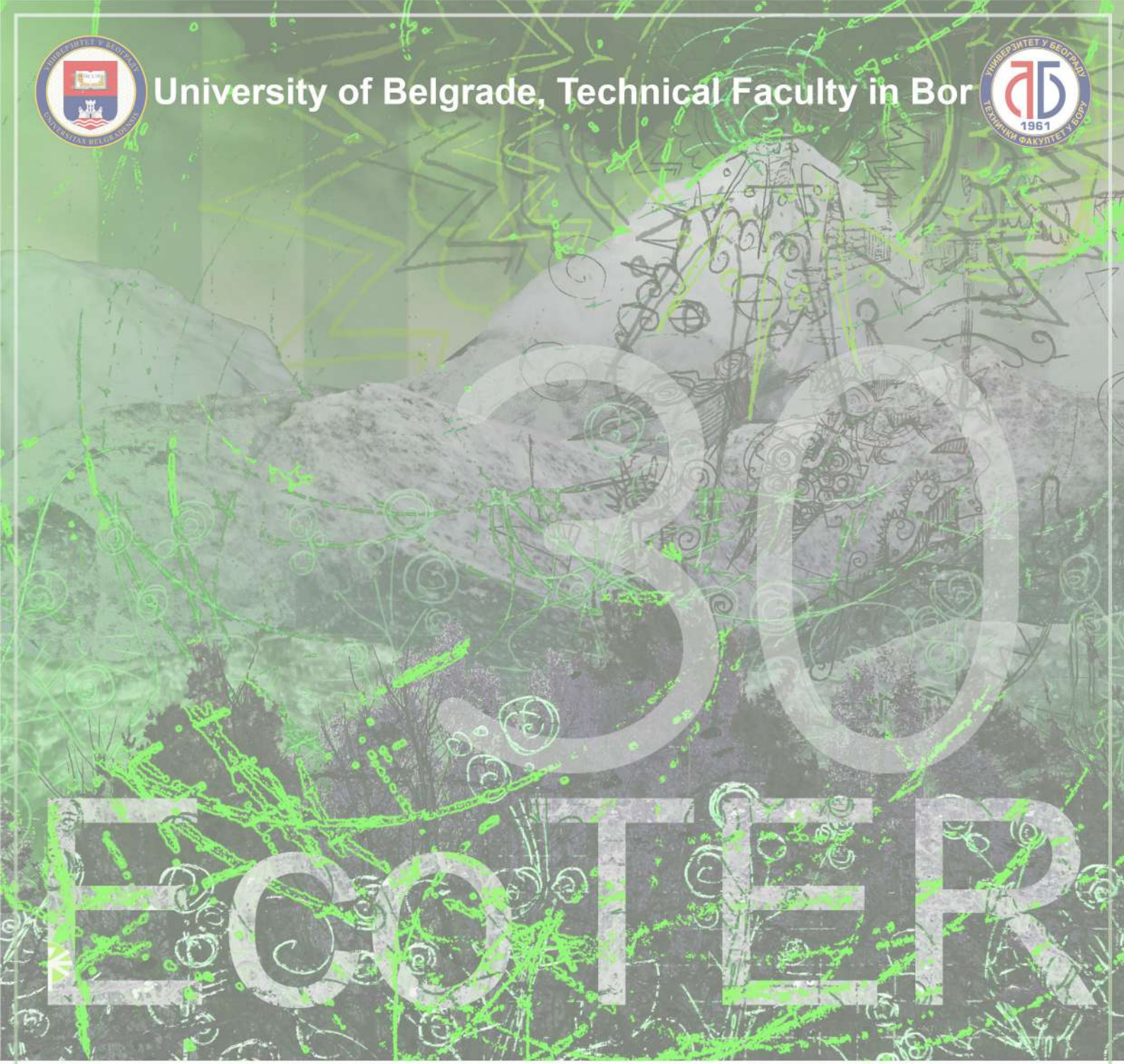

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PROCEEDINGS

30th INTERNATIONAL CONFERENCE

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PREFACE

The 30th international conference Ecological Truth & Environmental Research – EcoTER'23 kept three areas in focus: ecology, environmental protection and sustainable development. The conference will be held on Mt Stara Planina in hotel Stara Planina, Serbia, 20–23 June 2023. The monograph is published on the occasion of the 30th anniversary of the conference. On behalf of the scientific and organizing committee, it is a great honor and pleasure to wish all the participants a warm welcome to the conference.

The monograph is published on the occasion of the 30th anniversary of the conference.

We hope to convey the message of the conference, which is that a transformation of attitudes and behavior would bring the necessary changes. This is also an opportunity for the participants who are experts in this field to exchange their experiences, expertise and ideas, and also to consider the possibilities for their collaborative research.

The 30th international conference Ecological Truth & Environmental Research – EcoTER'23 is organized by the University of Belgrade, Technical Faculty in Bor, and co-organized by the University of Banja Luka, Faculty of Technology, the University of Montenegro, Faculty of Metallurgy and Technology – Podgorica, the University of Zagreb, Faculty of Metallurgy – Sisak, the University of Pristina, Faculty of Technical Sciences – Kosovska Mitrovica and the Association of Young Researchers, Bor.

These Proceedings 103 papers from the authors coming from the universities, research institutes and industries in 11 countries: Australia, USA, Brazil, Spain, Portugal, Libya, Italy, Bulgaria, Bosnia and Herzegovina, North Macedonia, and Serbia.

As a part of this year's conference, the 5th Student Session – EcoTERS'23 is being held. We appreciate the contribution of the students and their mentors who have also participated in the conference.

The support of the Gold donor and their willingness and ability to cooperate has been of great importance for the success of the EcoTER'23. The organizing committee would like to extend their appreciation and gratitude to the Gold donor of the conference for their donation and support.

We appreciate the effort of all the authors who have contributed to these Proceedings. We would also like to express our gratitude to the members of the scientific and organizing committees, reviewers, speakers, chairpersons and all the conference participants for their support to the EcoTER'23. Sincere thanks go to all the people who have contributed to the successful organization of the EcoTER'23.

Prof. Snežana Šerbula,

President of the scientific and organizing committee

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20–23 June 2023, Serbia

RECYCLING OF Li-ION BATTERIES FROM THE END-OF-LIFE VEHICLES: OPPORTUNITY OR LIABILITY IN THE FUTURE?

Ana Radojević[1*], Jelena Milosavljević[1], Snežana Šerbula[1], Tanja Kalinović[1], Jelena Kalinović[1]

[1]University of Belgrade, Technical Faculty in Bor, V.J. 12, 19210 Bor, SERBIA

[*]*aradojevic@tfbor.bg.ac.rs*

Abstract

The presence of precious metals with much higher content and purity compared to the corresponding ores, the end-of-life lithium-ion batteries (LIBs), used in electric vehicles (EVs), are known as "urban mine" and have become a research hotspot. The growing demand for the EVs will result in generation of significant quantities of waste LIBs, containing both economically and environmentally targeted materials. If not recycled, traction LIBs could be linked to the depletion of scarce metal reserves, such as lithium and cobalt. Low recycling rate, amounting to less than 5%, is currently affecting profitability of the recycling process. Two main recycling routes for LIBs – pyrometallurgy and hydrometallurgy, have numerous advantages and disadvantages. However, research studies have shown that different processes developed on a laboratory scale could result in good recycling outcomes, considering high recovery rates, achieved purity of products, profitability and eco-friendliness of the recycling process.

Keywords: Li-ion batteries, recycling, end-of-life vehicles, electric vehicles.

INTRODUCTION

The lithium (Li) market is predominantly oriented towards the production of all kinds of batteries for portable devices and electric vehicles, with 80% of the global share, followed by the production of ceramic and glass (7%) and lubricating greases (4%). In recent years, Li consumption for battery production considerably increased mostly due to the fast-growing market of electric vehicles (EVs) [1]. The EVs have a great potential to contribute to the significant reduction of the air pollution emissions in the mobility sector if they are powered on the renewable energy. However, traction Li-ion batteries (LIBs) may become problematic from a sustainability point of view, because of high need for scarce metals [2,3]. The most common nonferrous metals used in the production of the LIBs are Ni, Cu, Co, and Li, among which the Co and Li are denoted as a critical raw material since their resources are limited. According to the reports of the US Geological Survey, the majority of Li production comes from a few countries in the world: Australia, Brazil, Argentina, Chile, China, and smaller operations located in Canada, Portugal, the USA, and Zimbabwe [1,4]. More than half of cobalt production in the world originates from Congo, while China controls about 70% of graphite production, which is also needed for the LIB production [5]. Owing to continuing exploration, the total identified Li resources worldwide are around 98 million tons, mostly located in Bolivia ($21 \cdot 10^6$ t), Argentina ($20 \cdot 10^6$ t), the USA ($12 \cdot 10^6$ t) and Chile ($11 \cdot 10^6$ t). Nevertheless, Blomeke *et al.* [2] showed that the industrial recycling routes are both

economically and environmentally advantageous over the production of the battery components from the raw materials.

At the start of 2019, more than 5.6 million EVs in the world were in use, which is an increase of 64% compared to 2018. By 2040, it is expected that the EVs will account for 58% of all globally sold automobiles [6]. If the LIBs from the end-of-life (EOF) vehicles are hoarded or dumped, non-renewable natural resources are irretrievably wasted. Since the enormous size of the EV´s batteries (e.g. the Tesla Model 3 Long Range battery weighs 480 kg) [6], a substantial amount of waste with high economic value will be produced every year, reaching 21 million tons in the period 2015–2040 [5]. The profit of LIB recycling is up to 3300 €/t, with avoided impacts of emitted 5–7 t of $CO_{2(eq)}$ per tonne of EOF LIBs [6]. The current amount of EOL LIBs affects the profitability of the recycling process. However, the number of EOL LIBs is projected to increase by 2030 due to the rising number of the EVs that will reach their EOL [2], since the average usage of the LIBs is 8 to 10 years [5]. According to Wasesa *et al*. [7], the price of waste LIBs must be less than $227/t in order to be profitable at the lowest incentive condition, while the LIB recycling cost should be $1803/t at the highest incentive scenario. The variety of active cathode materials and the mixing of unknown elements from the production and utilization are the main reasons for difficult and expensive recovery processes [8]. Nevertheless, the reuse of recovered materials in manufacturing of traction LIBs would cause a reduction for all the considered environmental impacts [9]. According to the European legislation, EOL LIBs need to be recycled with a minimum recovery rate of 65% by 2025, which is the medium level of ambition option of the European Commission proposal. Furthermore, the material-specific recovery rates are also set for the valuable components, such as Co, Ni, and Cu, amounting to 90% as well as for Li amounting to 35% of the recovery rate from the EOL LIBs [10].

The paper aims to analyse the main aspects of available recycling processes for the recoverable materials from the EOL LIBs.

RESULTS AND DISCUSSION

Composition of the LIBs

The main components of the LIB structure are battery shell, cathode, anode, separator, and electrolyte. The battery shell is composed of stainless steel, aluminium or plastic. The cathode consists of conductive carbon, binder polyvinylidene fluoride, Al-foil, and active material. There are five types of cathode active material: lithium nickel manganese cobalt oxide ($LiNi_xCo_yMn_zO_2$), lithium iron phosphate ($LiFePO_4$), lithium cobalt oxide ($LiCoO_2$), lithium manganate ($LiMn_2O_4$) and lithium nickel cobalt alumina ($LiNi_xCo_yAl_zO_2$). The anode is usually composed of binder polyvinylidene fluoride, Cu-foil, graphite or conductive carbon, while the electrolyte consists of organic solvents, Li-conductive salts ($LiPF_6$, $LiBF_4$ or $LiClO_4$) and additives [8,11,12].

Available recycling technologies for the EOL LIBs

There are three feasible options for the EOL EVs batteries, depending upon their design, quality, and state of condition: remanufacturing, repurposing, and recycling. Remanufacturing and repurposing extend the usage of the LIBs, while recycling closes the loop [5].

The selection of the recycling process for the EOL LIBs is driven by the difference between the profit of the recycled products and the sum of the costs of the recycling process, other than the recycling rate efficiency and the environmental impact of the process [8]. Dunn *et al*. [13] showed that 15–18% of Co, 9–11% of Li, and 15–17% of Ni demand in 2035, could be met by the closed-loop recycling of the EOL LIBs. As the most valuable component of the LIB, the cathode active materials account for more than 1/3 of the total battery cost [14]. However, if organic solvents and lithium salts are completely recovered from the electrolytes, the profit with a total value of $3345/t of electrolyte could also be gained [15].

Recycling of cathodes could be done *via* smelting in pyrometallurgy, *via* leaching in hydrometallurgy, or without destruction of the crystalline structure of the electrode active material *via* direct recycling. Combination of the given processes could also be employed, such as co-precipitation route, which is a process between hydrometallurgy and direct recycling. The outcomes of the recycling process can differ considerably regarding the recovered materials, achievable purities of the recycled metals, recovery rates, required infrastructure, process emissions, investments, costs, and revenues [2,14,16].

The metallurgy-oriented methods are widely used on the industrial level, while the direct recycling is still in the research stage, but considered as a promising method during which the cathode active materials from the EOL LIBs are directly regenerated [14]. By November 2022, 44 companies in Canada and the USA, and 47 companies in Europe were already involved in recycling or planning to recycle the LIBs [1]. The pyrometallurgical route is implemented mainly in Europe, Japan and North America, while hydrometallurgical route has been mostly commercialized in China [14]. Latini *et al*. [16] reported the plant capacities of most of the recycling facilities in Europe, North America, and Asia. In Table 1, the data is given for the largest ones, according to the recent capacities.

Table 1 *The location, capacities and recycling technology of some LIB recycling facilities [16]*

Company (Location)	Capacity (t/year) for specific year	Recycling technology
Umicore (Begum)	7000 (2020)	Pyro/hydrometallurgy
Glencore* (Norway)	7000 (2020)	Pyrometallurgy
Nortvolt AB (Norway)	8000 (2022)	Mechanical/hydrometallurgy
Stena (Sweden)	10000 (2023)	Mechanical/hydrometallurgy
Neometals (Germany)	10000 (2022)	Mechanical/hydrometallurgy
Erasteel Recycling* (France)	20000 (2020)	Pyrometallurgy
INMETCO* (USA)	6000 (2020)	Pyrometallurgy
Glencore (Canada)	7000 (2020)	Pyrometallurgy
Li-Cycle (USA)	60000 (2023)	Hydrometallurgy/co-precipitation
Nippon Recycle Centre Corp.* (Japan)	5000 (2020)	Pyrometallurgy
SungEel Hi Tech (South Korea)	8000 (2020)	Mechanical/hydrometallurgy
Guanghua Sci-Tech (China)	10000 (2020)	Mechanical/hydrometallurgy
Highpower International* (China)	10000 (2020)	Mechanical, pyro/hydrometallurgy
GEM* (China)	30000 (2020)	Mechanical/hydrometallurgy
Huayo Cobalt (China)	65000 (2020)	Mechanical/ hydrometallurgy

* Facility is recycling batteries other than Li-ion batteries, such as Ni-metal hydride battery, NiCd, etc.

The pyrometallurgical route can utilize different LIB chemistries, as well as geometries, without pre-sorting. However, Li could not be recovered unless the formed slag is subsequently refined. The hydrometallurgy-based processes are more chemistry-specific, but necessarily require pre-treatment [16]. More advantages and disadvantages of pyro- and hydrometallurgical-oriented processes are listed in Table 2.

Table 2 *The key advantages and disadvantages of the pyro- and hydrometallurgical routes for recycling of end-of-life LIBs [2,8,12,16]*

Pyrometallurgy		**Hydrometallurgy**	
Advantages	Disadvantages	Advantages	Disadvantages
Versatile process	Valuable materials are burnt	High recovery rates	Sorting or pre-treatment needed
High capacity of facilities	Downcycling for Li	Recycling of the electrolytes	Lower capacity of facilities
No sorting or pre-treatment needed	Waste gases treatment needed	High purity of obtained products	Generation of wastewaters
High recovery rates for Co, Ni and Cu	High energy consumption (up to 1700 °C)	Lower energy consumption (up to 100 °C)	High operating costs
Industrial application	Low purity of metals	Low emission of waste gases	Complexity of process
Simple process	Further processing of slag is needed	Recovery of the most LIB components	Wastewater treatment needed

Pyrometallurgy is based on smelting of whole batteries in a furnace at extremely high temperatures in order to recover an alloy of precious metals, such as Ni, Co and Cu. The electrolyte is evaporated in the low-temperature zone of the furnace and proceeded to energy recovery unit, while plastics and graphite parts are burnt in the high-temperature zone. The slag, consisted of Li, Al, Mn, and partly of Fe, is typically forwarded to the low-quality markets, although it could be refined via hydrometallurgy to obtain high-purity metals [12].

Hydrometallurgy is based on the leaching in strong inorganic acids (e.g. H_2SO_4, HCl, HNO_3) or alkalis (NaOH), with an additional reducing agent (e.g. H_2O_2), for dissolving the cathode active materials, in order to recover them as single-phase metal salts through crystallization, selective precipitation, solvent extraction, or by electrochemical methods. As an environmentally friendly option, the LIBs could be treated by bio-hydrometallurgy (e.g. microbiological metal dissolution or fungal bioleaching), leached with organic acids (e.g. citric, ascorbic, oxalic or malic acid) or with water because Li-salts are soluble in water [12,17,18]. However, some methods are far from the industrial application. Prior to hydrometallurgical route, a pre-treatment is usually applied in order to increase the recovery rate and reduce the high costs [12]: (a) mechanical separation of electrode materials encapsulated in iron and plastic, including crushing, grinding, gravity and magnetic separation; (b) thermal treatment (e.g. vacuum pyrolysis) in which organic additives and binders are pyrolyzed at high temperatures in order to liberate electrode materials from the foils; (c) mechano-chemical treatment, as a grinding technique, is used to decompose the

crystal structure of metal active materials, extracting Co and Li by an acid leaching process at the room temperature; and (d) dissolution treatment used to weaken the adhesive substance by organic solvents. The electrochemical methods (such as electrolytic deposition, electrodialysis, aqueous electrolysis, and molten salt electrolysis), could replace the traditional chemical leaching of the EOL LIBs. In these methods, the recycling efficiency is greatly improved, and at the same time high reagent consumption and environmental hazards are reduced, which are the main disadvantages of the leaching process. However, many of the electrochemical methods are developed only on a laboratory scale [8,17].

The direct recycling consists of separating and regenerating the cathode and anode active materials without decomposing them into elements. The Li-inventory, lost during battery usage, is restored by keeping the particle morphology and crystalline structure, thus recycled product can be directly reused in new LIBs [12]. The direct recycling has the potential to be environmentally and economically sustainable because the energy and water consumption, as well as the emissions of greenhouse gasses and SO_x are much lower compared to the virgin production of the LIB components and pyro/hydrometallurgical recycling routes. Beyond the challenge of repairing the crystal structure of electroactive materials, there are still many obstacles facing the practical application of the direct recovery [19]. Solid-state sintering, hydrothermal regeneration, eutectic molten salts, and thermal, hydrothermal and electrochemical lithiation are some of the methods, mostly developed at the laboratory scale [16,19].

Zhang *et al*. [20] proposed the concept of carbothermal reduction in combination with three-stage leaching as easy and eco-friendly approach to recycle Li from the LIBs with high Al content, with the comprehensive recovery rate of 87.15%. According to Chakraborty and Saha [6], the optimum choice for the LIB recycling is blending of mechanical shredding, electrolyte extraction, electrode dissolution and Co electrochemical reduction. According to the available data, Dunn *et al*. [13] showed that the recycling of LIBs is less expensive in China than in the USA, neither by conducted transport by truck nor train. Blomeke *et al*. [2] emphasized that, in order to manage the high costs associated with transport of spent batteries as hazardous goods, it is crucial to establish a flexible and scalable recycling infrastructure that can meet the needs for the future market growth and utilization. Makwarimba *et al*. [17] concluded that the focus in the future should be on the development of abundant and non-toxic anode, cathode and electrolyte materials for the LIB production, as well on the new battery designs. Despite the progress made in upscaling conventional methods, there are still few medium to large-scale recycling facilities in the world.

CONCLUSION

In future years, it is crucial to establish a sustainable recycling system that will ensure infrastructure readiness and support the needs for much greater volumes of the LIBs – the valuable, but also hazardous kind of waste. The aim of further research must be feasible connection of a complex relationship among recycling companies, manufacturers of the LIBs, and suppliers of the raw materials on a global scale. If the concept of circular economy is applied on LIBs, the loop could be closed, by combining the supply chain management, sustainability assessment, and innovative engineering. Both, the pyro- and hydrometallurgy-

oriented processes have major drawbacks, such as low purity of the recycled products and downcycling of Li, or high complexity and smaller capacity of the processes, respectively. On the other hand, outcomes of various recycling processes on the laboratory scale, have confirmed that recycling of EOL LIBs is a great opportunity, but also a huge liability for the future.

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