Authors' responses to the Reviewers' comments

SUMMARY

The authors would like to express their gratitude to all the reviewers involved in the process of improving our manuscript. We appreciate all the remarks regarding the text clarification, stylistic and logical issues, and the article's proper assembling. We hope all the concerns are now fully addressed and thoroughly elucidated.

Here, the reviewers' commentaries are numbered and enclosed after a vertical bold line. The authors' responses are located right after the reviewers' comments. Portions of the response text are **bolded** for emphasis. Excerpts from the first manuscript submission are presented in a blue font. Changes made to the revised manuscript text in response to the reviewer's comments have been highlighted here and in the revised manuscript.

REVIEWER #3

Li-ion battery cathode recycling: an emerging response to growing metal demand and accumulating battery waste is an interesting review by the authors. I want to recommend for its publication after a few minor issues are fixed:

We appreciate the Reviewer for the careful evaluation of our work in the field of Li-ion batteries recycling processes. Below, we tried to address all the mentioned commentaries and support them with the changes in the text.

COMMENTARY 1

The authors should write clearly how this review work is different in terms of novelty and why this review is needed more deeply in the introduction.

The authors thank the Reviewer for such a valuable request to emphasize the practical difference of our review from the others devoted to Li-ion batteries (LIBs) recycling. Accordingly, we rephrased the main text (Pages 2-3, Lines 79-104):

In this review, from **the chemical point** of view, we analyze the LIBs utilization technologies with a particular emphasis on the **cathode** recycling. The current paper mainly reviews the most recent recycling trends (2020-2022). Throughout the review, we follow the logic to pay attention

to every recycling stage in a universal manner and consider every technological option for each stage: weather it seems outdated (discuss briefly) or emerging (provide more details and analysis). Such a broad and topical coverage distinguishes this review from the other ones, which focus separately on either the recycling of EV cathodes [1], $LiCoO_2$ cathodes [2], and all LIBs' components [3-4], pretreatment stage of the recycling [5], or the commercialized technologies [6]. In other words, we suggest this review as a **self-sustain**, independent study that is enough for a primary and sufficient immersion into the LIB cathode recycling field. Indeed, this review covers the whole spectrum of the cathode recycling processes starting from cell discharging and finishing with battery-grade salts extracting. After an introduction (environmental impact, existing recycling processes, section 1), we focus strictly on the LIB's processing methodology and the chemistry used within — discuss the recycling structure, provide specific conditions and reagents, and analyze advantages and limitations of each method (section 2). Reasonable attention is devoted to the hydrometallurgical branch of recycling as one of the most promising routes for the end-of-life (EoL) LIBs treatment. Further, we proceed through the latest cathode restoration techniques and, finally, finish with a summary and perspective for the LIB cathode recycling. With the current review article, we would like to provide interested scientists, especially, novices to the LIB recycling culture, with valuable and hands-on both theoretical and practical knowledge. We hope that the LIBs recycling technological aspects discussed in this paper will enhance the search and then implementation of novel sustainable recycling routes.

[1] Or, T., Gourley, S. W., Kaliyappan, K., Yu, A., & Chen, Z. (2020). Recycling of mixed cathode lithium-ion batteries for electric vehicles: Current status and future outlook. *Carbon Energy*, *2*(1), 6-43.

[2] Botelho Junior, A. B., Stopic, S., Friedrich, B., Tenório, J. A. S., & Espinosa, D. C. R. (2021). Cobalt recovery from Li-ion battery recycling: a critical review. *Metals*, *11*(12), 1999.

[3] Piątek, J., Afyon, S., Budnyak, T. M., Budnyk, S., Sipponen, M. H., & Slabon, A. (2021). Sustainable Li-ion batteries: chemistry and recycling. *Advanced Energy Materials*, *11*(43), 2003456.

[4] Wang, Y., An, N., Wen, L., Wang, L., Jiang, X., Hou, F., ... & Liang, J. (2021). Recent progress on the recycling technology of Li-ion batteries. *Journal of Energy Chemistry*, *55*, 391-419.

[5] Kim, S., Bang, J., Yoo, J., Shin, Y., Bae, J., Jeong, J., ... & Kwon, K. (2021). A comprehensive review on the pretreatment process in lithium-ion battery recycling. *Journal of Cleaner Production*, *294*, 126329.

[6] Baum, Z. J., Bird, R. E., Yu, X., & Ma, J. (2022). Lithium-ion battery recycling ─ overview of techniques and trends. *ACS Energy Lett.* 2022, 7, 2, 712–719.

COMMENTARY 2

I cannot find what the authors think about a framework for developing future recycling infrastructures.

We appreciate this valuable Reviewer's comment requesting to discuss the future trends and perspective for LIB recycling. Below, we introduce the corresponding discussion to the main text (page 32, lines 1121-1146).

With the rapid development of the EV and stationary energy storage markets, the demand for high-energy and high-power LIBs will continue growing in the next decades. Therefore, exploring efficient cathode recycling processes is essential for the further sustainable development of the LIBs industry. The global authorities forecast both extensive and intensive growth of the recycling infrastructure. By 2022, 32 ktons $year^{-1}$ recycling capacity has been already installed, whereas additional 70 ktons are planned for this decade to be set worldwide [1]. Besides new recycling gigafactories, novel technologies should be developed and introduced to boost the metals recovery efficiency and minimize the cost of the process.

Emerging LIB cell pretreatment technologies are expected to crucially influence future recycling. For instance, in the case of hydrometallurgy automatic cell dismantling would improve the safety and efficiency of the whole recycling process by minimizing active material losses and enhancing selectivity.

Selective metal leaching is another promising method to improve overall efficiency. It allows to extract specific metals, e.g., Co, from spent mixed cathodes or multi-component scrap leaving less valuable or toxic components in the residue. The economical and time issues of the NMC cathodes recycling would be overtaken significantly with this approach. Leaching organic acids, in addition to their eco-friendliness, also bear several other benefits. They can be used as sol-gel precursors in the cathode restoration processes and serve as precipitation and reducing agents.

The alternative concepts of cathode material resynthesis and its direct regeneration might be simpler and more cost-efficient for the LIBs production industry compared to the hydrometallurgical methods. Both approaches would deprive the recycling process with the complex steps of metal ions separation, extraction, and salt precipitation. The main issue of direct regeneration to overtake is the unification of the methodology to be applicable for a large variety of rapidly exploring types of cathode materials.

[1] Baum, Z.J., Bird, R.E., Yu, X., & Ma, J. (2022). Lithium-Ion Battery Recycling ─ Overview of Techniques and Trends. *ACS Energy Lett.* 7(*2*), 712–719.

COMMENTARY 3

The authors should write and discuss the topic of LIB recycling differentiation between academic approaches and industrial reality.

The authors express their gratitude to the Reviewer for the idea to emphasize the differences between lab-scale and industrial features of LIB recycling. Below, we have addressed this issue in more detail (page 9, lines 333-341).

Much attention will be devoted to the designed routines, process conditions, and chemicals used by leading research groups specialized in LIBs recycling over the last decade. Looking ahead, it is important to note there are recycling steps already suitable for use in industry (e.g., pyrometallurgy; cell crushing; sieving, and separation of crushed fractions), other methods are currently adopting to a large scale (inorganic acid leaching; metal ions separation; salt precipitation), whereas the rest part is emerging and capable only with the laboratory conditions (organic, alkaline, bioleaching; cell dismantling; cathodes direct regeneration; etc.). Throughout this section, we emphasize a correlation between an exact technology of a certain recycling stage and the acceptable application level, as well as discuss requirements and perspectives for the technology to be introduced into the industry.

COMMENTARY 4

Some of the important references must be discussed: Sustainable Materials and Technologies 32, e00396, 2022 and Journal of Cleaner Production 384, 135520, 2023.

The authors appreciate the Reviewer's suggestion and are thankful for providing us with highly relevant publications in the area of LIB recycling applications and general energy storage. We have introduced the proposed citations into the manuscript (page 2, lines 58).

It was not a surprise that over the last ten years, the demand for LIBs has drastically increased [1-2].

[1] Bejigo, K.S., Natarajan, S., Bhunia, K., Elumalai, V., & Kim, S.J. (2023). Recycling of value-added products from spent lithium-ion batteries for oxygen reduction and methanol oxidation reactions. *Journal of Cleaner Production*, *384*, 135520.

[2] Sahu, M., Hajra, S., Jadhav, S., Panigrahi, B. K., Dubal, D., & Kim, H. J. (2022). Bio-waste composites for cost-effective self-powered breathing patterns monitoring: An insight into energy harvesting and storage properties. *Sustainable Materials and Technologies*, *32*, e00396.

COMMENTARY 5

The authors should make a picture from one idea to make the readers understand LB recycling, its challenges, and what are possible ways to overcome them.

The authors would like to thank the Reviewer for the great idea to include discussing the challenges, possible ways for overcoming them, as well as their practical impact on the LIBs recycling outcomes. Below, we carefully address this request and introduce Figure 7 and the corresponding discussion in the manuscript (pages 32-33, lines 1147-1161.

In the end, we suppose it is necessary to summarize the main obstacles, promising solutions, and future targets for LIBs recycling. The current Challenges can be conditionally separated into three groups: technological, geographical, and economic (Figure 7). While the technology-related issues (e.g., sustainable aspects, unification recycling as well as ineffective metal separation) were thoroughly discussed in the current review, the geography and economy problems were just superficially addressed. Overall, attentive control over the amount of accumulating EoL LIBs, lobbying the recycling regulations, tuning raw/recycled materials supply chain for LIBs production, and promoting other circular economy principles [1] would complete transforming LIB recycling into an environmentally benign and cost-efficient procedure. Finally, a high volume (>80%) of LIB recycling would indemnify cells

manufacturing the effects of sudden raw material supply shortages and reduce the total cost of the LIBs production.

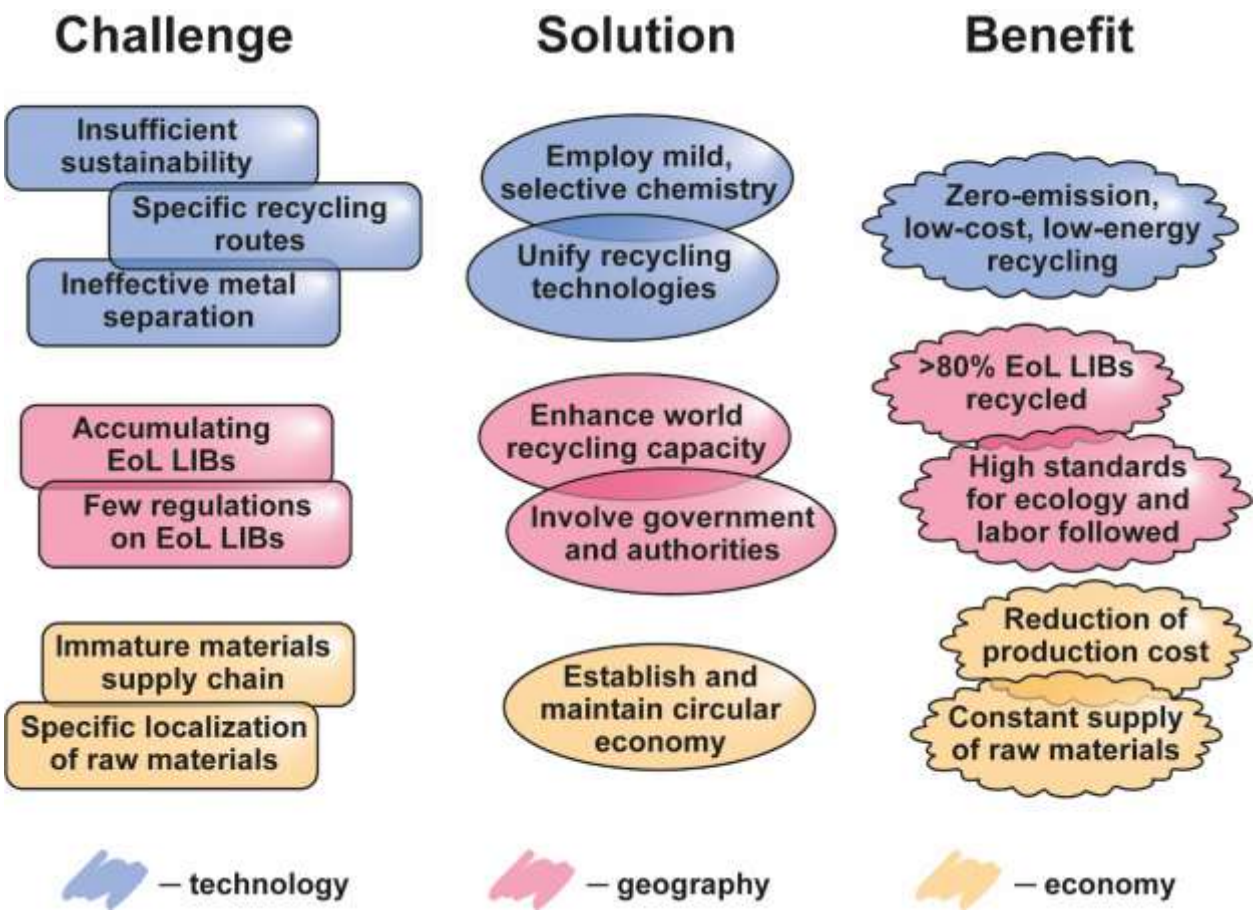


Figure 7. Perspective roadmap of EoL LIB recycling development illustrating the existing Challenges, proposing Solutions, and Benefits that can be gained as a result of solutions implementation.

[1] Piątek, J., Afyon, S., Budnyak, T. M., Budnyk, S., Sipponen, M. H., & Slabon, A. (2021). Sustainable Li-ion batteries: chemistry and recycling. *Advanced Energy Materials*, *11*(43), 2003456.

COMMENTARY 6

English grammatical errors need to be corrected throughout.

We thank the Reviewer for the kind request to check the English language throughout the review. Accordingly, we introduced the grammatical improvements as much as we could.n All changes are highlighted and tracked.