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 logic 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 reviewer's comments have been highlighted here and in the revised manuscript.

REVIEWER #1

Thank you for sending me the manuscript for review. The manuscript seems a good review on Li-ion battery recycling. My recommend is to include and discuss the recycling achieving for low-temperature Li-ion battery in a subheading. I think the manuscript discusses advances in Li-ion Batteries. The overall presentation, figures, conclusions and reference sections are fine. The manuscript may be considered for publication after revision.

We would like to thank the reviewer for elaboration of the manuscript and its appreciation. Below, we have addressed the reviewer's feedback to the manuscript.

COMMENTARY 1

I suggested the author include a topic on low temperature Li-ion batteries and discuss their recycling process.

The Reviewer has raised a really important question regarding low-temperature Li-ion batteries (LIBs) and their recycling. Indeed, at temperatures below 0 °C LIBs generally face severe energy and capacity losses, impeded interfacial ionic transfer, and poor ions diffusion in bulk electrodes. In general, the main problems for LIBs at low temperatures are associated with the graphite anode (slow Li intercalation, high resistance of SEI) and liquid electrolyte (solidification,

increased resistance). The anode-related issues are currently trying to be solved by the graphite oxidizing and metal nanoparticles' introduction (reduction of lithiation overpotential), particle size reduction, and structure and morphology modification (reduce Li plating). The spectrum of electrolyte improvement solutions is large and implies salt and solvent modification. A more thorough analysis of the specific trends for low-temperature LIBs can be observed in the latest review and research papers [1-3].

Meanwhile, the cathodes are not the most critical factor to consider when designing low-temperature LIBs. The modern cathodes, i.e., $LiCoO_2$, $LiMn_2O_4$, Li-NMC, were shown to demonstrate quite a decent intercalation/deintercalation kinetics even at sufficiently low temperatures (< −60 °C) [4-6]. Therefore, the choice of the **cathode and its recycling route** does not depend on the specific modification of the LIB (whether it is low-, medium-, or high-temperature oriented). If so, everything discussed in the current review manuscript can also be related to the low-temperature LIB cathodes.

Overall, considering the specific focus on **cathode** recycling in our review, we suppose the **whole** consideration of the recycling of low-temperature LIBs as a whole to be the subject of another (no less valuable) review paper.

[1] Zhang, N., Deng, T., Zhang, S., Wang, C., Chen, L., Wang, C., & Fan, X. (2022). Critical Review on Low-Temperature Li-Ion/Metal Batteries. *Advanced Materials*, *34*(15), 2107899.

[2] Xu, J., Wang, X., Yuan, N., Ding, J., Qin, S., Razal, J. M., ... & Gogotsi, Y. (2019). Extending the low temperature operational limit of Li-ion battery to− 80 C. *Energy Storage Materials*, *23*, 383-389.

[3] Collins, G. A., Geaney, H., & Ryan, K. M. (2021). Alternative anodes for low temperature lithium-ion batteries. *Journal of Materials Chemistry A*, *9*(25), 14172-14213.

[4] Chen, Кю, Yu, Z., Deng, S., Wu, Q., Zou, J., Zeng, X. (2015). Evaluation of the low temperature performance of lithium manganese oxide/lithium titanate lithium-ion batteries for start/stop applications. *Journal of Power Sources*, *278*, 411-419.

[5] Ramanujapuram, A., Yushin, G. (2018). *Adv. Energy Mater.*, *8*, 1802624.

[6] Sun, Z., Jiao, L., Fan, Y., Li, F., Wang, D., Hanab, D., & Niu, Li (2016). *RSC Adv*, *6*, 97818-97824