Authors' responses to the Reviewers' comments

Here, the reviewer's 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.

Following the Reviewer's strong recommendation, we added the following section into the manuscript regarding low-temperature Li-ion batteries recycling (pages 10-11, lines 353-390):

A special attention should be devoted to the low-temperature (LT) LIBs. 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 LTs 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 the salt and solvent modification [1-3]. An efficient electrolyte for LT LIBs can be obtained by

introducing F-contained chemistry (e.g., LiTFSI salt; fluoroethylene carbonate solvent additive, etc.) that would stabilize the SEI and provide the electrolyte with high ionic conductivity and low viscosity.

Improving cathodes is another path towards the LT LIBs development. Currently, besides the enhanced impedance, at LT the cathodes suffer from dissolution and re-deposition of transition metals on the surface of separator and anode [4]. The common solutions for the LT cathodes are surface coating (reduces contact resistance [5]), metal doping (improve the mobility of Li^+, stabilizes cathode-electrolyte interface (CEI) [6]), and particle size reduction (facilitates lithium insertion/extraction [7]). Overall, the modification of existing cathodes for LT LIBs requires a careful design of CEI by tuning the transition metal ratios and cathode surface pretreatment (e.g., by LiF-rich layers). In addition to the cathode modification of common materials (LCO, NMC, LFP), their substitution to emerging ones might be also beneficial for LT LIBs, such as $Li_3V_2(PO_4)_3$, Nb_2O_5, or Ni-based Prussian blue analogues [8].

Of course, both the cathode modification and use of completely new materials call for severe modification of the recycling process of LIB as a whole and that of cathode materials, in particular. Such, the LiF introduction requires a its responsible deactivation method during the recycling to avoid HF emission. The possible options are either selective capturing of the hazardous F-contained gases or LIB's components treatment in strong bases. e.g., NaOH [9]. The use of V-based cathodes additionally requires specific leaching conditions for reductive/oxidative dissolution of vanadium. The sulfuric acid leaching solutions enhanced with a reductive agent (H_2O_2, Na_2SO_3) are capable to recover stable V^{4+} in the form of $VOSO_4$ [10]. The use of expensive transition metals (e.g., Nb) as dopants or a main material bears additional economical challenges to the recycling of such cathodes. The losses of such high-cost materials should be minimized, otherwise the recycling profitability will be questioned. Overall, the LT LIBs development and, particularly, their recycling technology demands a deeper investigation in the nearest future and deserves a separate comprehensive overview. Meanwhile, in the current study we are mostly focused on the recycling of traditional cathode materials: LCO and NMC.

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