

Fast-charging and durable all-solid-state lithium-sulfur batteries enabled by pre-embedded solid-solid redox mediator

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This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Nature Communications.

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #3

(Remarks to the Author)

I am generally in favor of accepting the paper for publication. The rounds of revisions have significantly improved the quality of the manuscript. There are two suggestions for the authors in light of their responses to previous reviews.

1) Add stack pressure information of Table S1. In general, the value of comparing rate capability between different papers is limited. As the authors stated, there are too many factors behind the numbers. It is very difficult for an ordinary reader to appreciate the subtle differences behind the numbers. The authors need to add statements in the text to help the readers in this regard.

2) A similar statement is needed when the rate and cycling data are presented. It will be helpful to put these numbers into better context. This can be done by having a discussion at the end of the manuscript to lay out what remains challenging for the technology.

3) The authors should also put the proposed mechanism into better contexts. In the latest response, the authors proposed that the residual pores might encourage the redistribution of iodine. That is a reasonable proposal but in itself does not really confirm a mechanism. Perhaps adding a short discussion at the end of the manuscript on what questions remain on the mechanism (in light of the current findings) would help the readers.

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Reviewer #3 (Remarks to the Author):

I am generally in favor of accepting the paper for publication. The rounds of revisions have significantly improved the quality of the manuscript. There are two suggestions for the authors in light of their responses to previous reviews.

We sincerely appreciate the reviewer's positive and constructive feedback on our work. We have carefully considered all of the suggestions and have made corresponding revisions. Our responses are provided below.

1) Add stack pressure information of Table S1. In general, the value of comparing rate capability between different papers is limited. As the authors stated, there are too many factors behind the numbers. It is very difficult for an ordinary reader to appreciate the subtle differences behind the numbers. The authors need to add statements in the text to help the readers in this regard.

We thank the reviewer for this valuable suggestion. We have added the stack pressure information to Table S1 in the revised manuscript. A brief discussion has been included in the revised manuscript to emphasize that comparisons across different studies should be made cautiously because of differences in experimental conditions.

2) A similar statement is needed when the rate and cycling data are presented. It will be helpful to put these numbers into better context. This can be done by having a discussion at the end of the manuscript to lay out what remains challenging for the technology.

We appreciate the reviewer's insightful comment. Following the reviewer's suggestion, we have incorporated a brief explanation in the revised manuscript to provide better context for the rate capability and cycling performance. We have also highlighted the remaining challenges associated with the practical application of all-solid-state batteries, including high-energy-density negative electrode, lithium metal/electrolyte interfacial stability, and high-conductivity solid electrolyte.

3) The authors should also put the proposed mechanism into better contexts. In the latest response, the authors proposed that the residual pores might encourage the redistribution of iodine. That is a reasonable proposal but in itself does not really confirm a mechanism. Perhaps adding a short discussion at the end of the manuscript on what questions remain on the mechanism (in light of the current findings) would help the readers.

We appreciate the reviewer's insightful comment. The proposed mechanism is supported by multiple structural, chemical and electrochemical characterizations presented in this work. Nevertheless, we believe that the role of residual pores in promoting iodine redistribution represents a reasonable interpretation based on the current evidence. We have now added this part with proper words. Also, we have added a brief discussion at the end of the manuscript to highlight the remaining questions to be resolved in future studies, regarding how the iodine transport within the electrode and the kinetics of iodine reaction with Li_2S and lithium polysulfides.