

Design Strategies and Future Directions of Sparingly Solvating Electrolytes for Lithium-Sulfur Batteries

Corresponding Author: Dr Alexander Santiago

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

Dear Dr Santiago,

Thank you for submitting your manuscript titled "From Innovation to Application: A Perspective on the Combination of Sparingly Solvating Electrolytes and Electrocatalysts in Commercializing Lithium-Sulfur Batteries". Regrettably, we cannot publish it in Communications Materials.

Among the considerations that arise at this stage are the likelihood that the manuscript would seem of considerable interest to those working in the same area and the degree to which the results will stimulate new thinking in the field. In this case, while we do not question the validity of your work, I am afraid we are not persuaded that the content of your Perspective represent a sufficiently original or in-depth discussion to justify publication in Communications Materials.

We would, however, be willing to consider a revised manuscript, as long as additional personal views are added, and we suggest narrowing down the topic to only the electrolyte and excluding the catalyst part, as currently discussing both of these lacks depth. We suggest reformatting it into a 5000-word review to accommodate this.

Please use the following link if you wish to submit revised manuscript files:

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Best regards,

Golareh Jalilvand
External Editor
Communications Materials

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Version 1:

Decision Letter:

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Dear Dr Santiago,

Thank you again for submitting your Review "Sparingly Solvating Electrolytes for Lithium–Sulfur Batteries: Design Principles, Progress, and Perspectives" to Communications Materials. We have now received reports from 2 reviewers and, based on their comments, we have decided to invite a revision of your work. You will find the reviewers' reports below. While they find your work of interest, they have raised important points which must be addressed in a revised manuscript.

To allow us to move forward with your work, we also ask that you edit your manuscript according to the attached table.
Please read this document carefully as we will be unable to further assess your revised paper until these important points are addressed.

Please outline all revisions made in the right-hand column and return the completed table with your updated manuscript files as a Related Manuscript file.

When resubmitting, please also include:

- A point-by-point response to the reviewers' comments. If you are unable to address specific reviewer requests or find any points invalid, please explain
- A clean version of your revised manuscript with no mark-ups
- A marked-up version of your paper with all changes highlighted in a different colour

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We hope to receive your revised paper within six weeks, but we understand that the revisions may take longer. Please let us know if you find that the revision process will take substantially more time.

We are committed to providing a fair and constructive peer-review process. Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

John Plummer, PhD
Chief Editor
Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

This paper reviews the latest advances in sparingly solvating electrolytes (SSEs) for lithium–sulfur batteries (LSBs), highlighting their potential in addressing key challenges such as the lithium polysulfide (LiPS) shuttle effect. The paper points out that by regulating the solubility of polysulfides, promoting the formation of stable interfaces, and maintaining efficient ion transport, SSEs can significantly enhance battery performance. The authors emphasize the importance of rational electrolyte design and call for consideration of environmental sustainability during the development process. Finally, the paper proposes future research directions, including hybrid electrolyte systems and dynamic solvation control, to advance the transition of lithium–sulfur batteries from laboratory research to commercial applications. Nevertheless, I do have some concerns on the manuscript, and some revisions are required before it can be accepted for publication.

1. It better to expand Section 2 to incorporate a comprehensive schematic diagram illustrating the polysulfide shuttle effect. Additionally, include a flowchart that details the progression of chemical reactions during the discharge and charge cycles, with a focused emphasis on clarifying the formation pathway and migration mechanism of lithium polysulfides (LiPS). On this basis, further discuss the specific impact of LiPS migration on the system's Coulombic efficiency and capacity decay, and strengthen the persuasiveness of the discussion by citing specific relevant studies that provide quantitative data (e.g., numerical changes in Coulombic efficiency or capacity retention rates) to verify these effects.

2. In Section 4.1, deliver a detailed elaboration on the physicochemical descriptors applied in the design process of solid-state electrolytes (SSEs). Specifically, include key parameters such as maximum electrostatic potential (ESP_{max}), donor number (DN), and viscosity. Meanwhile, elaborate on how these parameters modulate the solvation structure of SSEs and further affect their ionic conductivity, with a focus on explaining the underlying mechanism of such influences. To enhance clarity and comparability of the information, it is advisable to supplement this section with a comparative table that lists different diluents, their corresponding physicochemical properties, and the associated performance implications.

3. In the conclusion section, it is necessary to clearly sort out the limitations of current solid-state electrolytes (SSEs), including issues such as high cost, insufficient scalability, and poor viscosity. At the same time, specific and operable future research directions should be proposed—for instance, carrying out the exploration of hybrid electrolyte systems and promoting the development of new low-cost, sustainable materials. This approach will provide a more clear and actionable

roadmap for subsequent research in this field.

4. The overall language is fluent, but it is advisable to carefully check for grammatical and spelling errors to enhance the professionalism of the manuscript.

5. Some review papers are related to this work and maybe helpful to the readers. (<https://doi.org/10.1002/cey2.641> ; Adv. Mater. 2024, 36, 2312812 ; Adv. Funct. Mater. 2025, 35, 2501761)

Reviewer #2 (Remarks to the Author):

In this manuscript, the authors critically assess recent advances in sparingly solvating electrolytes (SSEs), including highly concentrated, localized high-concentration, and weakly solvating systems, as strategies for enabling lean-electrolyte operation and long cycle life. The merits and limitations of each SSE class are discussed, with a focus on key design principles, environmental sustainability, and cell-level constraints. The manuscript is clear in logics and fundamental in solvation science. Therefore, the manuscript can be published in Commun. Mater. after minor revision. Here are my detailed comments as follows:

1. From the aspect of electrolyte, the cathodic conversion pathway can be divided into the solid–solid conversion and the solid–liquid–solid conversion. Which pathway can be the future technology especially under lean E/S ratio in authors' opinion?
2. From the aspect of cathode, there are mainly SPAN and C/S cathodes. The highly concentrated, localized high-concentration, and weakly solvating electrolytes exhibit different behaviors in the above two types of cathodes. Which one would be preferable to achieve both high energy density and long cycling lifespan?
3. Regulating the lithium polysulfide solvation structures and its solubility are the mainstream in previous researches. In authors' opinion, which one, either the solvation structure or solubility, is more significant for future electrolyte design?
4. How did authors depict the Figure 5? Are there any quantitative indicators? It would be better if the scale bars of the statistics in the figures are further explained.
5. To broaden the content of the manuscript, some relevant papers concentrating on the polysulfide solvation and regulations are suggested to be cited, e.g., J. Am. Chem. Soc. 2025, 147(18), 15435–15447; J. Am. Chem. Soc. 2025, 147(38), 34632–34640; Angew. Chem. Int. Ed. 2025, 64(32), e202504898; Angew. Chem. Int. Ed. 2022, 61, e202114671; Nat. Chem. Eng. 2024, 1, 588–596.

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Version 2:

Decision Letter:

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Dear Dr Santiago,

Thank you once again for submitting your manuscript, "Design Strategies and Future Directions of Sparingly Solvating Electrolytes for Lithium-Sulfur Batteries," to Communications Materials. It has now been seen again by the referees, whose comments are appended below. The concerns of our reviewers have now been addressed, but there are some amendments needed before we can accept your paper.

We ask that you edit your manuscript according to the attached table. **Please read this document carefully as we will be unable to further assess your revised paper until these important points are addressed.**

Please outline all revisions made in the right-hand column and return the completed table with your updated manuscript files as a Related Manuscript file.

Please use the link below to submit your revised files:

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When resubmitting, please provide a marked-up manuscript with all changes highlighted, as well as a clean version of your paper.

We hope to receive this updated version of your paper within 1 week, but please let us know if you find that you

need more time.

Best regards,

John Plummer, PhD
Chief Editor
orcid.org/0000-0003-4824-8497
Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Authors have revised the manuscript carefully and all the questions raised have been addressed well. Therefore I recommend it for publication at the present form.

Reviewer #2 (Remarks to the Author):

The authors have addressed my comments well. The manuscript is suggested to be published in the current form.

Version 3:

Decision Letter:

Dear Dr Santiago,

We are delighted to accept your manuscript titled "Design Strategies and Future Directions of Sparingly Solvating Electrolytes for Lithium-Sulfur Batteries" for publication in Communications Materials. Thank you for choosing to publish your interesting work with us.

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not be published elsewhere and there must be no announcement of the work in the media until the publication date. At this stage, you may wish to make your institution's press office aware of the forthcoming publication, if you wish to bring your work to the media's attention, so that they can start preparing any publicity. Please note that the paper is still under embargo until it is published in the journal. Further details of our embargo policy can be found here <http://www.nature.com/authors/policies/embargo.html>.

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We look forward to publishing your paper.

Best regards,

John Plummer, PhD
Chief Editor
orcid.org/0000-0003-4824-8497
Communications Materials

***As a new journal, we would greatly appreciate any comments you have about your experience at Communications Materials. I hope that we have been able to meet your expectations and look forward to working with you again in the future.

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