

Peer Review File

Coating layer design principles considering Lithium chemical potential distribution within solid electrolytes of Solid-State Batteries



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8th May 24

Dear Dr Kimura,

Thank you for submitting your manuscript, "Coating Layer Design Principles Considering Li Chemical Potential Distribution within Solid Electrolytes in Solid-State Batteries", to Communications Materials. It has now been seen by 3 referees, whose comments are appended below. You will see that while they find your work of interest, some important points are raised. We are interested in the possibility of publishing your study in Communications Materials, but would like to consider your response to these concerns in the form of a revised manuscript before we make a decision on publication.

We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type 'Revised Manuscript - Marked Up' when uploading the manuscript file to our online system.

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We hope to receive your revised paper within three months; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we will close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

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,

Andrew Westover

Guest Editor

Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The manuscript Coating Layer Design Principles Considering Li Chemical Potential Distribution within Solid Electrolytes in Solid-State Batteries gives fundamental insight into the mechanism of the way that coating layers work at an active material (AM)/solid electrolyte (SE) interface in solid-state batteries. The results and methods are clearly presented and the interdependence of different parameters on the working principle of the coating layer is discussed. The main part of the manuscript is clear and matches with what is known in literature (e.g. reference 23). I think the manuscript can be improved in terms of terminology and explanation of the fundamental concepts underlying the methodology. This would be helpful for the wider readership of the journal and I recommend major revisions particularly to these parts:

(1) Terminology:

The most important terms around the concepts of Fig. 1 need to be better explained. Use of the specific terms is crucial to avoid confusion. For the general reader it is helpful to have a brief introduction to the meanings of these terms, their relationship with electrochemical potential and why a high voltage corresponds with a low μ_{Li} . Please update the manuscript or SI with the information fundamental to these concepts:

-what does it mean 'chemical potential of lithium in a phase (cathode, SE)', what is the meaning of μ_{Li} ? Why is only μ_{Li} relevant for SE stability? What about the chemical potential of the other elements present in SE or cathode? Are they relevant?

-what is electrochemical stability, electrochemical window, oxidation limit; are they synonymous or are there differences? How is the electrochemical window of SE determined/calculated; meaning, what defines the limit of electrochemical window of SE based on μ_{Li} ?

-voltage, potential,... Which potential: galvanic, surface, electrode, cell,...? Against which reference?

(For example, in the sentence: 'When the SE near the cathode interface is exposed to high voltages, namely to low μ_{Li} , outside the electrochemical window, ...' Which voltage is meant exactly? What do the authors mean to be an electrochemical window of a material?)

(2) Cell convention, figure design:

-There is a convention that the negative electrode stands left and the positive electrode stands right in a cell design or drawing. The authors should also align their drawings with this convention so that they can be interpreted more intuitively.

-Figure 2a. the sign for SE is missing in the top diagram

(3) The interphase formation in Figure 1a, is this due to self-decomposition of the SE because of low μ_{Li} beyond the electrochemical window of SE as in ($\text{SE} \rightarrow \text{degradation products}$) or does it account for the chemical reaction between cathode and SE as in ($\text{cathode} + \text{SE} \rightarrow \text{degradation products}$)? What are the degradation products that form the interphase?

(4) Before contact between cathode and SE, the μ_{Li} in each phase is constant throughout the materials (horizontal lines at different μ_{Li} in each phase). But after contact μ_{Li} in cathode is still horizontal but in SE phase there is a slope. Why is μ_{Li} horizontal in cathode phase but sloped in SE phase? Shouldn't there be either a slope in both phases or in neither phase? Or, is μ_{Li} simply assumed to be constant in cathode as a boundary condition?

(5) During cycling, there is a lithiation gradient in the cathode. How does this influence the SE stability? Is only the μ_{Li} at the surface of the cathode relevant?

(6) in Figure 2c, why is there no case in which there is a step of μ_{Li} at the cathode then a horizontal line of μ_{Li} through the SE and then another step of μ_{Li} at the anode?

(7) In the reaction of ($\text{cathode} + \text{SE} \rightarrow \text{degradation products}$) there are many complex reaction paths and degradation products that determine the thermodynamic and kinetic stability of the interface 'cathode + SE'. What about oxidation of other elements in the SE by the cathode (e.g. in thiophosphate solid electrolytes, oxidation of sulfur and phosphorus are often observed)? Is this also dependent of μ_{Li} ?

(8) A possible degradation mechanism is the oxygen release of cathodes. So, formed oxygen gas may also react with the SE as in ($\text{O}_2 + \text{SE} \rightarrow \text{degradation products}$). How does this affect μ_{Li} and the requirements of the CL?

(9) page 8 line 231 '...the μ_{Li} at the interface surpassed the oxidation limit of the electrochemical window, indicating that the CL can inhibit the oxidative decomposition of the SE.' The oxidation limit appears to be at around -3.6 eV if I read the figures correctly: is this a fixed value and how was it obtained? What is the oxidation limit of other SE?

(10) Additional suggestion:

Could the authors provide a calculation tool (matlab, excel, origin, or so) as supplementary material that can be freely downloaded and modified (ionic, electronic, geometric values etc) by anyone, so that the calculations can be readily adapted to other materials systems and parameters?

Reviewer #2 (Remarks to the Author):

The work by Kimura et al. explores coating layer properties for SSBs to understand design principles associated with the thermodynamic stability of the solid electrolytes with the cathode active materials. The manuscript is well written, with the conclusions adequately supported by the results provided. The following points need to be considered prior to publication:

1. Mixed ionic and electronic conducting interlayers are crucial for achieving quasi-stable interphases for operations as related in the literature (Adv. Sci. 2023, 10, 2302263, Energy Mater Adv. 2021;2021:DOI:10.34133/2021/1519569, etc.). Can the authors relate their insight into design of MIECs specifically – they touch base on ionic and electronic conductivity separately, but how does the combination play out?
2. Having a table of typical coating layer materials with reported ionic, electronic and thickness values from the literature might be a good reference point.
3. It is understandable that the chemical stability is influenced by the thickness of the coating layer. However, why does the stability of the coating layer depend on the thickness of the solid electrolyte? Given that they are thermodynamic calculations – shouldn't the chemical stability of the interface be independent of the thickness of the SE itself.
4. The authors suggest increasing the thickness of the CL when the electronic conductivity is higher. Can they provide a justification for reduced ion transport across that CL in case of thick CL layers? Is there a maximum CL beyond which the ionic conduction drop would be so much, that the benefit of improving chemical stability might not justify the CLs?
5. The premise of coating SEs, opposed to the CAMs is interesting postulate. Are there any literature experimental studies that showcase that? If so, can you reference them. If not, can you provide some pathways to design these?

Reviewer #3 (Remarks to the Author):

Kimura et al. extensively investigated the thermodynamic and kinetic characteristics of cathode barrier layers for solid-state composite cathodes. The authors found that thickness and electronic conductivity of the barrier layers play a critical role in stabilizing anodic interfaces by continuum modeling. Here are my comments.

(1) The conditions for barrier coating found in this work is not different from what has been established in the field. Given the solid electrolyte will see full electrochemical potential of the cathode only by having ionic (Li^+) and electronic (e^-) percolation, improving electrochemical stability by making the interfaces insulating is obvious. It actually has been covered in many papers, such as Xiao et al., *Nat. Rev. Mater.* 5, 105-126 (2020) and Zhu et al., *ACS Appl. Mater. Inter.*, 7, 23685-23693 (2015).

(2) The manuscript used fixed modeling parameters (ionic and electronic conductivities) from known materials. As the calculation framework is set, it would be more useful and insightful to compare different systems to understand the optimum coatings. For instance, determine conditions required for the barrier coating for systems involving LLZO against NCM and Argyrodite against NCM.

(3) Above analysis may lead to a few candidate materials for coating.

Dear Reviewer 1

We sincerely appreciate you taking the time and energy to review our manuscript and provide your comments. We tried to be responsive to your concerns. We hope you find these answers meet your expectations.

Comment 1-1 (Terminology):

what does it mean 'chemical potential of lithium in a phase (cathode, SE)', what is the meaning of μ_{Li} ? Why is only μ_{Li} relevant for SE stability? What about the chemical potential of the other elements present in SE or cathode? Are they relevant??

Response:

First and foremost, we would like to express our sincere gratitude for your thorough understanding of the manuscript's content and your thoughtful consideration of how to make it more accessible to a broader readership. Regarding the terminology-related comments, we have made revisions to address your suggestions to the best of our ability. We believe that the revisions made based on your advice have significantly improved the quality of our manuscript.

Now, to address your specific question, 'the chemical potential of lithium in a phase (cathode, SE)' follows the standard definition in thermodynamics, representing the chemical potential of electrically neutral Li. When discussing the thermodynamic stability of materials, the chemical potential of neutral Li is crucial, rather than the chemical potential or electrochemical potential of Li ions. μ_{Li} represents the aforementioned chemical potential of neutral Li, which serves as the driving force for Li-related chemical reactions and Li diffusion. Furthermore, it essentially determines the electrode potentials of both electrodes in lithium-ion batteries.

As such, the meaning of the term "chemical potential of lithium" used in this paper is consistent with its usage in materials science. Therefore, for readers of *Commun. Mater.*, a journal specialized in materials science, particularly those of the special issue "Overcoming challenges in all-solid-state batteries," explicitly defining this term may seem unnecessary. In fact, within the same special issue to which we submitted this manuscript, more specialized terms such as "Electrochemical window" are treated as preliminary knowledge and used without definition¹. Furthermore, in general chemistry journals, papers that theoretically discuss SE stability² and coating layer stability³ also treat chemical potential as preliminary knowledge, using it without definition. Following these previous studies, we would like to treat 'chemical potential of lithium in a phase (cathode, SE)' and ' μ_{Li} ' as preliminary knowledge in this paper because we consider providing detailed explanations for these terms would be redundant and may obscure the main focus of this paper. However, we recognize the importance of properly defining key terms. Therefore, as explained below, we have added definitions

for more specialized terms such as 'Electrochemical window' in the main text. Additionally, to clarify that the chemical potential of lithium is the chemical potential of electrically neutral lithium and is thus directly related to thermodynamic stability, we have revised it to "chemical potential of neutral lithium."

In the original text, we did not claim that "only μ_{Li} is relevant for SE stability". In the introduction of the original text, we stated, "The chemical and electrochemical stability of the interface is essentially governed by the chemical potential of **the constituent elements of the SE, as well as the cathode and the anode**. Particularly, the chemical potential of mobile elements, *i.e.* the chemical potential of lithium (μ_{Li}), play a crucial role in defining the electrochemical stability of the interface in SSBs. (lines 20-23 on p.2 in the original manuscript)." We assert that μ_{Li} is particularly related to the electrochemical stability of SE. Here, the electrochemical stability of SE refers to the stability of SE against oxidation and reduction induced by the variation in the cathode or anode electrode potentials during (dis)charge. To clearly convey this to readers, we have added definitions of electrochemical stability and electrode potential, and the explanation for the relationship between electrode potential and μ_{Li} in our revised manuscript.

References in this response

1. Li, S. *et al.* Sulfide-based composite solid electrolyte films for all-solid-state batteries. *Commun. Mater.* **5**, 1–11 (2024).
2. Schwietert, T. K. *et al.* Clarifying the relationship between redox activity and electrochemical stability in solid electrolytes. *Nat. Mater.* **19**, 428–435 (2020).
3. Xiao, Y., Miara, L. J., Wang, Y. & Ceder, G. Computational Screening of Cathode Coatings for Solid-State Batteries. *Joule* **3**, 1252–1275 (2019).

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have included a definition of "electrochemical window" on lines 17-19 in p.2 in the revised manuscript: "The electrochemical window of most SEs, *i.e.*, the electrode potential range within which the SE remains stable without undergoing oxidative or reductive decomposition, are known to be narrow relative to the operating voltage of solid-state batteries^{19,20}."
- We have changed the phrase "chemical potential of lithium" and "chemical potential of mobile elements" to "chemical potential of **neutral** lithium" and "chemical potential of **neutral** mobile elements", respectively. The relevant revised portions can be found on lines 25 and 26 of p.2.
- We have included a definition of "electrochemical stability" on lines 26-28 in p.2 in the revised manuscript: "... electrochemical stability of the SE at the anode and cathode interfaces, namely, the stability of the SE at the interfaces against oxidation and reduction induced by the variation of the cathode or anode electrode potential during

(dis)charging²³.”

- We have included a definition of “electrode potential” on lines 15-17 in p.2 in the revised manuscript: “Here, the electrode potential is defined as the electromotive force between the electrode in question and a Li metal reference electrode, measured using a SE with a transference number of 1 as the electrolyte.”
- We have included an explanation for the relationship between electrode potential and μ_{Li} on lines 28-29 in p.2 in the revised manuscript: “This is because the electrode potential (E_{ele}) is essentially determined by the μ_{Li} of the electrode ($\mu^{\text{ele}}_{\text{Li}}$): $E_{\text{ele}} = -\mu^{\text{ele}}_{\text{Li}}/F$, where F is the Faraday constant.”

Comment 1-2 (Terminology):

what is electrochemical stability, electrochemical window, oxidation limit; are they synonymous or are there differences? How is the electrochemical window of SE determined/calculated; meaning, what defines the limit of electrochemical window of SE based on μ_{Li} ?

Response:

As mentioned in the previous response, the electrochemical stability of the SE refers to the stability of the SE against oxidation and reduction induced by the variation of the cathode or anode electrode potential during (dis)charge. The electrochemical window is defined as the electrode potential range within which the SE remains stable without undergoing oxidative or reductive decomposition. The oxidation limit represents the μ_{Li} corresponding to the oxidation threshold of the electrochemical window, beyond which the oxidative decomposition of the SE begins. These definitions have been added to the main text of the manuscript.

Theoretical attempts to explain at which μ_{Li} the SE decomposes are still in progress, in our opinion. Therefore, generally, the electrochemical window of the SE is experimentally determined using electrochemical measurement techniques such as linear sweep voltammetry¹. Efforts have also recently begun to theoretically determine the electrochemical window of the SE, and it has been reported that good agreement with the experimentally evaluated electrochemical window can be obtained by considering the reaction pathway via Li-insertion/extraction of the SE². In this work, we focus on discussing the conditions required for the CL to thermodynamically stabilize the SE under a given electrochemical window of the SE. Therefore, the details of the electrochemical window of SEs, including how it is determined, fall outside the scope of this paper. We thus would greatly appreciate your understanding in allowing us to refrain from elaborating on this point within the manuscript.

References in this response

1. Hassoun, J. *et al.* A structural, spectroscopic and electrochemical study of a lithium ion conducting $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ solid electrolyte. *J. Power Sources* **229**, 117–122 (2013).

2. Schwietert, T. K. *et al.* Clarifying the relationship between redox activity and electrochemical stability in solid electrolytes. *Nat. Mater.* **19**, 428–435 (2020).

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have included a definition of “electrochemical stability” on lines 26-28 of p.2 in the revised manuscript, as noted in the response to Comment 1-1.
- We have included a definition of “electrochemical window” on lines 17-19 of p.2 in the revised manuscript, as noted in the response to Comment 1-1.
- We have included a definition of “oxidation limit” on lines 10-11 of p.5 in the revised manuscript: “To prevent the oxidative decomposition of the SE, the $\mu_{\text{Li}}^{\text{int}}$ needs to be greater than the μ_{Li} corresponding to the oxidation threshold of the electrochemical window of the SE, beyond which the oxidative decomposition of the SE begins (oxidation limit),...”

Comment 1-3 (Terminology):

Voltage, potential,... Which potential: galvani, surface, electrode, cell,...? Against which reference? (For example, in the sentence: ‘When the SE near the cathode interface is exposed to high voltages, namely to low μ_{Li} , outside the electrochemical window, ...’ Which voltage is meant exactly? What do the authors mean to be an electrochemical window of a material?)

Response:

Thank you for your valuable comment. In the mentioned sentence, we intended to refer to the electrode potential with respect to the Li^+/Li reference. However, we agree that the expression may not have been accurate. In the revised manuscript, we have replaced the term "voltage" with "electrode potential" where necessary. Furthermore, we have revised the ambiguous expressions that used the terms "potential" and “voltage”. In addition, in the revised manuscript, we have included the definition of electrode potential in this work and an explanation for its relationship with μ_{Li} .

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have replaced the term "voltage" with "electrode potential" where necessary. Furthermore, we have revised the ambiguous expressions that used the terms "potential" and “voltage”. The relevant revised portions can be found:
 - * Original MS: “...compromised by the voltage change...” → Revised MS: “...compromised by electrode potential variation...” in line 15 of p.2
 - * Original MS: “...exposed to voltages...” → Revised MS: “...exposed to the electrode potential...” in line 19-20 of p.2
 - * Original MS: “...high voltages...” → Revised MS: “... high electrode potential...” in line 30-31 of p.2
 - * Original MS: “... anode and cathode potential...” → Revised MS: “... anode and cathode electrode potential...” in line 31 of p.2

p.6

* Original MS: "... oxidation potential limit..." → Revised MS: "...oxidation limit..." in line 4 of p.8.

* Original MS: "... terminal voltage..." → Revised MS: "... μ_{Li} at the cathode and anode interfaces, *i.e.*, the cathode and anode electrode potentials..." in line 2-3 of p.15.

* Original MS: "... the cathode terminal voltage..." → Revised MS: "... the cathode electrode potential..." in line 4-5 of p.15.

* Original MS: "... anode terminal voltage..." → Revised MS: "... anode electrode potential..." in line 7 of p.15.

* Original MS: "... anode terminal voltage on the interfacial stability..." → Revised MS: "...anode electrode potential on the SE/CL interfacial stability..." in line 8-9 of p.15.

* Original MS: "...anode terminal voltages..." → Revised MS: "... anode electrode potential..." in line 10 of p.15.

* Original MS: "...anode terminal voltages..." → Revised MS: "...anode electrode potential..." in line 13 of p.15.

* Original MS: "...anode terminal voltage..." → Revised MS: "...anode electrode potential..." in line 14 of p.15.

* Original MS: "...anode terminal voltage..." → Revised MS: "...anode electrode potential..." in line 15 of p.15.

* Original MS: "...anode terminal voltage..." → Revised MS: "...anode electrode potential..." in line 17 of p.15.

* Original MS: "...anode terminal voltage..." → Revised MS: "...anode electrode potential..." in line 19-20 of p.15.

* Original MS: "...anode terminal voltage..." → Revised MS: "...anode electrode potential..." in line 21 of p.15.

* Original MS: "...terminal voltage applied at a spatially distant position..." → Revised MS: "... electrode potential applied at a spatially distant position from the CL..." in line 23-24 of p.15.

* Original MS: "... fixed terminal voltages..." → Revised MS: "... fixed μ_{Li} at the cathode and anode interfaces..." in line 27 of p.15.

* Original MS: "...anode terminal voltages..." → Revised MS: "... anode electrode potentials..." in line 12 of p.19.

- We have included a definition of "electrode potential" on lines 15-17 of p.2 in the revised manuscript, as noted in the response to Comment 1-1.
- We have included an explanation for the relationship between electrode potential and μ_{Li} on lines 28-29 in p.2 in the revised manuscript, as noted in the response to Comment 1-1.

Comment 2-1 (Cell convention, figure design):

There is a convention that the negative electrode stands left and the positive electrode stands right in a cell design or drawing. The authors should also align their drawings with this convention so that they can be interpreted more intuitively.

Response:

Thank you for your comment. We were aware of the convention you mentioned, but in the original manuscript, we placed the cathode interface on the left side of the graph, *i.e.*, at the origin, as we believed it to be more appropriate when focusing on the cathode interface, as in this paper. However, following your suggestion, we have revised Figures 1 and 2 to place the anode on the left and the cathode on the right. Consequently, this modification necessitated changes in the layout of all other

figures as well as Eqs. (13) and (14), which have been updated accordingly.

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have revised all the figures to place the anode on the left and the cathode on the right.
- We have revised Eqs. (13) and (14) to take the anode interface as the origin.
- Additionally, we have modified some of the textual expressions to accommodate the changes made to the above equations and figures. The relevant revised portions can be found:

* Original MS: "...with the origin defined at the interface between the CL and CAM..." → Revised MS: "... with the origin defined at the interface between the SE and anode..." in line 3 of p.5

* Original MS: "... $\mu_{Li}^{int}(= \mu_{Li}(L_{CL}))$..." → Revised MS: "... $\mu_{Li}^{int}(= \mu_{Li}(L_{SE}))$..." in line 7 of p.5

* Original MS: "... $\mu_{Li}(L_{CL}+L_{SE})$..." → Revised MS: "... $\mu_{Li}(L_{SE}+L_{CL})$..." in line 13 of p.5

* Original MS: "... where the left and right ends correspond to the interfaces with the cathode (lower μ_{Li}) and the anode (higher μ_{Li})..." → Revised MS: "...where the left and right ends correspond to the interfaces with the anode (higher μ_{Li}) and the cathode (lower μ_{Li})..." in line 12-13 of p.6

* Original MS: "... μ_{Li} sharply increases near the cathode, approaching nearly equal to that at the anode side..." → Revised MS: "... μ_{Li} sharply decreases near the cathode..." in line 13-14 of p.6

* Original MS: "... , corresponding to the voltage applied to the left end of the CL..." → Revised MS: "... , which determines the μ_{Li} applied to the right end of the CL..." in line 4-5 of p.15

Comment 2-2 (Cell convention, figure design):

Figure 2a. the sign for SE is missing in the top diagram.

Response:

Thank you very much for pointing out the typographical errors in the figures and providing us with the opportunity to correct them. We have made the necessary amendments to the areas you mentioned. Additionally, we have thoroughly reviewed all other figures and made corrections wherever typographical errors were found.

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have added the sign for SE in Figure 2a.
- In Fig. 4b, the value that should have been listed as "20 μm " was incorrectly written as "10 μm ". We have corrected the relevant part of the figure. Accordingly, we have also fixed similar typographical errors in the main text and Figure caption. The relevant revised portions can be found:

* Original MS: "... 100, 50, and 10 μm ..." → Revised MS: "... 100, 50, and 20 μm ..." in line 13 of p.11

* Original MS: "... 100, 50, and 10 μm ..." → Revised MS: "... 100, 50, and 20 μm ..." in line 3 of p.12

- We have corrected the typographical errors in the caption of Figure 4. The relevant revised portions can be found:

* Original MS: "... , maintain the thickness..." → Revised MS: "... , maintain^{ing} the thickness..." in line 3 of p.12

* Original MS: "... where the thickness of the SE and CL are set at 100 μm and 10 nm, respectively, with σ_{Li^+} set at 0.5×10^{-3} S/cm for the SE and 1.0×10^{-6} S/cm for the CL..." → Revised MS: "... where the σ_{Li^+} is set at 0.5×10^{-3} S/cm for the SE and 1.0×10^{-6} S/cm for the CL..." in line 7 of p.12

Comment 3:

The interphase formation in Figure 1a, is this due to self-decomposition of the SE because of low μ_{Li} beyond the electrochemical window of SE as in (SE-->degradation products) or does it account for the chemical reaction between cathode and SE as in (cathode+SE-->degradation products)? What are the degradation products that form the interphase?

Response:

The interphase depicted in Figure 1a was intended to represent the one formed due to the self-decomposition of the SE. To clarify this, we have added the text "Interphase formed due to oxidative self-decomposition of SE" to Figure 1a. Furthermore, in the revised manuscript, we have adjusted the wording to clearly indicate that we are referring to the self-decomposition of the SE or the decomposition occurring within the SE, wherever appropriate.

The decomposition products forming the interphase vary depending on the SE and whether the decomposition is oxidative or reductive. For example, in the case of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$, it has been reported that Li_2O_2 , La_2O_3 , and $\text{Li}_6\text{Zr}_2\text{O}_7$ are formed during oxidative decomposition¹. However, the focus of this paper is on the conditions under which oxidative or reductive decomposition of the SE can occur, and the specific products formed are beyond the scope of this work.

References in this response

1. Schwietert, T. K. *et al.* Clarifying the relationship between redox activity and electrochemical stability in solid electrolytes. *Nat. Mater.* **19**, 428–435 (2020).

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have added the text "Interphase formed due to oxidative self-decomposition of SE" to Figure 1a.
- We have adjusted the wording to clearly indicate that we are referring to the self-decomposition of the SE or the decomposition occurring within the SE, wherever appropriate. The relevant revised portions can be found:

* Original MS: "... oxidative or reductive decomposition of the SE..." → Revised MS: "... oxidative or reductive self-

decomposition of the SE...” in line 20 of p.2

* Original MS: “... the respective regions...” → Revised MS: “... the corresponding regions in the SE...” in line 31 of p.2

* Original MS: “...leading to the decomposition...” → Revised MS: “... leading to the self-decomposition...” in line 41 of p.2

Comment 4:

Before contact between cathode and SE, the μ_{Li} in each phase is constant throughout the materials (horizontal lines at different μ_{Li} in each phase). But after contact μ_{Li} in cathode is still horizontal but in SE phase there is a slope. Why is μ_{Li} horizontal in cathode phase but sloped in SE phase? Shouldn't there be either a slope in both phases or in neither phase? Or, is μ_{Li} simply assumed to be constant in cathode as a boundary condition?

Response:

First, regarding the gradient of μ_{Li} within the SE, the SE is in contact with the anode that has a different μ_{Li} on the opposite side. Consequently, the μ_{Li} must vary spatially within the SE, which is represented by the gradient in the Figure 1. As for the distribution of μ_{Li} within the cathode, in this work, the focus is on the μ_{Li} at the SE and cathode interface. Therefore, for simplicity, the figure was intended to depict a situation where the μ_{Li} distribution is uniform within the cathode, such as after very low-rate charging. In real batteries, an inhomogeneous μ_{Li} distribution may also occur within the cathode¹. However, as you correctly pointed out, the distribution of μ_{Li} within the cathode is not particularly important for this work, as the μ_{Li} at the SE and cathode interface is the primary focus. To emphasize the importance of μ_{Li} at the SE and cathode interface, we have revised the figure by adding shading to the line representing the μ_{Li} distribution within the cathode, clearly highlighting the μ_{Li} at the interface.

References in this response

1. Kimura, Y. *et al.* 3D Operando Imaging and Quantification of Inhomogeneous Electrochemical Reactions in Composite Battery Electrodes. *J. Phys. Chem. Lett.* **11**, 3629–3636 (2020).

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have revised the Fig. 1 by adding shading to the line representing the μ_{Li} distribution within the cathode, clearly highlighting the μ_{Li} at the interface.

Comment 5:

During cycling, there is a lithiation gradient in the cathode. How does this influence the SE stability?
Is only the μ_{Li} at the surface of the cathode relevant?

Response:

As the reviewer correctly pointed out, the μ_{Li} at the interface between the cathode active material (CAM) and the SE is crucial. In this paper, we primarily focus on the steady-state cathode/SE interface in a one-dimensional SSB model under open-circuit conditions as a model case. This is because the main objective of this paper is to demonstrate that the protective capability of CLs is not solely determined by their intrinsic characteristics and chemical compatibility with SEs and AMs, but is also governed by the μ_{Li} distribution within the SE and CL. Therefore, the Li concentration gradient formed within the cathode during cycling is beyond the scope of this paper. However, as you mentioned, during cycling, the μ_{Li} distribution within the cathode can become inhomogeneous. In this case, the complexity arises from the SE present within the composite electrode. The stability of the SE within the composite electrode should vary depending on the μ_{Li} of the CAM particles which is in contact with the SE. To rigorously consider this, it is necessary to understand the three-dimensional spatial arrangement of each CAM particle and SE particle, as well as the state of charge (SOC) of each CAM particle, which requires more complex and extensive calculations. This is the next step beyond the scope of this paper. Nevertheless, even in such cases, the fundamental concepts presented in this paper still hold true, and by extending equations (1), (2), (8), and (9) to three dimensions, the μ_{Li} distribution in the SE particles within the composite electrode can be calculated under appropriate boundary conditions.

Comment 6:

In Figure 2c, why is there no case in which there is a step of μ_{Li} at the cathode then a horizontal line of μ_{Li} through the SE and then another step of μ_{Li} at the anode?

Response:

The μ_{Li} distribution that the reviewer has pointed out may indeed occur within a relatively short period after the SE is brought into contact with the anode and cathode. However, assuming that the SE has some degree of electronic conductivity, as assumed in this study, such a μ_{Li} distribution cannot be achieved in the steady state. We believe these assumptions are valid in actual SSBs since many SEs indeed have non-negligible electronic conductivity. We thus believe that the μ_{Li} distribution shown in Fig. 2c exists within the SE in the SSB, at least after a sufficient amount of time has passed in the open circuit condition. Although this will be discussed in detail in a separate paper, it can be demonstrated that for an SE with the electronic conductivity and thickness assumed in this paper, the steady-state μ_{Li} distribution shown in Fig. 2 can be achieved within a realistic time frame (ranging from a few hours to a few days).

Comment 7:

In the reaction of (cathode + SE --> degradation products) there are many complex reaction paths and degradation products that determine the thermodynamic and kinetic stability of the interface 'cathode + SE'. What about oxidation of other elements in the SE by the cathode (e.g. in thiophosphate solid electrolytes, oxidation of sulfur and phosphorus are often observed)? Is this also dependent of μ_{Li} ?

Response:

The focus of this paper is on the self-decomposition of the SE due to oxidation or reduction induced by the variation in the cathode or anode electrode potentials. Therefore, the reactions the reviewer have mentioned are beyond the scope of this work. Even if the cathode and SE are chemically stable and the reactions the reviewer have pointed out do not occur, the SE may still become thermodynamically unstable. The key question of this paper is to determine the design guidelines for the coating layer to prevent such instability. To clarify this point, in the revised manuscript, we have explicitly stated that we are focusing on the self-decomposition of the SE and not considering chemical reactions with the cathode. Furthermore, we have revised expressions similar to "stability of the interface" found in the original text to "stability of the SE at the interface" or similar phrases, because the expression "stability of the interface" can include changes in the cathode at the interface, which is not our intention.

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have explicitly stated that we are focusing on the self-decomposition of the SE and not considering chemical reactions with the cathode on lines 8-10 of p.8 in the revised manuscript:
"Throughout this work, we assume that the CL is chemically stable with both the CAM and SE; therefore, we do not consider decomposition reactions other than the oxidative self-decomposition of the SE, such as chemical reactions with the cathode."
- We have revised expressions similar to "stability of the interface" found in the original text to "stability of the SE at the interface" or similar phrases, because the expression "stability of the interface" can include changes in the cathode at the interface, which is not our intention. The relevant revised portions can be found:
 - * Original MS: "...ensure interfacial stability..." → Revised MS: "...ensure the thermodynamic stability of the SE at the interfaces..." in line 14-15 of p.1
 - * Original MS: "To thermodynamically protect the interface..." → Revised MS: "To thermodynamically protect the SE..." in line 16 of p.1
 - * Original MS: "...ensure interfacial thermodynamic stability..." → Revised MS: "...ensure the thermodynamic stability of the SE..." in line 22 of p.1
 - * Original MS: "... the chemical and electrochemical instability..." → Revised MS: "... the chemical and electrochemical instability of the SE at the anode and cathode interfaces..." in line 11-12 of p.2
 - * Original MS: "... stability of interfaces..." → Revised MS: "... stability of the SE at the anode and cathode interfaces..." in

line 21-22 of p.2

* Original MS: “The chemical and electrochemical stability of the interface...” → Revised MS: “The chemical and electrochemical stability of the SE at the anode and cathode interfaces...” in line 23-24 of p.2

* Original MS: “... stability of the interface...” → Revised MS: “... the SE at the anode and cathode interfaces...” in line 26-27 of p.2

* Original MS: “... from a thermodynamic stability perspective...” → Revised MS: “... from the perspective of its ability to thermodynamically protect the SE...” in line 13 of p.3

* Original MS: “... for thermodynamically protective CLs...” → Revised MS: “... of the CL necessary for thermodynamically protecting the SE...” in line 3-4 of p.4

* Original MS: “... electrochemical window...” → Revised MS: “...electrochemical window of the SE...” in line 9 of p.19

* Original MS: “... to protect the interface...” → Revised MS: “... to protect the SE at the interface...” in line 11 of p.19

Comment 8:

A possible degradation mechanism is the oxygen release of cathodes. So, formed oxygen gas may also react with the SE as in ($O_2 + SE \rightarrow$ degradation products). How does this affect μ_{Li} and the requirements of the CL?

Response:

We agree that the reaction between oxygen released from the cathode and the SE is an important issue. However, as mentioned in the response to comment 7, the focus of this paper is on the self-decomposition of the SE due to oxidation or reduction induced by the variation in the cathode or anode electrode potentials. Therefore, the reaction the reviewer has pointed out is beyond the scope of this paper.

Changes (line and page numbers are those of the revised manuscript, if not specified):

- The revisions made in response to this comment are the same as those described in the response to Comment 7.

Comment 9:

Page 8 line 231 ‘...the μ_{Li} at the interface surpassed the oxidation limit of the electrochemical window, indicating that the CL can inhibit the oxidative decomposition of the SE.’ The oxidation limit appears to be at around -3.6 eV if I read the figures correctly: is this a fixed value and how was it obtained? What is the oxidation limit of other SE?

Response:

We referred to the reported oxidation limit of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZ) (3.6 V vs. Li^+/Li) in the literature¹, where the oxidation limit of LLZ was determined using first-principles calculations. This was mentioned in the beginning of the Results and Discussion section of the original manuscript, citing the relevant literature (lines 3-4 in p.8 in the original manuscript, "For the SE, we select a garnet-type SE ($\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$), assuming its σ_{Li^+} and σ_{ele} to be 0.5×10^{-3} S/cm and 1.0×10^{-11} S/cm, respectively, with its oxidation potential limit of 3.6 V vs. Li^+/Li ."). In this work, we discuss the conditions required for the CL to thermodynamically stabilize the SE, under the given oxidation or reduction limit of a SE. Therefore, we do not delve into the details of the oxidation and reduction limits of other SEs, which have been extensively investigated through experiments and first-principles calculations¹⁻⁴.

However, we did not explicitly mention the corresponding μ_{Li} value for the oxidation limit of 3.6 V vs. Li^+/Li , as the reviewer pointed out. Therefore, in the revised manuscript, we have clearly stated that the μ_{Li} corresponding to the oxidation limit is -3.6 eV.

References in this response

1. Schwietert, T. K., Vasileiadis, A. & Wagemaker, M. First-Principles Prediction of the Electrochemical Stability and Reaction Mechanisms of Solid-State Electrolytes. *JACS Au* **1**, 1488–1496 (2021).
2. Hassoun, J. et al. A structural, spectroscopic and electrochemical study of a lithium ion conducting $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ solid electrolyte. *J. Power Sources* **229**, 117–122 (2013).
3. Zhu, Y., He, X. & Mo, Y. Origin of Outstanding Stability in the Lithium Solid Electrolyte Materials: Insights from Thermodynamic Analyses Based on First-Principles Calculations. *ACS Appl. Mater. Interfaces* **7**, 23685–23693 (2015).
4. Richards, W. D., Miara, L. J., Wang, Y., Kim, J. C. & Ceder, G. Interface Stability in Solid-State Batteries. *Chem. Mater.* **28**, 266–273 (2016).

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have clearly stated that the μ_{Li} corresponding to the oxidation limit is -3.6 eV on line 4 of p.8: "... , which corresponds to μ_{Li} of -3.6 eV".

Comment 10 (Additional suggestion):

Could the authors provide a calculation tool (matlab, excel, origin, or so) as supplementary material that can be freely downloaded and modified (ionic, electronic, geometric values etc) by anyone, so that the calculations can be readily adapted to other materials systems and parameters?

Response:

We agree with the reviewer's suggestion. We will provide the code used to calculate the μ_{Li} distribution within the SE and CL as part of the Supplementary Information.

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We will provide the code used to calculate the μ_{Li} distribution within the SE and CL as part of the Supplementary Information.
- We have explicitly stated that the relevant code is included in the Supplementary Information on line 28-29 of 19:

“Code availability

The computational code to calculate the μ_{Li} distribution within the SE and CL, written in the Julia programming language and running on a Jupyter Notebook, is provided in the Supplementary Information.”.

Dear Reviewer 2

We are truly thankful for your time and effort in offering us your valuable feedback that strengthens our discussion. We have carefully considered your concerns and attempted to address them. We hope our responses meet your expectations.

Comment 1:

Mixed ionic and electronic conducting interlayers are crucial for achieving quasi-stable interphases for operations as related in the literature (Adv. Sci. 2023, 10, 2302263, Energy Mater Adv. 2021;2021:DOI:10.34133/2021/1519569, etc.). Can the authors relate their insight into design of MIECs specifically – they touch base on ionic and electronic conductivity separately, but how does the combination play out?

Response:

Thank you for your valuable comment. We have read the papers the reviewer suggested, and we agree that mixed ionic and electronic conducting (MIEC) interlayers are indeed important for efficient and stable Li deposition/dissolution. However, it appears that the role of MIEC interlayers in the papers is to enable efficient and mechanically-electrochemically stable Li deposition/dissolution rather than protecting the solid electrolyte (SE) from reductive decomposition due to contact with Li. If the role of MIEC interlayers were the latter, the insights obtained in this study would be directly applicable, but in the former case, it would be beyond the scope of this paper.

Regarding the combination of ionic and electronic conductivities, our study showed that the ability of the coating layer (CL) to thermodynamically protect the SE depends on the electronic conductivities of the CL and SE but is almost independent of their ionic conductivities. This is because, in a situation where the ionic conductivities of the CL and SE are significantly high compared to their electronic conductivities, the Li chemical potential (μ_{Li}) distribution within the CL and SE is primarily determined by their electronic conductivities. If the ionic and electronic conductivities of the SE and CL were comparable, their combination would influence the μ_{Li} distribution within the CL and SE and, consequently, the ability of the CL to thermodynamically protect the SE. However, materials with comparable ionic and electronic conductivities cannot be used as SEs. Furthermore, as demonstrated in this study, the CL is required to have a lower electronic conductivity than the SE to thermodynamically protect it. Using a material with an extremely low ionic conductivity comparable to its electronic conductivity as the CL would result in a high battery resistance, making it an unrealistic choice. Therefore, in practical SSBs, the influence of the combination of ionic and electronic conductivities on the thermodynamic protective capability of the CL appears to be negligible.

Comment 2:

Having a table of typical coating layer materials with reported ionic, electronic and thickness values from the literature might be a good reference point.

Response:

Thank you for your suggestion. The ionic conductivity of CLs has been well summarized in previous literature (for example, Y. Xiao *et al.*, *Joule* **3**, 1252–1275 (2019)). However, to the best of our knowledge, there is currently a scarcity of data on the electronic conductivity of CLs. Furthermore, although we performed calculations under the assumption of a constant electronic conductivity in this work, in reality, the electronic conductivity of the CL can vary depend on the μ_{Li} or Li activity to which the CL is exposed. Therefore, ideally, the electronic conductivity of the CL should be measured while specifying the Li activity to which the CL is exposed. However, to the best of our knowledge, no such reports currently exist. We are thus hoping that this work will inspire an increase in the measurement data of electronic conductivity of CLs under specified Li activities. Due to these circumstances, we feel that currently, there is insufficient data on CLs to make a table which serves as a reference point.

Comment 3:

It is understandable that the chemical stability is influenced by the thickness of the coating layer. However, why does the stability of the coating layer depend on the thickness of the solid electrolyte? Given that they are thermodynamic calculations – shouldn't the chemical stability of the interface be independent of the thickness of the SE itself.

Response:

Thank you for highlighting a crucial point in this paper. To be precise, it is not the stability of the CL that depends on the thickness of the SE, but rather the ability of a CL with a given electronic conductivity and thickness to thermodynamically protect the SE. Whether a CL can thermodynamically protect the SE is determined by whether the μ_{Li} at the CL/SE interface falls within the range where the SE can exist stably. As mentioned in this paper, the μ_{Li} at the CL/SE interface is determined by the overall μ_{Li} distribution formed within the CL and SE. The μ_{Li} distribution formed within the CL and SE is significantly influenced not only by the electronic conductivity and thickness of the CL, but also by those of the SE, and even by the μ_{Li} at the cathode and anode interfaces. Consequently, whether a CL can thermodynamically protect the SE depends not only on the thickness of the CL but also on the thickness of the SE, as well as the electronic conductivities of the CL and SE and the μ_{Li} of the anode and cathode interfaces.

To clarify further, the calculation of the overall μ_{Li} distribution within the SE and CL is not strictly a thermodynamic calculation; rather, it should be considered a calculation in a non-equilibrium

steady state. This is because, in the composite system consisting of the anode, SE, CL, and cathode under investigation in this study, the μ_{Li} does not become uniform throughout the entire composite system. Therefore, the entire composite system cannot be considered to be in a thermodynamic equilibrium state. Consequently, it is reasonable to consider the composite system (after long open-circuit holding) to be in a non-equilibrium steady state, where local equilibrium is established throughout the system, and a macroscopically constant state is maintained over time. Even so, because local equilibrium is established, it is possible to define μ_{Li} in local regions, such as at the SE/CL interface, and to discuss the thermodynamic state at that location. Due to these circumstances, to answer the reviewer's question more abstractly, the thermodynamic state in local regions, such as at the SE/CL interface, depends on the non-equilibrium steady state of the entire composite system. Therefore, the thickness of the SE, which is one of the factors determining the non-equilibrium steady state, influences the thermodynamic stability of the SE/CL interface.

We believe that obtaining these non-obvious results is one of the novel aspects of this study. In the revised manuscript, we have revised the text to emphasize this point.

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have revised the text to emphasize the aforementioned point.

Original MS: “Nevertheless, the required thickness of a CL to protect an SE varies significantly depending on the thickness of an SE.” → Revised MS: “It is important to emphasize, however, that the required thickness of a CL to protect an SE varies significantly depending on the thickness of an SE..” in line 11-12 of p.11

Comment 4:

The authors suggest increasing the thickness of the CL when the electronic conductivity is higher. Can they provide a justification for reduced ion transport across that CL in case of thick CL layers? Is there a maximum CL beyond which the ionic conduction drop would be so much, that the benefit of improving chemical stability might not justify the CLs?

Response:

Thank you again for highlighting a critical point in this paper. The issue you have raised is precisely the content we wanted to convey in the section "Effect of σ_{Li+} of a CL on its Protective Capability and the Battery Performance." Increasing the thickness of the CL can lead to a significant internal resistance of the battery due to the low ionic conductivity of the CL while a thicker CL is favorable in terms of protecting the SE. Therefore, it is necessary to optimize the thickness of the CL to keep the increase in the internal resistance of the battery within an acceptable range while ensuring the stability of the interface. In the aforementioned section, we quantitatively calculate the conditions required for the CL to maintain the μ_{Li} at the CL/SE interface within the thermodynamically stable range of the SE

while suppressing the increase in the total resistance caused by the increased thickness of the CL within the desired value. To further emphasize this point, we have added more specific calculation examples in Fig. 5d. This revision also serves as a response to Reviewer 3's comment 2.

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have added specific calculation examples of acceptable absolute value range for the electronic conductivity, ionic conductivity, and thickness of the CL required to ensure the thermodynamic stability of the SE while limiting the increase in total ohmic resistance in Fig. 5d.
- We have added a discussion regarding Fig. 5d to the revised manuscript on line 29-41 of p.13 and line 1-3 of p.14: “As a practical example, we calculated the acceptable absolute value range for the σ_{ele} , σ_{Li^+} , and thickness of the CL required to ensure the thermodynamic stability of the SE while limiting the increase in total ohmic resistance to less than 10% compared to the resistance of the SE alone, considering two scenarios with different SE (Fig. 5d). In the first scenario, LLZ is assumed as the SE with σ_{Li^+} and σ_{ele} of $0.5 \times 10^{-3} \text{ S/cm}^{46}$ and $1.0 \times 10^{-11} \text{ S/cm}^{39}$, respectively, and an oxidation limit of 3.6 V vs. $\text{Li}^+/\text{Li}^{20}$ (green). The second scenario assumes LPSC as the SE with σ_{Li^+} and σ_{ele} of $2.4 \times 10^{-3} \text{ S/cm}^{49}$ and $5.2 \times 10^{-8} \text{ S/cm}^{48}$, respectively, and an oxidation limit of 2.2 V vs. $\text{Li}^+/\text{Li}^{20}$ (blue). In both cases, the thickness of the SE is set to 100 μm . As shown in this figure, the scenario assuming an LLZ as the SE nominally has a slightly wider acceptable range for the σ_{Li^+} of the CL. This is because LLZ has a lower σ_{Li^+} than LPSC, allowing for a larger increase in resistance before reaching a 10% increase compared to the resistance of the SE alone. On the other hand, regarding the acceptable range for the σ_{ele} of the CL, the scenario assuming LPSC as the SE exhibits a wider range as discussed in Fig. 3d, despite the considerably lower oxidation limit (2.2 V vs. Li^+/Li). This example demonstrates that a narrow electrochemical window of the SE does not necessarily render it unusable. A relatively high σ_{ele} of the SE may expand the range of options for the CL, allowing for the selection of an appropriate CL that can sufficiently stabilize the SE thermodynamically. As described above, the protective capability of the CL depends not only on its intrinsic properties and geometry but also on those of the SE. Therefore, when designing a CL, it is crucial to consider the relative values of the three essential parameters (σ_{ele} , σ_{Li^+} , and thickness) of the CL with respect to those of the SE to be used, as well as the interdependence among these parameters.”

Comment 5:

The premise of coating SEs, opposed to the CAMs is interesting postulate. Are there any literature experimental studies that showcase that? If so, can you reference them. If not, can you provide some pathways to design these?

Response:

To the best of our knowledge, there have been no reports showing directly that coating SEs instead of CAMs improves the performance of composite electrodes. However, atomic layer deposition (ALD) technique can be considered as a promising method for coating SEs. This technique has been applied

to SEs such as $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$, and it has been reported to improve the stability of the interface with electrodes (Y. Ren *et al.*, *J. Electrochem. Soc.* **169**, 040529 (2022)). Furthermore, Z. D. Hood *et al.* reported that coating $\text{Li}_6\text{PS}_5\text{Cl}$ powder with Al_2O_3 using ALD enhances cycling performance (Z. D. Hood *et al.*, *Adv. Mater.* **35**, 2300673 (2023)). We have cited these references in the main text.

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have cited the literature that reports the coating of SEs using atomic layer deposition on line 37-38 of p.17: “..., using a technique such as the atomic layer deposition^{52,53}.”
- We have cited following literature in the revised manuscript.
 - *52. Ren, Y. & Wachsman, E. D. *J. Electrochem. Soc.* **169**, 040529 (2022).
 - *53. Hood, Z. D. *et al. Adv. Mater.* **35**, e2300673 (2023).

Dear Reviewer 3

We are sincerely grateful for the time and effort you have invested in providing us with your valuable comments that enrich our discussion. We have made a sincere attempt to address your concerns. We hope you find these answers meet your expectations.

Comment 1:

The conditions for barrier coating found in this work is not different from what has been established in the field. Given the solid electrolyte will see full electrochemical potential of the cathode only by having ionic (Li^+) and electronic (e^-) percolation, improving electrochemical stability by making the interfaces insulating is obvious. It actually has been covered in many papers, such as Xiao et al., *Nat. Rev. Mater.* 5, 105-126 (2020) and Zhu et al., *ACS Appl. Mater. Inter.*, 7, 23685-23693 (2015).

Response:

Thank you for your comment. We were aware that the need to reduce electronic conductivity (σ_{ele}) of the coating layer (CL) has been mentioned in the papers the reviewer cited. However, that is not the most important point of this work. Until now, there has been no method proposed to specifically calculate the required σ_{ele} or the degree of insulation necessary for a CL to thermodynamically protect the solid electrolyte (SE). In contrast, this work proposes a method to quantitatively calculate the required σ_{ele} of the CL for thermodynamically protecting the SE. Furthermore, we demonstrated that the required σ_{ele} of the CL is heavily dependent on the thickness of the CL, the σ_{ele} and thickness of the SE, and even on both the cathode and anode electrode potentials. This is because the local thermodynamic stability at the SE/CL interface is determined by the overall μ_{Li} distribution formed across the entire SE and CL. Those insights and findings have not been asserted in previous papers. Therefore, we believe that our paper makes a valuable contribution to advancing the understanding of the thermodynamic protective capability of CLs, and the insights and findings presented in this work offer sufficient novelty and significance to merit publication in *Commun. Mater.*

Changes (line and page numbers are those of the revised manuscript, if not specified):

- To emphasize that the necessity of reducing the electronic conductivity of CLs has been mentioned in previous literature, we have made the following changes to the manuscript, citing the papers the reviewer introduced:

Original MS: "Therefore, it is essential for CLs to possess low σ_{ele} , not solely to kinetically inhibit the decomposition of an SE, but also from a thermodynamic perspective." → Revised MS: "Therefore, it is essential for CLs to possess low σ_{ele} as pointed out in previous literature^{14,22}, not solely to kinetically inhibit the decomposition of an SE, but also from a thermodynamic perspective." in line 37-38 of p.8

Comment 2:

The manuscript used fixed modeling parameters (ionic and electronic conductivities) from known materials. As the calculation framework is set, it would be more useful and insightful to compare different systems to understand the optimum coatings. For instance, determine conditions required for the barrier coating for systems involving LLZO against NCM and Argyrodite against NCM.

Response:

Thank you very much for your highly informative suggestions. We agree that it is insightful to perform calculations for different systems. In the revised manuscript, we have included calculation results for the case of using argyrodite as an SE in Fig. 3d and Fig. 5d. These results highlight that the required characteristics of the CL vary significantly depending on the characteristics of the SE, and that even SEs like argyrodites with a narrow electrochemical window can be thermodynamically safeguarded by introducing a CL if the electronic conductivity of the SE is relatively high. We believe that these additional discussions, prompted by the reviewer's suggestion, have greatly contributed to enhancing the value of our paper.

Changes (line and page numbers are those of the revised manuscript, if not specified):

- We have included calculation results for the case of using argyrodite as an SE in Fig. 3d and Fig. 5d.
- To accompany the addition of the above figures, we have included the following discussion in the revised manuscript:

**For example, $\text{Li}_6\text{PS}_5\text{Cl}$ (LPSC), an argyrodite, is known to have a relatively high σ_{ele} of $5.2 \times 10^{-8} \text{ S/cm}^{48}$. Fig. 3d shows the μ_{Li} distribution within the SE and CL, assuming the use of LPSC as the SE and setting its σ_{ele} to $5.2 \times 10^{-8} \text{ S/cm}$ while keeping the other parameters the same as in Figs. 3a,b. Although LPSC is known to have a very low oxidation limit of approximately 2.2 V vs. $\text{Li}^+/\text{Li}^{20}$, even the CL with relatively high σ_{ele} of $1.0 \times 10^{-12} \text{ S/cm}$ can sufficiently maintain the μ_{Li} at the interface within the range where the SE is thermodynamically stable.” on lines 12-16 of p.9.

**As a practical example, we calculated the acceptable absolute value range for the σ_{ele} , σ_{Li^+} , and thickness of the CL required to ensure the thermodynamic stability of the SE while limiting the increase in total ohmic resistance to less than 10% compared to the resistance of the SE alone, considering two scenarios with different SE (Fig. 5d). In the first scenario, LLZ is assumed as the SE with σ_{Li^+} and σ_{ele} of $0.5 \times 10^{-3} \text{ S/cm}^{46}$ and $1.0 \times 10^{-11} \text{ S/cm}^{39}$, respectively, and an oxidation limit of 3.6 V vs. $\text{Li}^+/\text{Li}^{20}$ (green). The second scenario assumes LPSC as the SE with σ_{Li^+} and σ_{ele} of $2.4 \times 10^{-3} \text{ S/cm}^{49}$ and $5.2 \times 10^{-8} \text{ S/cm}^{48}$, respectively, and an oxidation limit of 2.2 V vs. $\text{Li}^+/\text{Li}^{20}$ (blue). In both cases, the thickness of the SE is set to 100 μm . As shown in this figure, the scenario assuming an LLZ as the SE nominally has a slightly wider acceptable range for the σ_{Li^+} of the CL. This is because LLZ has a lower σ_{Li^+} than LPSC, allowing for a larger increase in resistance before reaching a 10% increase compared to the resistance of the SE alone. On the other hand, regarding the acceptable range for the σ_{ele} of the CL, the scenario assuming LPSC as the SE exhibits a wider range as discussed in Fig. 3d, despite the considerably lower oxidation limit (2.2 V vs. Li^+/Li). This example demonstrates that a narrow electrochemical window of the SE does not necessarily render

it unusable. A relatively high σ_{ele} of the SE may expand the range of options for the CL, allowing for the selection of an appropriate CL that can sufficiently stabilize the SE thermodynamically. As described above, the protective capability of the CL depends not only on its intrinsic properties and geometry but also on those of the SE. Therefore, when designing a CL, it is crucial to consider the relative values of the three essential parameters (σ_{ele} , σ_{Li^+} , and thickness) of the CL with respect to those of the SE to be used, as well as the interdependence among these parameters.” on lines 29-41 of p.13 and lines 1-3 of p.14.

- To accommodate the addition of Fig. 3d and Fig. 5d, we have modified the figure captions of Fig.3 and Fig.5:

* Original MS: “...the CL to SE. The thicknesses of the SE and CL are set to 100 μm and 10 nm thickness, respectively, with σ_{Li^+} set at 0.5×10^{-3} S/cm for the SE and 1.0×10^{-6} S/cm for the CL.”

→ Revised MS: “...the CL to SE. In Fig. 3 (a)-(c), the oxidation limit of the SE was set to 3.6 V vs. Li^+/Li . (d) μ_{Li} distribution within the SE and CL when the σ_{ele} of the CL was set to 1.0×10^{-11} , 10^{-12} , 10^{-13} , and 10^{-14} S/cm while the σ_{ele} of the SE was set to 5.2×10^{-8} S/cm and the oxidation limit of the SE was set to 2.2 V vs. Li^+/Li . In all figures, the σ_{Li^+} was set at 0.5×10^{-3} S/cm for the SE and 1.0×10^{-6} S/cm for the CL, and the thicknesses of the SE and CL were set to 100 μm and 10 nm, respectively.” on lines 4-7 of p.10

* Original MS: “...of the SE alone.”

→ Revised MS: “...SE alone. (d) Acceptable absolute value range for the σ_{ele} , σ_{Li^+} , and thickness of the CL required to ensure the thermodynamic stability of the SE while limiting the increase in total ohmic resistance to less than 10% compared to the resistance of the SE alone. Two scenarios are considered: (1) LLZ is assumed as the SE with σ_{Li^+} and σ_{ele} of 0.5×10^{-3} S/cm and 1.0×10^{-11} S/cm, respectively, and an oxidation limit of 3.6 V vs. Li^+/Li (green); (2) LPSC is assumed as the SE with σ_{Li^+} and σ_{ele} of 2.4×10^{-3} S/cm and 5.2×10^{-8} S/cm, respectively, and an oxidation limit of 2.2 V vs. Li^+/Li (blue)” on lines 9-13 of p.14

- We have included following literature to refer ionic and electronic conductivity of argyrodites.

*48. Deiseroth, H.-J. *et al.* *Z. Anorg. Allg. Chem.* **637**, 1287–1294 (2011).

*49. Zhou, L. *et al.* *ACS Energy Lett.* **4**, 265–270 (2019).

Comment 3:

Above analysis may lead to a few candidate materials for coating.

Response:

As we mentioned in the response to comment 2, by analyzing different systems, we were able to incorporate important insights into the paper. We would like to express our gratitude once again for your valuable suggestion.

Changes:

The modifications made in response to Comment 3 are the same as those described in our response to Comment 2.

Thank you once again for your comments and suggestions. We worked hard to be responsive to them and we strongly hope that these answers help to solve your concerns.

Sincerely,

Yuta Kimura

Assistant professor

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yuta.kimura.b2@tohoku.ac.jp

26th Jun 24

Dear Dr Kimura,

Thank you for submitting your manuscript, "Coating Layer Design Principles Considering Li Chemical Potential Distribution within Solid Electrolytes in Solid-State Batteries", to Communications Materials. It has now been seen by 3 referees, whose comments are appended below. For the most part all of the reviewer concerns from the previous iteration have been addressed. However, Reviewer 1 has one last minor comment. We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type 'Revised Manuscript - Marked Up' when uploading the manuscript file to our online system.

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We hope to receive your revised paper within one month; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we will close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

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,

Andrew Westover

Guest Editor

Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Thank you for the detailed and clear explanations and changes to the manuscript. Well revised! As the study describes open-circuit conditions as a model case, I have one additional question as to whether the same protection principles apply during current flow. It seems that the protection capability depends strongly on the electronic conductivity difference of coating layer and SE and on electrons having to pass this layer, which is the case in open-circuit conditions. But, when the external circuit is closed during current flow, electrons may simply flow around the protective coating (i.e. through the electronic circuit, the anode and the SE). Does this influence the chemical potential profile of neutral lithium across the cell, thus negatively impacting the protection capability of the coating? A note on what is expected during current flow should be added to the manuscript.

Reviewer #2 (Remarks to the Author):

The reviewer appreciates the responses from the authors on the comments. The reviewer has no further questions.

Reviewer #3 (Remarks to the Author):

The authors have addressed my comments sufficiently. Recommended for publication.

Response to the comment received on 26th June, 2024

Dear Reviewer 1

We would like to express our sincere gratitude for your interest in our manuscript and for engaging in the discussion with us. Your comments have truly enriched our understanding of this study. To be quite honest, the point raised in your comment is the very same one that have piqued our curiosity. In fact, we have already discussed this topic to some extent in our original manuscript and within the Supplementary Information. Please allow us to elaborate on this point and provide you with a more comprehensive explanation here.

Comment:

Thank you for the detailed and clear explanations and changes to the manuscript. Well revised! As the study describes open-circuit conditions as a model case, I have one additional question as to whether the same protection principles apply during current flow. It seems that the protection capability depends strongly on the electronic conductivity difference of coating layer and SE and on electrons having to pass this layer, which is the case in open-circuit conditions. But, when the external circuit is closed during current flow, electrons may simply flow around the protective coating (i.e. through the electronic circuit, the anode and the SE). Does this influence the chemical potential profile of neutral lithium across the cell, thus negatively impacting the protection capability of the coating? A note on what is expected during current flow should be added to the manuscript.

Response:

We are grateful for your comment and are delighted to learn that you are satisfied with the revisions we have made!

Once again, we would like to express our gratitude for pointing out the very important point. As mentioned at the beginning, the variation of the neutral lithium chemical potential (μ_{Li}) distribution within the solid electrolyte (SE) and coating layer (CL) under an external current was a point of great interest to us as well. Consequently, we had discussed this topic in our original manuscript and the Supplementary Information.

Unfortunately, the Wagner theory, which we employed for the calculation of the μ_{Li} distribution, cannot be strictly applied to non-steady-state conditions, *i.e.*, during charging and discharging. Despite this limitation, we utilized the aforementioned theory

as a first-order approximation to calculate the μ_{Li} distribution under an external current (p. 15, lines 26-36 of the previously submitted manuscript, and Supplementary Note 1). We speculate that these calculation results are somewhat close to the situation during charging and discharging at least at extremely low rates.

As you have pointed out, the amount of ionic and electronic currents flowing through the SE and CL can vary depending on an external current. Such changes can be accounted for in our calculation framework as the r parameter (Eq. (11) in the manuscript). Although it is not straightforward to determine the r parameter corresponding to a given charging/discharging current, i_{tot} (Eq. (10) in the manuscript), the calculation program we previously provided in the Supplementary Information allows for the determination of an appropriate r parameter and the calculation of the μ_{Li} distribution within the SE and CL by performing an optimization calculation using the Nelder-Mead algorithm, once an external current is given as an input.

In Fig. S1 of Supplementary Note 1, we have included the μ_{Li} distribution when the charging/discharging current is set to 20 mAh/cm² and 200 mAh/cm². These charging/discharging currents correspond to 1C and 10C, respectively, when using a composite electrode with the currently reported highest cathode active material (CAM) loading (Li, Y. *et al. Science* **381**, 50–53 (2023)). As shown in Fig. S1, while the μ_{Li} distribution does indeed change when a current is applied, the change is minimal, even when such large currents are applied. Under the calculation conditions adopted in Supplementary Note 1, the change in μ_{Li} at the SE/CL interface is only about 0.2 eV, even when a current of 200 mAh/cm² is applied. Furthermore, in the actual charging and discharging of solid-state batteries (SSBs), electrode overpotentials restrict the μ_{Li} at the CAM/CL interface to values equal to or higher than that under open-circuit conditions. Therefore, in practical charging and discharging, the influence of the change in the μ_{Li} distribution due to an external current can be even smaller than that shown in Fig. S1. Based on these considerations, we believe that the influence of an external current on the μ_{Li} distribution in the SE and CL, and hence on the protective capability of the CL, can be regarded as insignificant in practical SSBs.

However, as mentioned at the beginning, strictly speaking, the μ_{Li} distribution during (non-steady-state) charging/discharging cannot be calculated using the Wagner theory. Therefore, we would like to limit the discussion presented here to a small part of the main text and the Supplementary Information, as in the previously submitted manuscript. Consequently, we would greatly appreciate your understanding in allowing us to refrain from adding further note on this point within the manuscript.

We sincerely hope that our response has adequately addressed your concerns. As this work is a purely theoretical one, it is relatively straightforward for others to reproduce. Therefore, we believe that the timely publication of our findings is important. In order to reciprocate your immense efforts in reviewing our manuscript, we are committed to enhancing the timeliness of our work and thereby maximizing the impact of our work in the field. Once again, we would like to express our heartfelt gratitude for your interest in our manuscript and for providing us with your invaluable comments. It has been an absolute pleasure to have Reviewer 1 assess our work, as your insights have greatly contributed to the improvement of our paper's quality.

Sincerely,

Yuta Kimura

Assistant professor

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yuta.kimura.b2@tohoku.ac.jp

5th Jul 24

Dear Dr Kimura,

Your manuscript titled "Coating Layer Design Principles Considering Li Chemical Potential Distribution within Solid Electrolytes in Solid-State Batteries" has now been seen again by our referees, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to edit your manuscript to comply with our journal policies and formatting style in order to maximise the accessibility and therefore the impact of your work.

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Best regards,

Andrew Westover

Guest Editor

Communications Materials

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Thank you very much for carefully addressing my additional question. I recommend publication of this manuscript.