

Peer Review File

Three-dimensional reconstruction and computational analysis of a structural battery composite electrolyte



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28th Apr 23

Dear Professor Asp,

Thank you for submitting your manuscript, "3D reconstruction and computational analysis of the electrolyte for a structural battery composite", 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.

In particular, Reviewer 2 asks where the origins of the SBE surface tilt is from and questions about the SEM measurement. All reviewers requests for additional information and clarification, relating to sample preparation, model validation, and calculations.

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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- An updated <https://www.nature.com/documents/nr-editorial-policy-checklist.zip> Editorial Policy checklist, uploaded as a 'Related Manuscript File' type. This checklist is to ensure your paper complies with all relevant editorial policies. If needed, please revise your manuscript in response to these points. Please note that this form is a dynamic 'smart pdf' and must therefore be downloaded and completed in Adobe Reader. Clicking this link will download a zip file containing the pdf.

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- If applicable, a statement regarding data available with restrictions
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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.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

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,

Dr Jet-Sing Lee
Associate Editor
Communications Materials
orcid.org/0000-0002-6740-8700

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors have presented a way to reconstruct, in 3D, the pore structure of a structural battery bicontinuous electrolyte based on BPA with an EC/PC liquid electrolyte phase. This work is important not only in the context of structural batteries but can also be extended to other forms of structural gel electrolytes. The characterisation of these porous structure is oftentimes challenging but understanding it will provide insight on engineering effective porous structures for bicontinuous electrolytes. There are a few comments and questions that the authors can consider:

1. The voxel dimensions are 10 x 12.2 x 20 nm which might be larger than some of the micropores present. If that is the case, what impact would that have? In addition, the authors could also explain briefly here how the voxel dimensions were derived.
2. In line 221, did the authors consider freeze drying or using critical point drying (after solvent exchange) to obtain dried specimens?
3. As a minor note, in line 410 (methods), the authors should note that the voltage is an rms value and is against the OCV.

In summary, the article is well written and will be important in the field not only of structural batteries but battery electrolytes in general. I would recommend the publication of this article after minor revision.

Reviewer #2 (Remarks to the Author):

Based on previous studies on the performance of structural batteries using light-weight carbon fibers for the composite, the authors present here FIB/SEM to obtain three-dimensional information

of the composite material via tomography/reconstruction to gain insight into the pore structure. In addition, the authors use these data for finite element modeling, which is used to predict e.g., ion conductivity and modulus in comparison to experimental results. The study is interesting; however, it seems that the battery community so far has not yet explored the full potential of these structural batteries (regarding that limited numbers of references cited in this manuscript).

In the introduction the authors state that “FIB-SEM has not yet been applied on the SBE, which is more challenging since the SBE is a soft and non-conductive material.” Furthermore, the pores in the SBE are very small, only a few to hundreds of nanometers.” In general, the authors should add more references on state-of-the-art approaches and strategies for FIB/SEM tomography of soft materials. Nanoscale resolution e.g., on soft polymeric material has been already demonstrated (see Nanoscale, 2017). Also, it is stated, a “special SEM imaging mode is required to capture the pores”, however this “special mode” is not further discussed.

FIB/SEM tomography suffers from issues like curtaining or shine-through artefacts, which can be minimized by embedding in appropriate polymers. Why is this not applied/addressed within this study?

Regarding Figure 1d: it appears that the surface of the SBE is tilted. The authors should explain where this tilt is coming from. Shouldn't be the surface a straight line after successful alignment of the image stack (fig 1d “after registration”)? Is it really possible to align the SEM images with a reference pattern on the Pt as shown in Fig1 b? This is especially interesting, as the authors report huge problems with drift during the measurement. The authors should explain and state in the manuscript, why this procedure was chosen, as it appears not to be the best choice to do image alignments with features within the region of interest.

In addition, for all SEM images – a scale bar should be provided (e.g., 1c and d)

The authors describe that they apply a threshold to separate pore space from polymer. Can the authors comment, why a threshold value was chosen in a way that the achieved pore space matches the “theoretical porosity calculated from the liquid phase in the SBE mixture”? Thereby, the actually measured pore space was adjusted to a predicted value. Some further explanations should be added to the manuscript, why the pore space was measured if it is adjusted afterwards (“model driven science”)?

In addition, Reference 23 refers to NMR and threshold calculation (Cattaruzza, M. et al.) is only submitted for publication (2023). A manuscript submitted for publication cannot be cited. In this case, it is not clear how the pore space was calculated from NMR data. This is particularly problematic as the value was then used as threshold value.

Page 10 The information on achievable resolution and the issue of pores, which are too small to be visualized via FIB/SEM tomography should be discussed more prominently, earlier in the text.

The authors should comment on the swollen state of the polymer and what the expected differences in pore space and polymer volume would be? Are there any measurements confirming changes in volume during drying? How can the presented data be compared/connected with the possibilities for ion transport in the actual wet state when the authors expect that the pore channels may be even closed? A suggestion would be to perform measurements under cryogenic conditions and add such data to the manuscript. How useful are these simulations if the real state and the investigated state of the material is quite different?

Technical comments:

Some typos throughout the manuscript.

Regarding light-weight composite electrodes, the authors may add a few more references.

Reviewer #3 (Remarks to the Author):

Comments to the Authors:

The paper entitled “3D reconstruction and computational analysis of the electrolyte for a structural battery composite” provides a valuable contribution to the characterization of a biphasic battery electrolyte for structural battery applications. A clear description of the experimental procedure implemented to characterize the topology of the solid electrolytes is given, including details of the Fib-SEM set-up and the images post-processing. The proposed approach

Moreover, statistically equivalent representative volume elements are extracted from the 3D reconstruction of the RVE and are used in finite element simulations to predict their elastic properties and the ionic conductivity of the RVE. Both the experimental procedure and the workflow for the characterization of the structural electrolyte are valuable contributions to the study of these new types of multifunctional materials.

The reviewer suggests minor review before publishing the paper in “Communications Materials”:

- 1) Validation of SVE: A statistical analysis of the extracted 20 SVEs after meshing to demonstrate that they preserve the same channels properties and density would be important to assess the robustness of the simulations.
- 2) Including extra information about the finite element model size would help in understanding the need of hpc cluster to perform the simulations. Moreover, the authors attribute the difference in the estimation of the ionic conductivity to the fact that the simulations do not consider the increase in polymer backbone volume due to the absorption of the liquid electrolyte. Would Including this effect, with self-contact have been impractical from a computational point of view due to the excessive model complexity?
- 3) A table including the input used for the calculation of the ionic conductivity presented in Tab.2 would facilitate the understanding of the results.
- 4) Tab. presents the line of “Elastic Modulus” that is not aligned
- 5) Fig. 5 f and g quality could be improved

Based on previous studies on the performance of structural batteries using light-weight carbon fibers for the composite, the authors present here FIB/SEM to obtain three-dimensional information of the composite material via tomography/reconstruction to gain insight into the pore structure. In addition, the authors use these data for finite element modeling, which is used to predict e.g., ion conductivity and modulus in comparison to experimental results. The study is interesting; however, it seems that the battery community so far has not yet explored the full potential of these structural batteries (regarding that limited numbers of references cited in this manuscript).

In the introduction the authors state that “FIB-SEM has not yet been applied on the SBE, which is more challenging since the SBE is a soft and non-conductive material.” Furthermore, the pores in the SBE are very small, only a few to hundreds of nanometers.” In general, the authors should add more references on state-of-the-art approaches and strategies for FIB/SEM tomography of soft materials. Nanoscale resolution e.g., on soft polymeric material has been already demonstrated (see Nanoscale, 2017). Also, it is stated, a “special SEM imaging mode is required to capture the pores”, however this “special mode” is not further discussed.

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measurements under cryogenic conditions and add such data to the manuscript. How useful are these simulations if the real state and the investigated state of the material is quite different?

Technical comments:

Some typos throughout the manuscript.

Regarding light-weight composite electrodes, the authors may add a few more references.

Response letter for :

3D reconstruction and computational analysis of the electrolyte for a structural battery composite

*Shanghong Duan, Martina Cattaruzza, Vinh Tu, Robert M. Auenhammer, Ralf Jänicke, Mats K.G. Johansson, Fang Liu, Leif E. Asp**

We are grateful for the review comments provided by the editor and the reviewers. We have done our best to respond to these and revise the manuscript as requested. Below, you find detailed responses to each question raised by the reviewers. Text introduced into the manuscript is highlighted.

Thank you!

Reviewer #1

1. The voxel dimensions are 10 x 12.2 x 20 nm which might be larger than some of the micropores present. If that is the case, what impact would that have? In addition, the authors could also explain briefly here how the voxel dimensions were derived.

Reply: The voxel dimension is larger than pores less than 20 nm. The pores smaller than 1 voxel are unfortunately not identified. These small pores are most likely isolated from the large pores, otherwise they belong to the large pore. They are therefore not likely to contribute much to the ion transport. The contribution from the small and isolated pores to the overall ionic conductivity can therefore be neglected when much larger pores exist and form a connected network. In addition, a recent NMR study by Cattaruzza et al. [26] found that in the small nanochannels there are a lot of interactions between the electrolyte species and the polymer wall (specially for the solvents molecules) that hinder the molecule and ions transport. The contribution from unidentified, small, pores is therefore expected to be small. Concerning the effect of neglected pores on elastic modulus the effect is expected to be negligible.

The voxel dimension/resolution is based on image resolution (x-,y-) and FIB-milling depth (z-). The pixel size of the SEM image can go further down to 5 nm, and the milling depth can as low as 10 nm. However, due to the drifting issue of the non-conductive sample, if the milling depth is set too small, the final dimension of the volume of study will be too limited.

We added sentences on the voxel resolution in introduction part:

“..... The pixel size in the SME image can be as low as 5 nm, which determines the achievable resolution in image, whereas the resolution through the thickness is determined by the FIB-milling depth. Pores smaller than the achievable cannot be captured by the proposed FIB-SEM technique.”

and the impact from missing of some nano-scale pores in discussion part:

“ $5.43 \times 6.31 \times 1.20 \mu\text{m}^3$. The pores with size smaller than 20 nm will unfortunately not be identified. However, the impact from such small pores on the elastic properties can be neglected. In contrast, the effect of neglecting small pores on ionic conductivity predictions is more ambiguous. On the one hand, ionic

conductivity is expected to be controlled by the connected network of large pores. Also, the undetected small pores are likely to be isolated from the connected network. Cattaruzza et al. explored SBE with different porosity and found the interactions between the electrolyte species and the polymer matrix hinder the molecule and ions transport.^[26] On the other hand, if the amount of non-identified small pores is large enough, the volume of the network of connected large pores, and hence the ionic conductivity, will be overestimated”

2. In line 221, did the authors consider freeze drying or using critical point drying (after solvent exchange) to obtain dried specimens?

Reply: Thanks for the suggestion. We did not consider freeze drying in this study. Since the shrinkage is due to the extraction of the solvent from the polymer phase, it may occur after solvent exchange. However, we are planning to use a cryo-SEM for these type of studies in the future. That will allow us to skip the solvent exchange step.

3. As a minor note, in line 410 (methods), the authors should note that the voltage is an rms value and is against the OCV.

Reply: The authors do not understand this point raised by the reviewer. The subject discussed in line 410 concerns the image analysis and does not address voltage or OCV.

Reviewer #2

1. In the introduction the authors state that “FIB-SEM has not yet been applied on the SBE, which is more challenging since the SBE is a soft and non-conductive material. “Furthermore, the pores in the SBE are very small, only a few to hundreds of nanometers.” In general, the authors should add more references on state-of-the-art approaches and strategies for FIB/SEM tomography of soft materials. Nanoscale resolution e.g., on soft polymeric material has been already demonstrated (see *Nanoscale*, 2017). Also, it is stated, a “special SEM imaging mode is required to capture the pores”, however this “special mode” is not further discussed.

Reply: Thanks for comment. We agree that the *Nanoscale*, 2017 paper is an important reference and we have added it to the list of references. We have also introduced a few sentences to address the issue and also added references to additional work in the literature on soft material using FIB-SEM. We briefly state the method in *Nanoscale*, 2017, and point out the differences from our work.

The special mode refers to the electron immersion mode which explained later in the method section. To make it obvious to reader, we have replaced the “special mode” by “electron immersion mode” in introduction section.

“..... SBE is a soft and non-conductive material. These features cause issues such as curtaining, charging and low contrast of images.^[15,16,17,18,19] In general, artifacts induced by FIB milling on such specimen are more conspicuous and can be compensated for in the analysis. Furthermore, the pores in the SBE are very small, ranging from a few to hundreds of nanometers. Most previous studies investigated micro-scale pore structures. However, Neusser et al. performed FIB-SEM on soft polymer MIP at the nano-scale.^[19] To enhance contrast, the polymer structure was stained with osmium tetroxide and then filled with epoxy. In the current study, we take high resolution SEM images using the so-called electron immersion mode where electrons are caged in the imaging region. The pixel size in the SME image can be as low as 5 nm, which determines the achievable resolution in image, whereas the resolution through the thickness is determined by the FIB-milling depth. Pores smaller than the achievable cannot be captured by the proposed FIB-SEM technique.”

2. *FIB/SEM tomography suffers from issues like curtaining or shine-through artefacts, which can be minimized by embedding in appropriate polymers. Why is this not applied/addressed within this study?*

Reply: We agree with the reviewer that when analysing porous structure, FIB/SEM suffers a few pronounced artefacts. Filling the pores with epoxy has a few challenges: Firstly, it is difficult to ensure all pores are filled; Secondly, the contrast of epoxy and SBE polymer will be lower. To solve this, Neusser et al. (Nanoscale, 2017) first stain the polymer, which is for sure a brilliant idea. However, since epoxy is a different polymer system than the SBE, curtaining effect could still occur. Here, we have the option to directly capture the pores with the electron immersion mode, and then use the image processing method to treat the curtaining artifact. Therefore, we kept the polymer unstained.

To address this point, we added a short description of the method used by Neusser and co-workers in Nanoscale, 2017. (see reply to comment 1 by Reviewer 2 above).

3. *Regarding Figure 1d: it appears that the surface of the SBE is tilted. The authors should explain where this tilt is coming from. Shouldn't be the surface a straight line after successful alignment of the image stack (fig 1d 'after registration')? Is it really possible to align the SEM images with a reference pattern on the Pt as shown in Fig1 b? This is especially interesting, as the authors report huge problems with drift during the measurement. The authors should explain and state in the manuscript, why this procedure was chosen, as it appears not to be the best choice to do image alignments with features within the region of interest.*

In addition, for all SEM images – a scale bar should be provided (e.g., 1c and d)

Reply: Thanks for pointing this out, which might mislead readers. The “reference pattern” in Fig 1b was only used during data acquisition in the FIB/SEM, so FIB milling and SEM image acquisition were performed with better precision. However, this is still too coarse, later on a more accurate alignment was performed during image processing. This additional step is based on comparing the individual difference between the current image and the next one, which is efficient, as we do see the distorted pores are fixed and the whole structure shows isotropy regarding tortuosity.

The tilting is not from misalignment. Because if misalignment causes such large tilting, the pores should also be tilted in that direction. From Fig. 1d, the pores are appeared to be spherical. The tilting of the edge should result from the non-parallel alignment of the top surface of the SBE and substrate.

To clarify this, following part is modified:

“..... as shown in Figure 1d). Therefore, a box region of the image stack is used to performed additional alignment. The additional image alignment is performed to correct the distorted pore shapes in a registration step (Figure 1d)) as described in the Method section. The edge of the SBE sample is a tilting line due to the sample placement angle. Thereafter, the pore”

Finally, the scale bars are added into Fig. 1c) and d). Thanks for pointing that out.

4. The authors describe that they apply a threshold to separate pore space from polymer. Can the authors comment, why a threshold value was chosen in a way that the achieved pore space matches the “theoretical porosity calculated from the liquid phase in the SBE mixture”? Thereby, the actually measured pore space was adjusted to a predicted value. Some further explanations should be added to the manuscript, why the pore space was measured if it is adjusted afterwards (“model driven science”)?

Reply: We agree with the reviewer that the ideal scenario would be to base the 3D model only on the FIB/SEM images. However, there are transition regions between the pore and polymer phases which contain a width with a few pixels that cannot be unambiguously assigned to a pore or polymer. Additionally, some transition regions could appear even brighter than the polymer phase due to the charging effect at the edge as seen in Fig. 2a. The pixels in these transition regions need to be decided based on an objective value. For example, Neusser (Nanoscale, 2017) uses the **most popular pixel gray value**, which does not have any physical meaning or proof to be correct. Therefore, instead of using an arbitrary threshold value, we use the pore volume fraction obtained by another

method. This approach suits the goal of our study well – using the FIB/SEM to obtain the information on pore structure, and then base modelling on the information on porosity.

Additionally, variation of the threshold value only influences the size of the pore by a few pixels and not the 3D network. From Figs. 5f) and 5g), it can be seen the material properties are quite linear with porosity for the porosity range between 30 to 40 %, which indicates the porosity is not critical to the 3D connection in this region.

Regarding to the “model driven science”, the pore space was **not** measured later in the paper. It is the **individual pore size** that was measured. It is still a study of the 3D structure. Therefore, we **did not** apply the theoretical value and measured it again.

As a summary to the comment, the FIB-SEM technique is used to investigate the 3D structure of the SBE. The threshold value is chosen based on an objective value to allow determination of the phases at the soft transition region and it is also later found not critical to the 3D structure from simulation. To clarify this, we added a few sentences in the manuscript:

“..... separated by threshold segmentation. The transition regions between polymer, and pore phases are a few pixels due to the low contrast, different depths of the pores and charging at the pore edges. The best option is to use the true porosity instead of an arbitrary threshold. However, experimental measurements of porosity..... ”

5. *In addition, Reference 23 refers to NMR and threshold calculation (Cattaruzza, M. et al.) is only submitted for publication (2023). A manuscript submitted for publication cannot be cited. In this case, it is not clear how the pore space was calculated from NMR data. This is particularly problematic as the value was then used as threshold value.*

Reply: Luckily the paper is now published. The citation is changed to:

“31. Cattaruzza, M., Fang, Y., Furó, I., Lindbergh, G., Liu F., & Johansson, M. Hybrid polymer–liquid lithium ion electrolytes: effect of porosity on the ionic and molecular mobility. *J. Mater. Chem. A* **11**, 7006-7015 (2023).”

6. *Page 10 The information on achievable resolution and the issue of pores, which are too small to be visualized via FIB/SEM tomography should be discussed more prominently, earlier in the text.*

Reply: Thanks for the suggestion. Now the introduction part on FIB-SEM contains resolutions issue of pores (see reply to comment 1 by Reviewer 1).

7. *The authors should comment on the swollen state of the polymer and what the expected differences in pore space and polymer volume would be? Are there any measurements*

confirming changes in volume during drying? How can the presented data be compared/connected with the possibilities for ion transport in the actual wet state when the authors expect that the pore channels may be even closed? A suggestion would be to perform measurements under cryogenic conditions and add such data to the manuscript. How useful are these simulations if the real state and the investigated state of the material is quite different?

Reply: As discussed, the swollen state is a possible reason for the lower predicted ionic conductivity compared to that measured. As stated in the manuscript, as the polymer backbone swells the porosity decreases, and small pores may even close due to the swelling of the surrounding polymer. Experiment under cryogenic condition is indeed part of our plan for future studies. We are purchasing the equipment and hope to have it available next year. The simulation is still very useful. Firstly, the elastic modulus prediction is accurate. Secondly, the model for ion transportation can be updated according to future results. Thirdly, comparison between different structures or porosities is still valid.

8. Technical comments:

Some typos throughout the manuscript.

Regarding light-weight composite electrodes, the authors may add a few more references.

Reply: Thanks. We have added a sentence to introduce state of the art structural electrodes in the introduction section of the manuscript.

“ structural batteries must be of high stiffness. Structural electrodes are generally utilizing carbon fibers. [8, 9] In the negative electrode, carbon fibers are used as active material, i.e., host of lithium, current collector, and reinforcement. [6, 10] In the positive electrode, active material, e.g., lithium iron phosphate is coated on the carbon fiber that acts as current collector and reinforcement. [11, 12]. For the same reason, the liquid electrolyte...”

The following references have been added:

8. Asp LE, Johansson M, Lindbergh G, Xu J, & Zenkert D. Structural Battery Composites: A Review, *Funct. Compos. Struct.* **1**, 042001 (2019).

9. Jin T., Singer G., Liang K. & Yang Y. Structural batteries: Advances, challenges and perspectives. *Mater. Today*. **62**, 151–167 (2023).

10. Carlstedt D, et al. Experimental and computational characterization of carbon fibre based structural battery electrode laminae. *Compos. Sci. Technol.* **220**, 109283 (2022).

11. Hagberg J. et al. Lithium iron phosphate coated carbon fibre electrodes for structural lithium ion batteries. *Compos. Sci. Technol.* **162**, 235–43 (2018).

12. Sanchez JS, et al. Electrophoretic coating of LiFePO₄/graphene oxide on carbon fibers as cathode electrodes for structural lithium ion batteries. *Compos. Sci. Technol.* **208**, 108768 (2021).

Reviewer #3

1) *Validation of SVE: A statistical analysis of the extracted 20 SVEs after meshing to demonstrate that they preserve the same channels properties and density would be important to assess the robustness of the simulations.*

Reply: In the simulations, we do actually keep track of the composition of solid/liquid for each SVE as shown in Figure 5 (see the volume fractions on x-axis for each SVE). Based on this information, it becomes a trivial task to compute the effective density for the homogenized medium by, e.g., using a weighted arithmetic mean approach. As already mentioned in the section for “Results and discussions” , line 245; the samples do have a small variation in the microstructure volume fractions. However, this variation is deemed to be small enough to classify the SVEs as consistent representations (the spread corresponds to roughly +/- 5 percentage points from the average value), or, in other words, to be representative in a statistical sense.

In general, there are no differences between the raw voxel data from the scans and the meshed SVEs in FEM due to the fact that the raw voxel data is currently being used directly as the computational domain. Indeed, the reason this approach has been taken here is precisely because it is difficult to re-mesh the geometry while at the same time fully preserve the morphology of the microstructure.

2) *Including extra information about the finite element model size would help in understanding the need of hpc cluster to perform the simulations. Moreover, the authors attribute the difference in the estimation of the ionic conductivity to the fact that the simulations do not consider the increase in polymer backbone volume due to the absorption of the liquid electrolyte. Would Including this effect, with self-contact have been impractical from a computational point of view due to the excessive model complexity?*

Reply: As previously mentioned in the section “Methods” , line 449; the SVEs are voxel based geometries. Since each scanned voxel is represented by a hexahedron Finite Element in the simulations, we end up with SVEs that are discretized with up to 400,000 elements, corresponding to roughly 12 million degrees of freedom each. It should be noted, however, that quadratic shape functions were used for the hexahedron elements, resulting in a high FEM accuracy to guarantee mesh convergence.

Indeed, the multi-physics modeling of the swelling of the solid phase in the SBE is by no means a trivial task as it requires a multidisciplinary effort to mathematically describe. Furthermore, due to the added non-linearity stemming from computational

contact mechanics, it will also dramatically increase the computational effort required to solve the problem at hand. Currently, one of the computational constraints is that the raw voxel data is directly used as the computational mesh for FEM. The massive number of voxels that are scanned will, in this case, always automatically result in an extremely high-resolution FEM model. Therefore, for simplicity, the constitutive laws have been restricted to linear relations as the model size has already surpassed 10 million degrees of freedom. Nevertheless, modeling the swelling of the polymer for effective ionic conductivity predictions is definitely of great interest, and is certainly a problem that will be tackled in future research.

3) A table including the input used for the calculation of the ionic conductivity presented in Tab.2 would facilitate the understanding of the results.

Reply: The computationally predicted ionic conductivity in FEM depends solely on (i) the raw voxel data for geometry creation and (ii) the intrinsic ionic conductivity of the pure liquid electrolyte. Therefore, all input data that is required to predict the effective ionic conductivity has already been presented in the paper (Tab. 2).

4) Tab. presents the line of “Elastic Modulus” that is not aligned

Reply: Thanks. The line is now aligned.

5) Fig. 5 f and g quality could be improved

Reply: Thanks for the suggestion. We now update the image with the same format as Fig. 5 a)-b) and updated the figure caption.

8th Jun 23

Dear Professor Asp,

Your manuscript titled "3D reconstruction and computational analysis of the electrolyte for a structural battery composite" 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).

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Best regards,

Dr Jet-Sing Lee
Associate Editor
Communications Materials
orcid.org/0000-0002-6740-8700

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have answered the comments and I recommend the publication of the article.

Reviewer #2 (Remarks to the Author):

The authors addressed a majority of the raised issues and hence, the manuscript is now recommended for publication.

Reviewer #3 (Remarks to the Author):

The authors clarified all the reviewer's comments and the manuscript is ready to be published in "Communications Materials".