

Peer Review File

Analyzing microstructure relationships in porous copper using a multi-method machine learning-based approach



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Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality

13th Sep 23

Dear Dr Brunner,

Thank you for submitting your manuscript, "A Machine Learning-Based Methodology to Uncover the Structure-Property Relationship in Porous Materials", 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 potential interest, they have raised substantial concerns that must be addressed. In light of these comments, we cannot accept the manuscript for publication, but are interested in considering a revised version that addresses these serious concerns.

In particular, Reviewer 2 suggests that there is no significant improvement compared to prior reports, highlighting that a similar machine learning pipeline has been previously proposed, which should be carefully commented upon. In addition, both Reviewers 1 and 2 suggest clarifying the significance/objective of this study and its originality compared to similar works. All reviewers request for further clarification on the methods used.

We hope you will find the referees' comments useful as you decide how to proceed. Should further experimental data or analysis allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. However, please bear in mind that we will be reluctant to approach the referees again in the absence of major revisions. If the revision process takes significantly longer than three months, we will 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 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.

-A clean version of the manuscript. Please select the file type 'Article File'.

-An updated 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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Please do not hesitate to contact me if you have any questions or would like to discuss the required revisions further. 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 submitted the article titled: "A Machine Learning-Based Methodology to Uncover the Structure-Property Relationship in Porous Materials". The topic is interesting, and the article is well written. Here are some suggestions for authors to improve their manuscripts:

1. The introduction part should be significantly improved. Please emphasize the significance and novelty of this research.
2. The author should check the title of each section. The section "results" should be "methods".
3. It is necessary to provide a description of the dataset used. The author should clarify the number of data generated by the FIB-SEM method and the number or percentage of data used for training.
4. Each figure should have a number.
5. Table 1. Why does the author use U-net for segmentation? The accuracy of segmentation results is low. The author can try more advanced semantic segmentation methods to improve prediction accuracy.
6. Fig. 5 is not clear. I see multiple input parameters for multivariable linear regression (MVLRL), but I don't know how the authors extract these features from the 3D model. Are these parameters estimated from each picture of the 3D model? Please add a section to describe it.
7. Line 706. How many datasets does MVLRL use? The authors simply stated that three different microstructure sets (not used as training datasets) were used in MVLP predictions. Please add a dataset description.
8. Please explain how to determine the quality of images generated by conditional GAN.

Reviewer #2 (Remarks to the Author):

Comments for Author

This paper proposes a machine learning based pipeline to reveal the structure-property relationship of porous materials

The proposed method consists of three parts.

The first part is a hybrid method for semantic segmentation of FIB-SEM 3D images, which combines a naive threshold-based heuristic and a U-Net-based model.

The binary labels for the semantic segmentation are material and pore.

Each pixel of the FIB-SEM image is annotated into two classes by its grey level, whose threshold is defined from the histogram of a greyscale image.

To improve the labeling accuracy, superpixel algorithm is applied to the raw image and the grey value in each superpixel is averaged before annotating the label.

Then, a U-Net based semantic segmentation model is trained on the annotated images.

The U-Net hybrid method, which uses both superpixel annotation and U-Net, shows better performance in terms of four statistical metrics, namely Jaccard index, precision, recall and accuracy, than the non-hybrid methods.

The various structural features, such as relative density, specific surface area, etc., are calculated from the segmented images.

The second component is a regression model that predicts electrical conductivity using the extracted structural features.

A multi-variable linear regression model is used because it is easy to interpret the variable contribution to the prediction.

From the leave one out cross-validation, the authors found that the model using specific surface area and two high-level features consisting of relative density, tortuosity and curvatures has the largest R2 metric.

The third component is a conditional generative model that generates sectioned FIB-SEM images conditioned on electrical conductivity.

A conditional generative adversarial network is used for the conditional generative model.

The realistic FIB-SEM images, which have similar structural properties to real materials, are successfully generated and can be controlled by the electrical conductivity as a condition.

The critical point of this paper is that there is no originality and no significant improvement.

The similar machine learning pipeline has been proposed previously in many articles.

Moreover, the readers could not understand why the chosen models, supervised U-Net, MVLR, CGAN, are suitable for the present objective.

For example, you can use LIME or SHAP for feature interpretation, which are widely used for the XAI community.

These XAI methods can be used for the non-linear regression models.

A linear model is certainly easy to understand, but is not applicable to the complex tasks.

In addition, the CGAN results are trivial and redundant for this paper.

The readers already know that the CGAN can generate realistic images conditioned on the input features.

To improve the present paper, the followings are recommended:

1. Clarify the objective and originality

Most materials researchers know that material design is difficult because material synthesis and property measurements require a lot of resources.

The ultimate goal is to create a material with the desired property with limited time, instrumentation, energy and human resources.

Therefore, one of them should be improved compared to the existing methods and the authors should show the evidence.

What problem does the proposed method solve and what is its original idea?

Many similar machine learning based pipelines have been proposed to reveal the structure-property relationship.

What is the main proposal of the present model that differs from the existing methods?

2. Focus on one objective.

Three machine learning methods are used in this paper.

However, they are not related to each other.

If the authors really want to show the relationship between structural features and material properties, one suggestion is to use the invertible model such as normalising flow and diffusion model, because the invertible model directly learns a relationship between the conditions and generation targets.

If not, it is recommended that you focus on one of the three tasks.

For example, you can focus on "Fast and accurate semantic segmentation from FIB-SEM images of porous materials" and use the electrical conductivity prediction as an evaluation of the proposed segmentation method.

The combination of superpixel preprocessing and U-Net could be an original and interesting idea if some reasonable or theoretical explanation is added as to why this combination is effective.

Comparison with other existing segmentation methods is of course required in the Experiments section.

If you want to focus on the analysis of the porous materials itself, rather than the machine learning methodology, new insights from the observations should be clarified.

The results of the geometric analysis of porous copper reported in this paper are trivial.

Minor comments

1. (p.10, lines 238-240) What is the difference between "specimens" and "samples"? 18 samples would be three types of copper paste, HPA, HPB and NPC, at six different temperatures. Why is the number of specimens three times the number of samples?
2. (p.12, lines 283-287) Why is the predicted electrical conductivity used for the condition of the CGAN instead of the measured value of the bulk material?
3. (p.14) The authors say "Usually, machine learning models work with categorical variables", but this is not true because the machine learning models have been widely applied to the non-categorical variables.
4. (p.16, line 394) Batch normalisation is not suitable for batch size 1 because the statistics in minibatch are used in this layer. If you do not have enough memory, you can use layernorm as instead.
5. (p.17, line 414-p.18, line 425) The authors discretise the electrical conductivity to use for the condition, but the discretisation is not necessary because the continuous variable can be used for the condition.
6. (Table 1) Why can the U-Net improve the semantic segmentation performance? In general, the supervised model cannot outperform the supervisor because the annotated label is treated as the ground truth. Did the authors use any technical training procedure?
7. What is the sintering time of the sample? What is the temperature for electron conduction measurements?
8. Is the electrical conductivity uniformly distributed in $[0, 250]$ $\mu\text{S}\cdot\text{cm}^{-1}$? In Fig. 3(c), Fig. 6(c)-(e) and Supplementary Fig. 6, the distribution of the electrical conductivity does not appear to be uniform, being concentrated in $[0, 50]$ $\mu\text{S}\cdot\text{cm}^{-1}$. What is the R^2 value of the prediction for each of the three sample types? Scatter plots are recommended for Fig. 5 (b)-(d) rather than line plots. The meaning of the shading in these figures is not clear.

Reviewer #3 (Remarks to the Author):

This paper presents a comprehensive study of applying machine learning tools to segmentation, microstructural feature extraction, and quantification. The relation between the electrical conductivity and the microstructural features are examined using the multi-variable linear regression models. The paper also studied the use of conditional generative adversarial networks to generate microscopic images in correlation with the electrical conductivity.

In general, the paper is very well written with excellent collections of technologies employed in the characterization of microstructures. In addition to the comprehensiveness, the paper also reports some novel approaches, for example, the two-stage hybrid pore segmentation approach based on the histograms second derivative, and the MVLR models as well as the introduction of the combinational parameters α and β .

While this is a well-written comprehensive manuscript, I do have some questions as highlighted below:

- 1) The rationale of using the first maximum of the histogram's second order derivative (Supplementary Fig. 2a) could be explained in further detail. In addition, this figure may deserve to be in the main text instead of in the supplementary note.
- 2) The compositions (e.g., sizes of the copper particles) of the three samples (HPA, HPB, and NPC) need to be clearly given. The rationale of choosing the three samples need to be addressed. What are the differences between HPA and HPB, and why they behave differently?
- 3) The imaging/tomography volume is only $10\mu\text{m} \times 10\mu\text{m} \times 10\mu\text{m}$. What is the imaging volume in comparison with the domain size (e.g., the pores and the copper struts)? Will there be any truncation error, i.e., the size of the features may be approaching the size of the imaging. This may not be an issue for NPC but could be an issue for HPA and HPB. It also may become an issue as the size of the copper particles grow as the sintering temperature or duration increases.

Point-by-point Referee Response

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors submitted the article titled: "A Machine Learning-Based Methodology to Uncover the Structure-Property Relationship in Porous Materials". The topic is interesting, and the article is well written. Here are some suggestions for authors to improve their manuscripts:

We thank the reviewer for the very positive reply and suggestions to improve the manuscript. We think that the manuscript benefited very much from the suggestions.

1. The introduction part should be significantly improved. Please emphasize the significance and novelty of this research.

According to your comment we rewrote the introduction and tried to emphasize the novelty of the work. Further, we extended the presented methodology by successfully utilizing the recently developed denoising diffusion probabilistic model (DDPM) for porous materials and compare the DDPM and GAN performance for the synthetic image reconstruction.

In particular in this work we develop a methodology capable to understand the processing-structure-property relationship for differently processed porous materials. We utilize a multi-method machine learning approach incorporating tomographic image data acquisition, segmentation, microstructure feature extraction, feature importance analysis and synthetic microstructure reconstruction. Enhanced segmentation with an accuracy of about 95 % based on an efficient annotation technique provides the basis for accurate microstructure quantification, prediction and understanding of the correlation of the extracted microstructure features and electrical conductivity as well as temperature. We show that a diffusion probabilistic model, superior among a GAN model provides synthetic microstructure images including physical information in agreement with real data, an essential step to predict properties of unseen conditions.

2. The author should check the title of each section. The section "results" should be "methods".

We deliver results based on the developed methodology in the results section. Further technical information with respect to the methodology is presented in the method section. Thank you for the comment we revised the manuscript to increase the readability.

3. It is necessary to provide a description of the dataset used. The author should clarify the number of data generated by the FIB-SEM method and the number or percentage of data used for training.

We provide now more information with respect to the data set description in the method section. The image slices generated by SEM are 450 slices per dataset. The training images used for U-Net is 20% (90 slices) per dataset. For cGAN and denoising diffusion probabilistic models (DDPM), each temperature class contains 1350 images. In total, the number of images is 8100 images. Please see the information in the method section of the manuscript.

4. Each figure should have a number.

Thank you for the comment. We added the figure number on each Fig as well as the title of the figure.

5. Table 1. Why does the author use U-net for segmentation? The accuracy of segmentation results is low. The author can try more advanced semantic segmentation methods to improve prediction accuracy.

The accuracy of the utilized U-net segmentation is about 94% which is quite high in comparison to recent literature, (usually 79 -91%¹⁻³). We are able to achieve such a high accuracy due the developed annotation process presented in the manuscript, which is based on a U-NET trained with the hybrid segmentation. We modified the text accordingly. Please see, further information in the modified first section of the result section “A need for an efficient annotation and deep learning microstructure segmentation.” As well as in the modified Fig.1. We think that the accuracy is appropriate for the presented study but of course can be increased in the future by other models.

6. Fig. 5 is not clear. I see multiple input parameters for multivariable linear regression (MVLRL), but I don't know how the authors extract these features from the 3D model. Are these parameters estimated from each picture of the 3D model? Please add a section to describe it.

Thank you for the comment. In particular we added further information about the extraction of the microstructure features in the method section as well as modified the manuscript text. The microstructure features utilized as an input for the MVLRL are extracted from the experimental tomographic data obtained by FIB-SEM.

7. Line 706. How many datasets does MVLRL use? The authors simply stated that three different microstructure sets (not used as training datasets) were used in MVLRL predictions. Please add a dataset description.

Sorry that this was not clearly described in the manuscript. For the MVLRL all microstructure features were evaluated from the segmented VOI with $10 \times 10 \times 10 \mu\text{m}^3$ obtained from the FIB-SEM tomography. We revised and improved the manuscript text in this context as well as provided more information in the method section.

There are three types of porous copper materials (HPA, HPB, and NPC) used in this paper. Each porous copper type is sintered at six different final sintering temperatures. Therefore, there are 18 samples used as the training dataset each with a VOI of $10 \times 10 \times 10 \mu\text{m}^3$. We maximize the use of the training set with a leave one out cross validation (LOOCV) to obtain reliable and unbiased results. As a result, 18 conductivity predictions of each unseen validation sample are produced from one leave one LOOCV-set.

A further test dataset with 3 different materials is used not used in the training. Those materials again provide each a VOI of $10 \times 10 \times 10 \mu\text{m}^3$. Additional information is provided in Supplementary Note 8 and Supplementary Fig. 8.

8. Please explain how to determine the quality of images generated by conditional GAN.

We completely revised the results in context to the synthetic microstructure reconstruction. In addition to the GAN we also utilize a more recently developed denoising diffusion probabilistic model and compare their performances. With respect to the determination of the quality we take the following remarks from literature⁴. ‘It should be mentioned that different metrics such as Frechét inception distance (FID) and precision and recall (PR) are common in computer vision for quality assessment. However, they are not interpretable as statistical microstructural descriptors (SMDs), and usually need a large dataset to compare the similarity between two distributions (i.e., Pdata and Pmodel) in high dimensions. Hence, they only show the general performance of the model to match the real data and do not ensure that each reconstructed image has the same structural and

topological properties as your real images.’ Therefore, also from a material science point of view we use as a measure of the performance the physical descriptors or microstructure features (relative density, specific perimeter or shape index) extracted from the segmented synthetic and real images. The quantified features from the real and synthetic images are compared with each other and in addition a regression analysis is performed and the linearity between the real and synthetic results utilizing the R^2 values evaluated. See the new chapter “Synthetic image reconstruction of the porous microstructures for different sinter temperatures and electrical conductivity values” and “Evaluation of the model performance” in this context as well as Fig. 6.

Reviewer #2 (Remarks to the Author):

Comments for Author

This paper proposes a machine learning based pipeline to reveal the structure-property relationship of porous materials. The proposed method consists of three parts.

The first part is a hybrid method for semantic segmentation of FIB-SEM 3D images, which combines a naive threshold-based heuristic and a U-Net-based model. The binary labels for the semantic segmentation are material and pore. Each pixel of the FIB-SEM image is annotated into two classes by its grey level, whose threshold is defined from the histogram of a greyscale image. To improve the labeling accuracy, superpixel algorithm is applied to the raw image and the grey value in each superpixel is averaged before annotating the label. Then, a U-Net based semantic segmentation model is trained on the annotated images. The U-Net hybrid method, which uses both superpixel annotation and U-Net, shows better performance in terms of four statistical metrics, namely Jaccard index, precision, recall and accuracy, than the non-hybrid methods. The various structural features, such as relative density, specific surface area, etc., are calculated from the segmented images. The second component is a regression model that predicts electrical conductivity using the extracted structural features. A multi-variable linear regression model is used because it is easy to interpret the variable contribution to the prediction. From the leave one out cross-validation, the authors found that the model using specific surface area and two high-level features consisting of relative density, tortuosity and curvatures has the largest R^2 metric. The third component is a conditional generative model that generates sectioned FIB-SEM images conditioned on electrical conductivity. A conditional generative adversarial network is used for the conditional generative model. The realistic FIB-SEM images, which have similar structural properties to real materials, are successfully generated and can be controlled by the electrical conductivity as a condition. The critical point of this paper is that there is no originality and no significant improvement. The similar machine learning pipeline has been proposed previously in many articles. Moreover, the readers could not understand why the chosen models, supervised U-Net, MVLR, CGAN, are suitable for the present objective. For example, you can use LIME or SHAP for feature interpretation, which are widely used for the XAI community. These XAI methods can be used for the non-linear regression models. A linear model is certainly easy to understand, but is not applicable to the complex tasks. In addition, the CGAN results are trivial and redundant for this paper. The readers already know that the CGAN can generate realistic images conditioned on the input features.

We think that the paper was not well written in order to see the novelty. Therefore, we modified the manuscript completely. We thank the reviewer in particular for the suggestion to utilize a diffusion model-based ansatz but also mention SHAP for the feature interpretation. We think that in particular the implementation of the diffusion model and comparison with GAN provides further novel insights and improves the paper again. Since the reviewer did not provide any citation we

cannot argue about similar pipelines which have been previously proposed in articles. The analysis of micron sized porous samples with features down to the nm range is not trivial and needs advanced experimental setups like FIB-SEM as well as a sophisticated analysis algorithm toolbox, especially to uncover the correlation of the microstructure with the electrical property or the processing parameter temperature, as presented here.

To improve the present paper, the followings are recommended:

1. Clarify the objective and originality

Most materials researchers know that material design is difficult because material synthesis and property measurements require a lot of resources. The ultimate goal is to create a material with the desired property with limited time, instrumentation, energy and human resources. Therefore, one of them should be improved compared to the existing methods and the authors should show the evidence. What problem does the proposed method solve and what is its original idea? Many similar machine learning based pipelines have been proposed to reveal the structure-property relationship. What is the main proposal of the present model that differs from the existing methods?

We thank the referee for the questions. Material design and the understanding of the processing-structure-property relationship by utilizing artificial intelligence-based approaches represents a quite new topic in material science. Here we present a methodology capable to predict properties of unseen conditions. According to our knowledge this has not been shown so far although attempts have been made recently showing partly success with individual steps in this direction (like segmentation or the synthetic image generation) but have never illustrated a full methodology as presented her with all the necessary steps for porous materials showing microstructures on nm- and μm -scales.

We revised the manuscript with respect to your comments in particular implemented according to your suggestion methods like SHAP and the recently developed diffusion model.

Within the paper we develop a methodology and not a single method capable to understand the processing-structure-property relationship for differently processed porous materials. A single method will fail, it will take a combination of approaches in material science to succeed as shown here. We utilize a multi-method machine learning approach incorporating tomographic image data acquisition, segmentation, microstructure feature extraction, feature importance analysis and synthetic microstructure reconstruction. The evaluation of the microstructure features, their physical analysis, as well as their correlation to the material property display crucial ingredients for accelerated material design and to uncover the processing-structure -property relationship. Enhanced segmentation with an accuracy of about 95 % based on an efficient annotation technique provides the basis for accurate microstructure quantification, prediction and understanding of the correlation of the extracted microstructure features and electrical conductivity. We show that a diffusion probabilistic model superior among a GAN model, provides synthetic microstructure images including physical information in agreement with real data, an essential step to predict properties of unseen conditions.

Our findings highlight not only the importance of synthetic image generation and of accurately retrieving a set of microstructural features with statistical confidence for accelerated material design but also scrutinizing the features physical meaning, a key to conclude about the process-structure-property relationship.

To our knowledge this has been not shown so far for the material science community. In particular less has been shown in context to diffusion models for the synthetic reconstruction of the microstructure, microstructure feature extraction and correlation analysis.

2. Focus on one objective.

Three machine learning methods are used in this paper. *However, they are not related to each other. If the authors really want to show the relationship between structural features and material properties, one suggestion is to use the invertible model such as normalising flow and diffusion model, because the invertible model directly learns a relationship between the conditions and generation targets.* If not, it is recommended that you focus on one of the three tasks. For example, you can focus on "Fast and accurate semantic segmentation from FIB-SEM images of porous materials" and use the electrical conductivity prediction as an evaluation of the proposed segmentation method. *The combination of superpixel preprocessing and U-Net could be an original and interesting idea if some reasonable or theoretical explanation is added as to why this combination is effective. Comparison with other existing segmentation methods* is of course required in the Experiments section. If you want to focus on the analysis of the porous materials itself, rather than the machine learning methodology, new insights from the observations should be clarified. The results of the geometric analysis of porous copper reported in this paper are trivial.

We thank the reviewer for her/his valuable input. We strongly modified the manuscript according to her/his comments.

We added more information with respect to the superpixel preprocessing and Unet in the manuscript as suggested. Please see Fig. 1 as well as the corresponding result section. As well as implemented the diffusion model in our methodology.

We argue that a single method approach usually will be not sufficient to trigger accelerated material design. Rather a methodology comprising several methods will provide a solution. We point out that not only the importance of synthetic image generation and of accurately retrieving a set of microstructural features with statistical confidence for accelerated material design but also scrutinizing the features physical meaning, is essential to conclude about the process-structure-property relationship.

Therefore, several methods are necessary and aligned for the given material. This is exactly what we do in this paper for porous materials exemplarily for sintered porous copper which display an interesting candidate for novel interconnect technologies and where efficient material design becomes important. Various steps are necessary to gain an understanding of the underlying correlations delivering essential knowledge to provide design guidelines. We therefore utilize a multi-method machine learning approach incorporating tomographic image data acquisition, segmentation, microstructure feature extraction, feature importance analysis and synthetic microstructure reconstruction. Each presented method is connected within the multi-method workflow. We modified the manuscript text accordingly to show this more prominent.

Minor comments

1. (p.10, lines 238-240) What is the difference between "specimens" and "samples"? 18 samples would be three types of copper paste, HPA, HPB and NPC, at six different temperatures. Why is the number of specimens three times the number of samples?

Thank for the comment and careful reading there was a mistake in the text. You are fully right the number is 18.

2. (p.12, lines 283-287) Why is the predicted electrical conductivity used for the condition of the CGAN instead of the measured value of the bulk material?

Thank you for your question suggesting. We changed the methodology and are using now electrical conductivity or temperature levels as the labels.

3. (p.14) The authors say "Usually, machine learning models work with categorical variables", but this is not true because the machine learning models have been widely applied to the non-categorical variables.

Thank you for the comment, we changed the sentence in the manuscript.

4. (p.16, line 394) Batch normalisation is not suitable for batch size 1 because the statistics in minibatch are used in this layer. If you do not have enough memory, you can use layernorm as instead.

Thank you for the comment we changed the misleading sentence in the manuscript accordingly. Thank you for the input. We are using Layernorm layers now by replacing the Batchnorm.

5. (p.17, line 414-p.18, line 425) The authors discretise the electrical conductivity to use for the condition, but the discretisation is not necessary because the continuous variable can be used for the condition.

Thank you for the suggestion. In the current version we dropped the discretization method. Instead, we use the obtained conductivities or temperatures as the labels.

6. (Table 1) Why can the U-Net improve the semantic segmentation performance? In general, the supervised model cannot outperform the supervisor because the annotated label is treated as the ground truth. Did the authors use any technical training procedure?

According to your question the manuscript text in this context was misleading. We changed the text accordingly. The softmax activation function at the end of the U-Net architecture provides a pixel's class probability. Therefore, we think the U-Net segmentation can rectify some inaccurate pixels if we have high training pixel accuracy.

7. What is the sintering time of the sample? What is the temperature for electron conduction measurements?

The sintering time is done with the ramping time of 5° C/min between room temperature and final sintering temperature. There is no holding time after the temperature is reached. We measure the specific resistivity/conductivity in situ, please see the method section.

8. Is the electrical conductivity uniformly distributed in [0, 250] uS.cm⁻¹? In Fig. 3(c), Fig. 6(c)-(e) and Supplementary Fig. 6, the distribution of the electrical conductivity does not appear to be uniform, being concentrated in [0, 50] uS.cm⁻¹. What is the R² value of the prediction for each of the three sample types? Scatter plots are recommended for Fig. 5 (b)-(d) rather than line plots. The meaning of the shading in these figures is not clear.

Please see more information in the modified version of the manuscript, method section. We replace the formula of β (see the main text). The R² values of each material are: R² HPA = 0.97, R² HPB = 0.61, R² NPC = 0.99. We modified Fig. 5 according to your comment.

Reviewer #3 (Remarks to the Author):

This paper presents a comprehensive study of applying machine learning tools to segmentation, microstructural feature extraction, and quantification. The relation between the electrical conductivity and the microstructural features are examined using the multi-variable linear regression models. The paper also studied the use of conditional generative adversarial networks to generate microscopic images in correlation with the electrical conductivity.

In general, the paper is very well written with excellent collections of technologies employed in the characterization of microstructures. In addition to the comprehensiveness, the paper also reports some novel approaches, for example, the two-stage hybrid pore segmentation approach based on the histograms second derivative, and the MVL models as well as the introduction of the combinational parameters α and β .

We thank the referee for the very positive report. Please see our answers to your questions below.

While this is a well-written comprehensive manuscript, I do have some questions as highlighted below:

1) The rationale of using the first maximum of the histogram's second order derivative (Supplementary Fig. 2a) could be explained in further detail. In addition, this figure may deserve to be in the main text instead of in the supplementary note.

We thank the reviewer for this question. We added more details in the manuscript text, as well as changed the corresponding figure (Fig.1) according to the question.

2) The compositions (e.g., sizes of the copper particles) of the three samples (HPA, HPB, and NPC) need to be clearly given. The rationale of choosing the three samples need to be addressed. What are the differences between HPA and HPB, and why they behave differently?

The copper pastes consist of copper micro- and nanoparticles. The nanoparticles are in the size range of about 150 nm before sintering, and the micron-sized particles are in the range of a micrometer.

HPA: At ambient conditions and with a shear rate of 50/s, HPA has a viscosity of 30.6 Pa.s, and its solids content is 78.8 %.

HPB: At ambient conditions and with a shear rate of 50/s, HPB has a viscosity of 4.0 Pa.s, and its solids content is 76.0 %.

NPC: At ambient conditions and with a shear rate of 50/s, NPC has a viscosity of 19.6 Pa.s, and its solids content is 84.0 %.

From the information above, the low viscosity of the HPB paste makes the HPB porosity higher. We use these materials to study the performance of porous copper for die-attach applications.

3) The imaging/tomography volume is only $10\mu\text{m} \times 10\mu\text{m} \times 10\mu\text{m}$. What is the imaging volume in comparison with the domain size (e.g., the pores and the copper struts)? Will there be any truncation error, i.e., the size of the features may be approaching the size of the imaging. This may not be an issue for NPC but could be an issue for HPA and HPB. It also may become an issue as the size of the copper particles grow as the sintering temperature or duration increases.

Very good question. We cannot fully neglect any truncation error. There is a trade off between the cost and the measurement volume. However, we compared the cross-section data from the FIB-SEM for different ROIs. The table below compares the ROI for $10\mu\text{m} \times 10\mu\text{m}$ and $50\mu\text{m} \times 50\mu\text{m}$ and the evaluated the microstructure features (relative density, specific perimeter and shape index) at 400

°C. No strong deviation by comparing the microstructure features for the two different ROIs can be observed. Overall the samples were fabricated to show on wafer level a rather recurring microstructure, that is a homogenous microstructure for large length scales. This also explains the results in the table below.

Sample	Physical descriptors	2D Data	Cross section
HPA 400°C	Relative density	71.3 ± 4.8	71.5 ± 2.6
	Specific perimeter	3.0 ± 0.3	3.6 ± 0.3
	Shape index	$0.094 \pm \pm 0.017$	0.10 ± 0.017
HPB 400°C	Relative density	45.6 ± 4.3	44.0 ± 1.75
	Specific perimeter	2.3 ± 0.2	2.3 ± 0.1
	Shape index	0.007 ± 0.01	0.01 ± 0.002

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6th Mar 24

Dear Dr Brunner,

Your manuscript titled "Multi-Method Machine Learning-Based Approach to Uncover Complex Microstructure Inferences in Porous Materials" has now been seen again by our referees, whose comments appear below. Reviewer 1 was not available to comment but we asked one of the other reviewers to check your replies to their comments. They mentioned to incorporate a dedicated paragraph in the Introduction section discussing the contribution of this paper to enhance clarity regarding its novelty. 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 revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you 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,

Dr Jet-Sing Lee

Associate Editor

Communications Materials

orcid.org/0000-0002-6740-8700

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Another reviewer says in regard to the revisions to Reviewer 1:

I would suggest to the authors to incorporate a dedicated paragraph in the Introduction section discussing the contribution of this paper to enhance clarity regarding its novelty.

Reviewer #2 (Remarks to the Author):

The authors have carefully addressed the reviewers' comments, resulting in significant improvements to the manuscript. Their work provides valuable insights, such as the importance of feature α for conductivity and the superiority of DDPM over CGAN for reproducing material properties. Furthermore, the importance of the proposed models to the authors' objectives is elucidated. If there are no objections, I consider the manuscript suitable for publication.

Reviewer #3 (Remarks to the Author):

The authors have answered all my concerns and addressed all my comments.

Rebuttal Letter

Dear Editor, Dear Reviewer,

Thank you very much for the positive reply.

Please see below our point-by-point response.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Another reviewer says in regard to the revisions to Reviewer 1:

I would suggest to the authors to incorporate a dedicated paragraph in the Introduction section discussing the contribution of this paper to enhance clarity regarding its novelty.

We modified the manuscript accordingly in the introduction to highlight the novelty of this work.

Reviewer #2 (Remarks to the Author):

The authors have carefully addressed the reviewers' comments, resulting in significant improvements to the manuscript. Their work provides valuable insights, such as the importance of feature α for conductivity and the superiority of DDPM over CGAN for reproducing material properties. Furthermore, the importance of the proposed models to the authors' objectives is elucidated. If there are no objections, I consider the manuscript suitable for publication.

We thank reviewer 2 for the positive reply.

Reviewer #3 (Remarks to the Author):

The authors have answered all my concerns and addressed all my comments.

We thank reviewer 3 for the positive reply.