

Supplementary Information

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

Andi Wijaya¹, Julian Wagner¹, Bernhard Sartory¹, Roland Brunner^{1*}

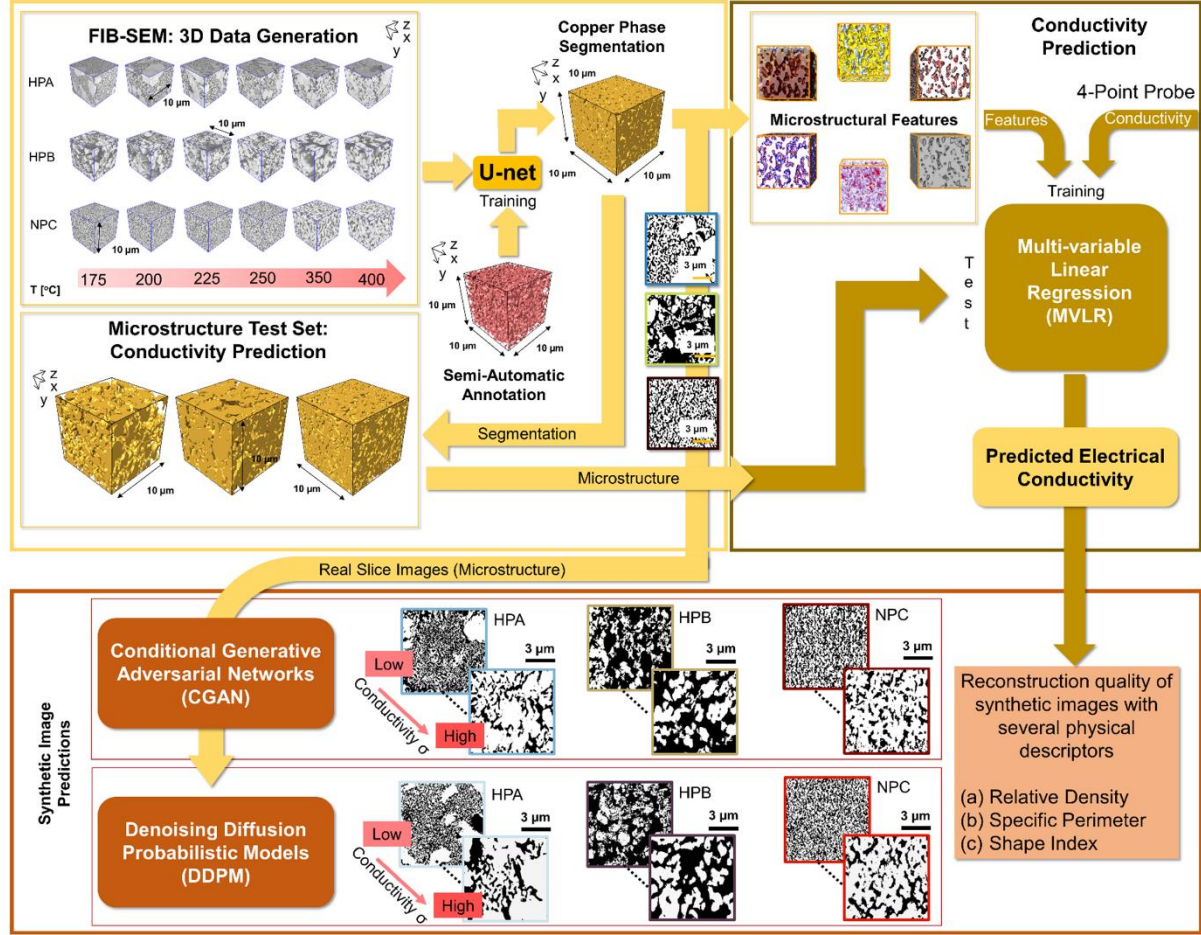
¹Materials Center Leoben Forschung GmbH, 8700 Leoben, Austria

*corresponding author: roland.brunner@mcl.at

Supplementary Note 1: Machine learning driven methodology to determine the interplay between the microstructure and electrical conductivity.

Supplementary Figure 1 illustrates the methodology, utilizing various interlinked supervised machine learning tools to achieve reliable segmentations from the volumetric image datasets, determine the interplay between the microstructural features and the electrical conductivity, and generate high-quality synthetic images.

Here, we aim for an accurate classification of two phases, associated with pore and copper, respectively, using a U-Net deep learning architecture¹. Accurate segmentation of the 3D copper datasets enables the quantification of the microstructural features from the segmented volumetric image data such as relative density, strut diameter, surface area, curvatures, and tortuosity. Further, within the methodology the understanding of the features are highly endorsed since those deliver essential ingredients for the multi-variable linear regression models to predict the electrical conductivity of a defined microstructure. Finally, we utilize cGAN and DDPM architectures to predict the specified material microstructures allocated to the conductivities. For the validation, we compare the physical descriptors, e.g, relative density, specific perimeter, and shape index, of the reconstructed images with the experimental data.

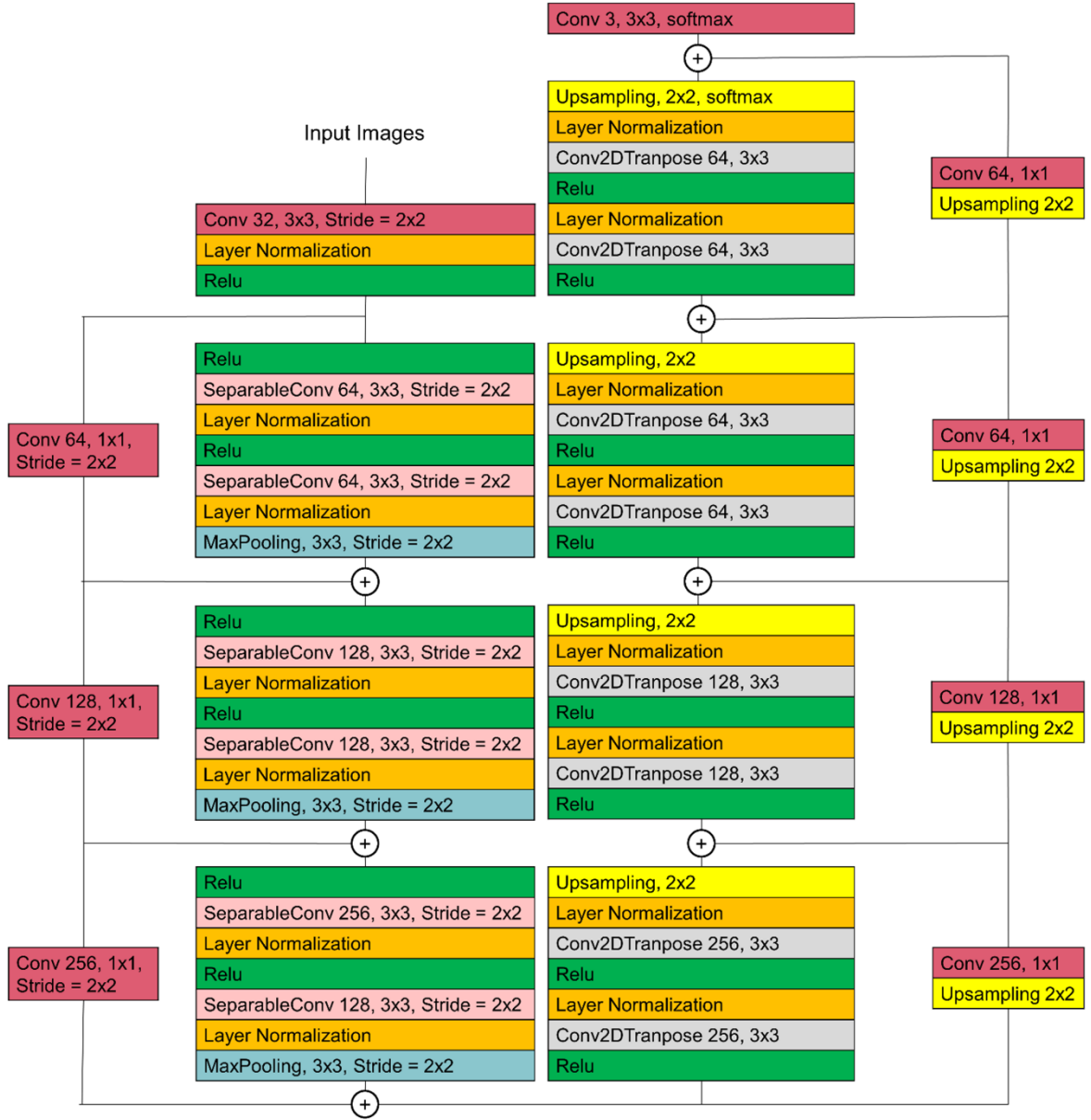


Supplementary Figure 1. ML-assisted methodology to infer the structure-property relationship of porous materials. Three different types of copper pastes, which are called HPA, HPB, and NPC, are sintered at six different temperatures. SEM-FIB nano-tomography is performed to obtain the volumetric image data of each sample. Each volume of interest (VOI) has a size of $10 \times 10 \times 10 \mu\text{m}^3$ with a voxel size of $18.6 \times 18.6 \times 25 \text{ nm}^3$. Accurate semi-automatic data annotation is performed to train the U-net model for proper image segmentation. For the annotation of the training data, a combination of thresholding and the super pixel segmentation method is utilized. The trained U-Net architecture is capable to segment the pore. From the segmented volumetric image datasets, we retrieve statistically relevant microstructural features. Microstructural features are used to train the multi-variable variable linear regression (MVLRL) model. Electrical conductivities are obtained experimentally by 4-point-probe measurements. MVLRL-based prediction models of the electrical conductivities are

performed on all three different porous materials datasets. Utilizing conditional generative adversarial networks (CGANs) and denoising diffusion probabilistic model (DDPM) in combination with an accurate segmentation method provide an opportunity to reconstruct high quality synthetic porous microstructure images. Reconstructed slice images and real data are compared for validation with several physical descriptors.

Supplementary Note 2: U-Net architecture.

The model is based on the U-net-like architecture from Chollet². The architecture has contraction and expansion paths, which are shown in Supplementary Figure 2. The difference in this method is that the normal convolution is replaced by the separable convolution, thereby reducing the filters dimensionality³. Another difference is the concatenation of the contraction residual, as seen in the U-Net model¹, is replaced by the addition operation between the current up-sampling operations and the residual of the previous up-sampling operation.

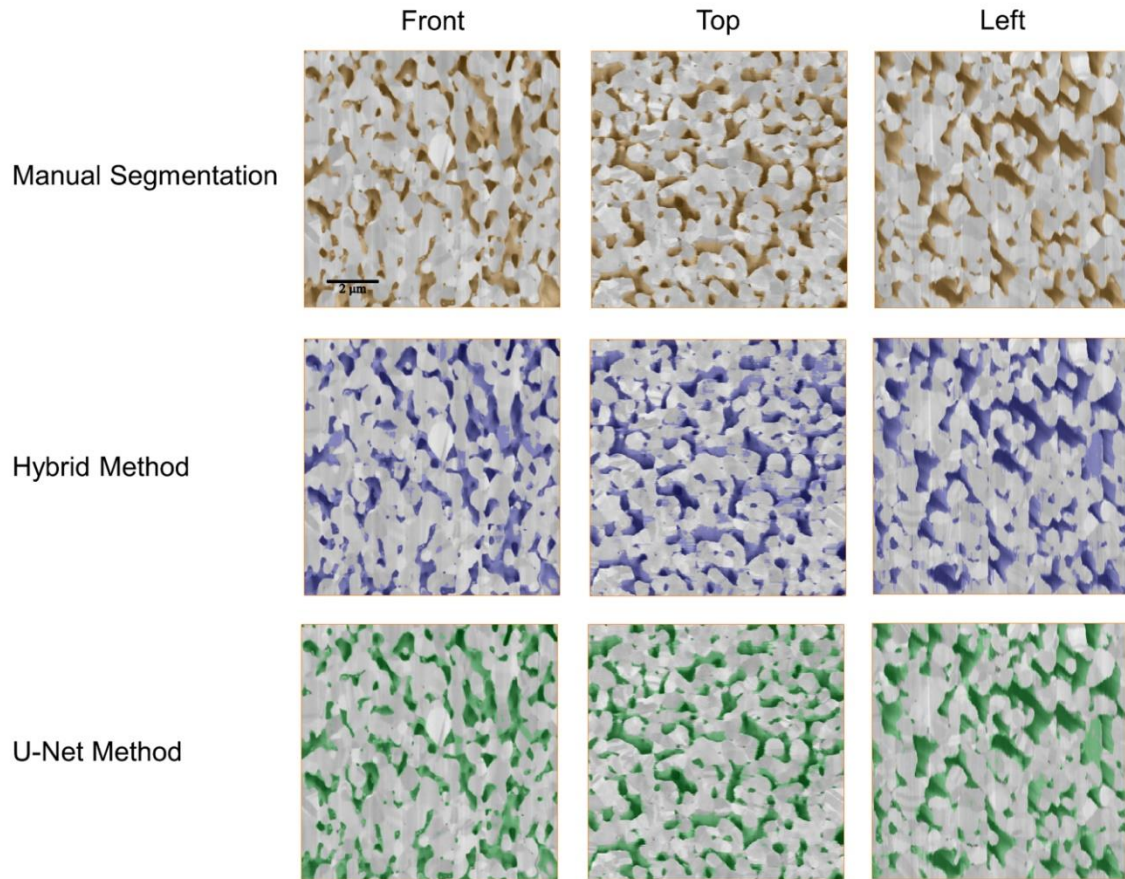


Supplementary Figure 2. The image segmentation architecture is proposed by Chollet.

The image is fed into the contraction path and the output is produced after the expansion path. The color of the box corresponds to the same type of operation. There are two main difference with the original U-Net model. First, the filters dimensionality is reduced. Second, the concatenation of the contraction residual is replaced by addition operation between current up-sampling operations and the residual of the previous up-sampling operations.

Supplementary Note 3: Manual Segmentation and Comparison with Threshold and U-Net like Segmentations.

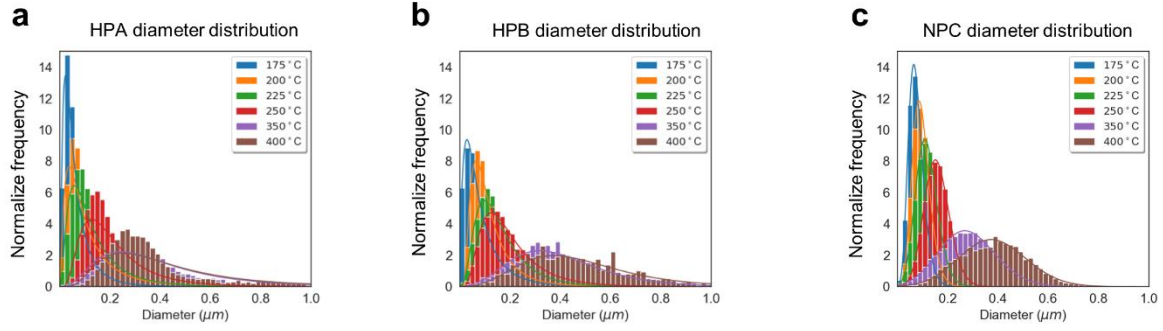
Manual segmentation of the images is performed with Avizo 3D software. It is a very time consuming and labor-intensive process so that we only segment the front, top, and left-most images of the 18 samples. We think this method is acceptable because we do not see the variability of segmentation quality within the same sample. For instance, the front, top, and left sides segmentation overlays, which are shown in Supplementary Figure 3, produce similar quality visually. They illustrate how the U-Net segmentation differs from manual segmentation and hybrid segmentation. In short, U-Net segmentation improves quality of the hybrid segmentation.



Supplementary Figure 3. Comparison of manual segmentation on the front, top, and left-most images with hybrid and U-Net like methods. Our hybrid method produces good segmentation but U-Net segmentation improves the segmentation quality. For instance, some segmentation artefacts are removed.

Supplementary Note 4: Strut diameter measurements.

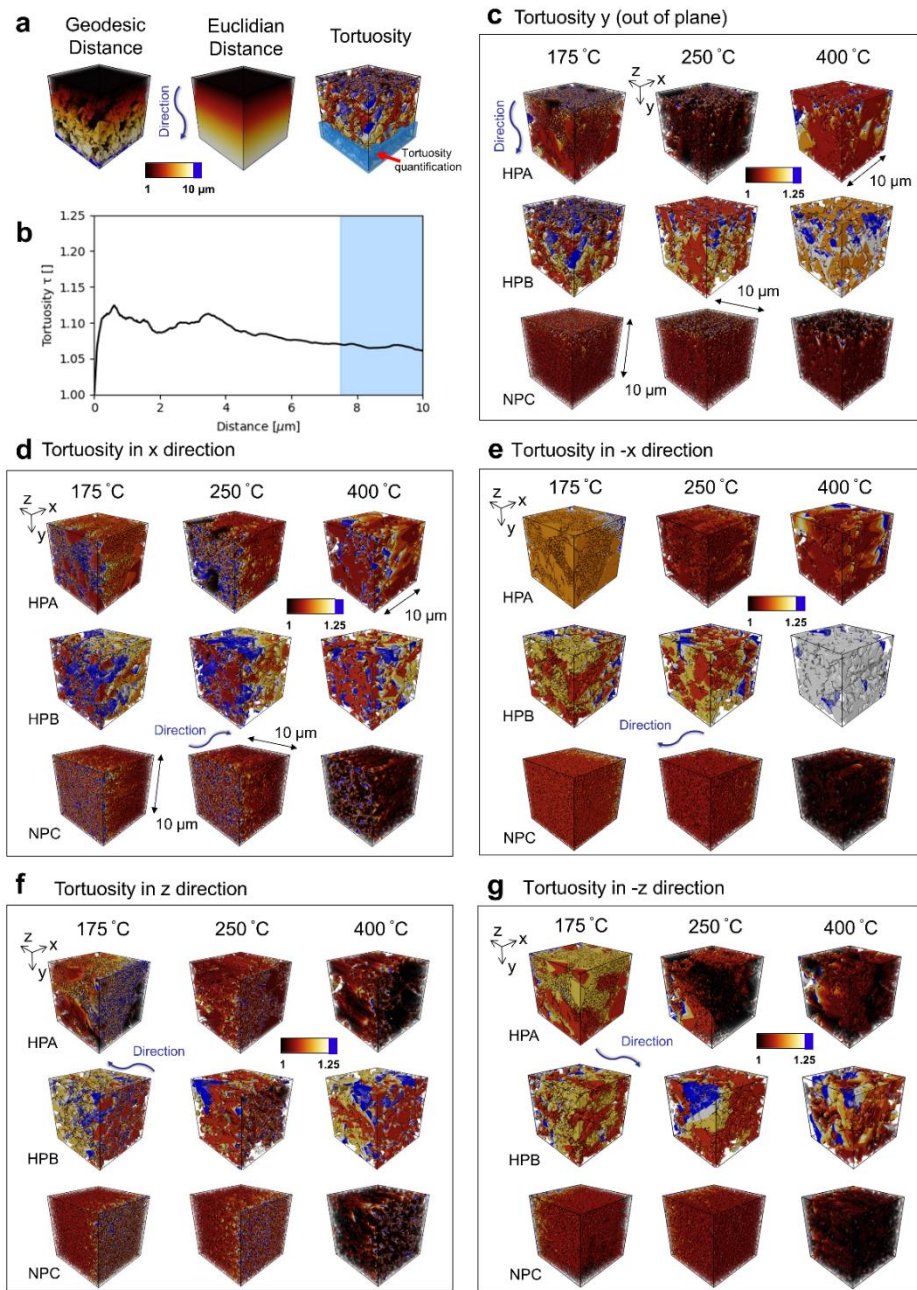
Supplementary Figure 4a-c presents the histogram of the strut diameter and the log-normal distribution. The fitted distribution contains the mean and the standard deviation.



Supplementary Figure 4. Strut diameter distribution. (a) The copper strut diameter distribution of HPA is fitted with a lognormal distribution. (b) The copper strut diameter distribution of HPB is fitted with a lognormal distribution. (c) The copper strut diameter distribution of NPC is fitted with a lognormal distribution.

Supplementary Note 5: Tortuosity Measurement.

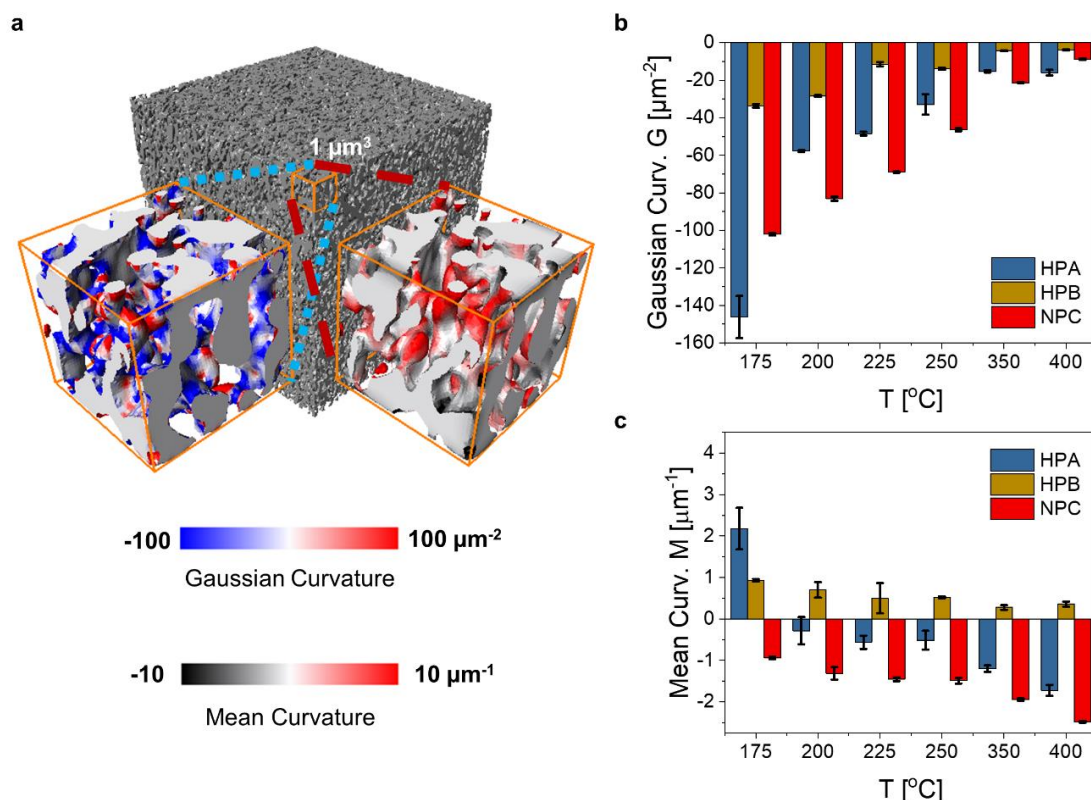
The geometric tortuosity τ^4 is a measure of the interconnectivity of copper materials. It is calculated by dividing the geodesic distance by the Euclidean distance (Supplementary Figure 5a). To determine the tortuosity, we take the average ratio of the last 25% of the sample's length⁵ (Supplementary Figure 5b) in five directions (x, y, z, -x, -z). The y-direction tortuosity is depicted in Supplementary Figure 5c, while Supplementary Figure 5d-g show the tortuosity in the x, -x, z, and -z directions, respectively.



Supplementary Figure 5. (a) Tortuosity is defined as a ratio of geodesic distance to Euclidean distance. (b) The tortuosity in the y direction is a distance function from the top surface. The last 25% tortuosity is averaged for quantification. (c) 3D tortuosity analysis in the y direction to quantify the connectivity of the copper along the direction from the surface to the substrate, with high tortuosity (blue) and low tortuosity (black). (d), (e), (f), and (g) show the x, -x, z, and -z directional tortuosity.

Supplementary Note 6: Gaussian and Mean Curvatures Measurements.

Supplementary Figure 6a illustrates the evaluated G and M for the copper interface. Figure 3d depicts the joint distributions of Gaussian and mean curvatures. The average data of both curvatures are shown in Supplementary Figure 6b and 6c.



Supplementary Figure 6. Gaussian and mean curvatures. (a) The changes in porous structures are described by Gaussian curvature and mean curvature measurements based on the segmented data. (b) Gaussian curvature measurements with respect to temperature for HPA (blue), HPB (gold), and NPC (red). (c) Mean curvature measurements with respect to temperature for HPA (blue), HPB (gold), and NPC (red). The data presented is the averaged curvatures of each sample.

Supplementary Note 7: New Features Generation from Existing Features.

Supplementary Figure 7a shows the impact of raw microstructural features on multi-variable linear regression (MVLRL) model by conducting SHapley Additive exPlanations (SHAP). We conduct a MVLRL model prediction with the top three features (specific surface area, mean curvature, and relative density). They do not predict the conductivity of the test samples well ($R^2 = 0.73$). Our goal is also to improve the accuracy of the test sample predictions. Supplementary Figure 7b illustrates the dependence of the conductivity on the temperature.

Models A, C, and I (Main text's Fig. 4a) consist only raw features and have good R^2 . They do not predict well in low and high conductivity regions (Main text's Fig. 4a); therefore, we engineer new features originated from raw features. They can be obtained from mathematical transformation of the raw features. We find two new features whose constituents are highly correlated with conductivity.

First, Bruggeman correlation⁶, $\tau = \text{Porosity}^{-\alpha}$, between tortuosity τ and porosity can be used. Porosity is replaced with relative density D because we measure the copper's tortuosity. Then, Bruggeman exponent α can be determined with the following formula:

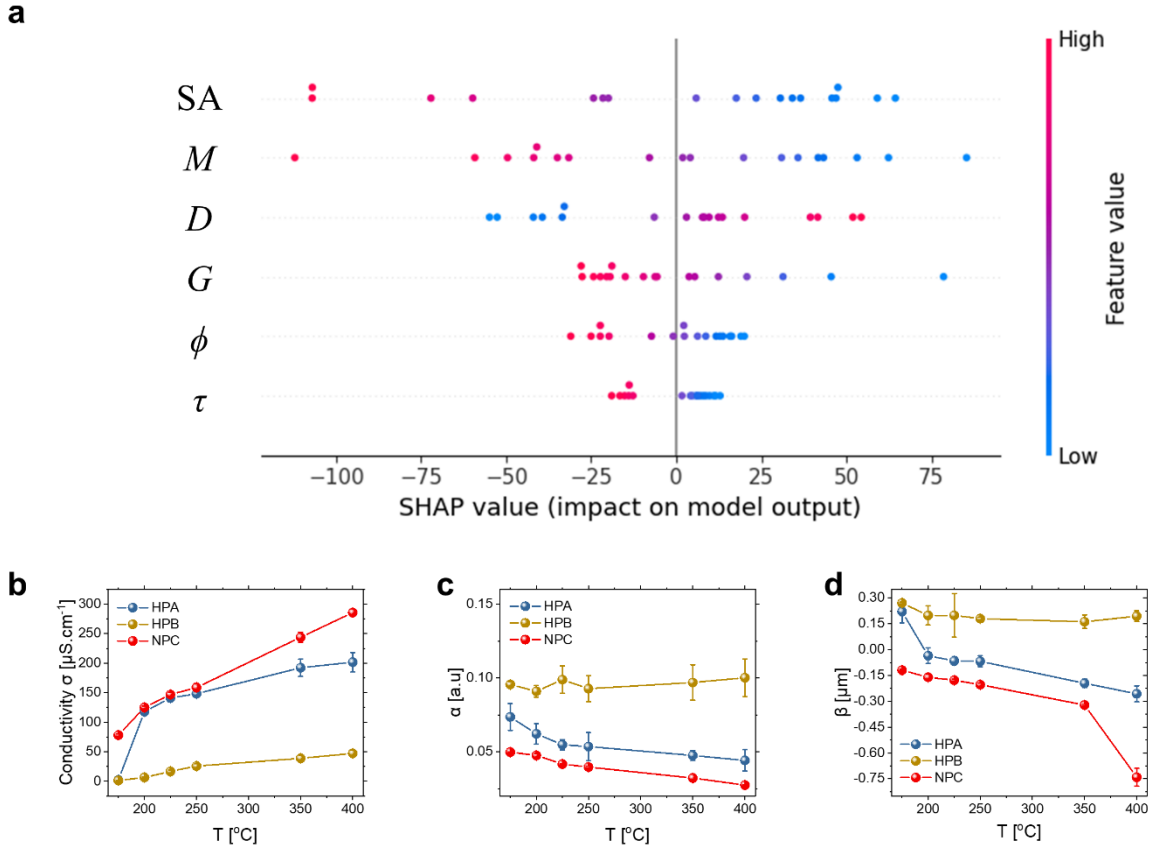
$$\alpha = -\left(\frac{\ln \tau}{\ln D}\right)$$

The plot of α with respect to temperature T is shown in in Supplementary Figure 7c.

Second feature β illustrated in Supplementary Figure 7d is

$$\beta = \frac{M}{|G| \cdot \phi}$$

Supplementary Figures 7b, 7c, and 7d depict some very interesting characteristics. α_{HPA} diverges at 175°C and β_{NPC} diverges at 175°C and 400°C. Supplementary Figure 7b shows the conductivity plot with respect to temperature. We observe there are divergences at 175°C for HPA and NPC and at 350-400°C for NPC. These characteristics help the multi-variable linear model to achieve linearity across the sintering process.



Supplementary Figure 7. New features generation from existing features. (a) The SHAP summary plot reveals the impact of microstructural features on the multi-variable linear regression model. **(b)** The conductivity trend with respect to temperature shows the divergence between HPA and NPC at 175°C and 250-400°C. **(c)** Bruggeman exponent α of each sample is plotted against the temperature. At lower temperature, the pattern depicts similarity with conductivity. **(d)** The second feature is $\beta = M / (|G| \cdot \phi)$. It shows the HPC's divergence at high temperature. The patterns observed in **(c)** and **(d)** are similar to conductivity. α and β definitely increase the conductivity prediction as shown in the main text.

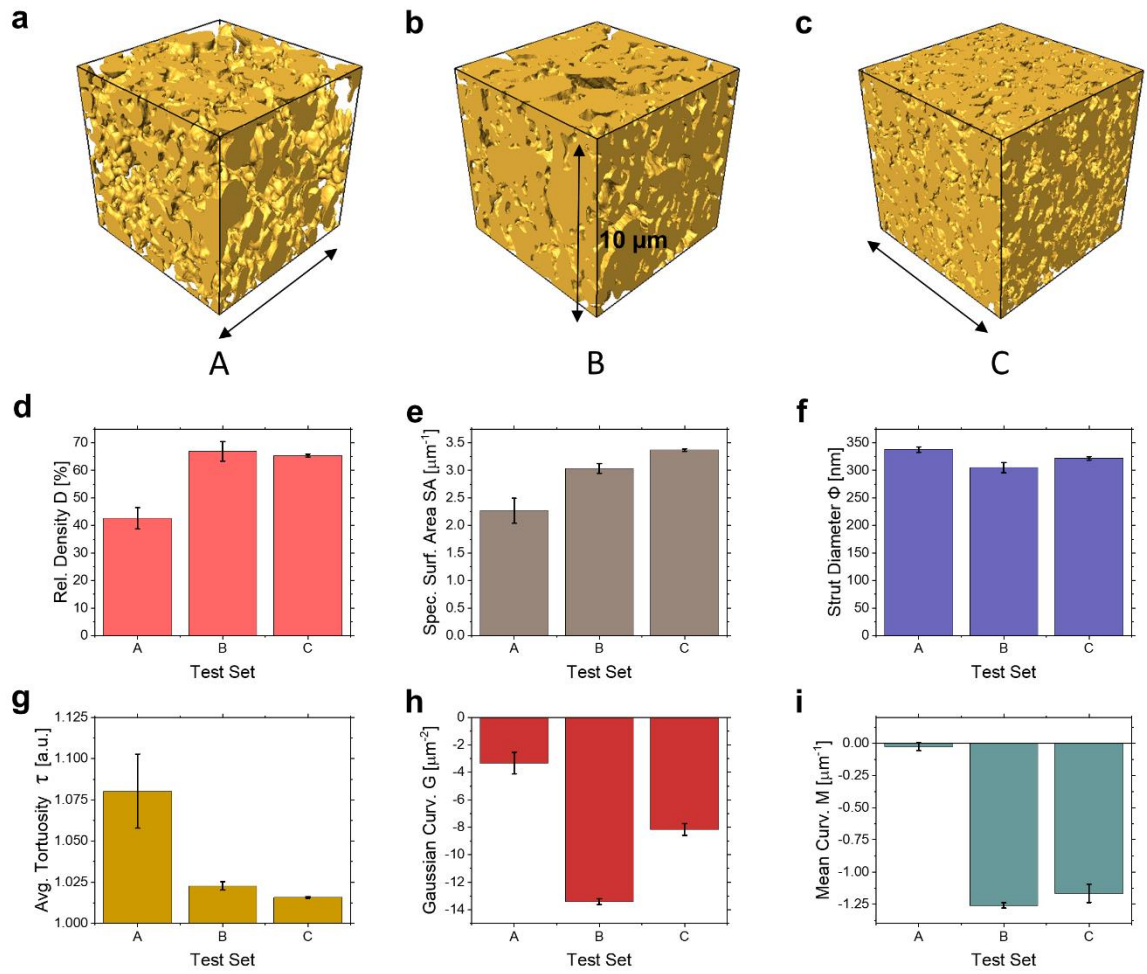
Supplementary Note 8: Multi-variable Linear Regression Models and the Test Set

The coefficient sign of the features with respect to the electrical conductivity. For instance, a plus (+) sign means that the feature in Supplementary Table 1 scales with the conductivity, a negative sign (-) means that it scales oppositely.

Supplementary Table 1. Features' regression coefficient signs.						
Raw Features	Relative Density D	Specific surface area SA	Skeleton diameter ϕ	Average Tortuosity T	Gaussian curvature G	Mean Curvature M
Sign of the Coefficient	+	-	+	-	+	-
Definition of the signs for different features' coefficients in the multi-variable linear regression model. A plus (+) sign means that the feature in Table 2 scales with the conductivity, a negative sign (-) means that it scales oppositely.						

There are many permutations to choose from eight features of the multiple linear regression models; therefore, only the relevant regression results that indicate leave one out cross validation and testing results are shown in Supplementary Table 2.

We also need to test the model with unseen data. There are three porous copper from our preliminary experiments in manufacturing porous copper. The porous structures of samples A, B, and C are similar to HPB, HPA, and NPC, respectively (Supplementary Figures 8a-c), but they do not share the same values as those of HPA, HPB, and NPC. We characterized with similar techniques in the main paper (Supplementary Figures 8d-i).



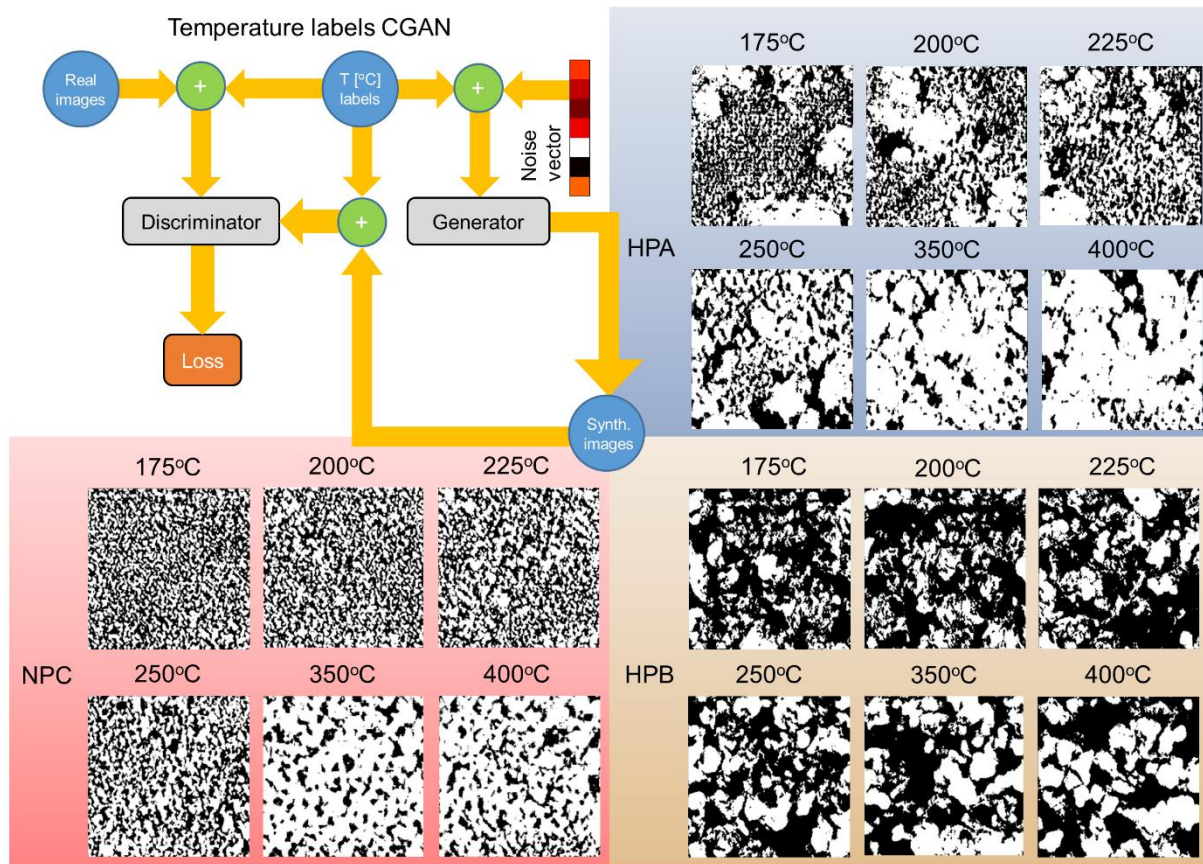
Supplementary Figure 8. Test set and their microstructural features characterization. (a), (b) and (c) represent the VOIs of test samples A, B, and C, respectively. (d) The relative density of test samples A, B, and C. (e) The specific surface area of test samples A, B, and C. (f) The strut diameter of test samples A, B, and C. (g) The average tortuosity of test samples A, B, and C. (h) The Gaussian curvature of test samples A, B, and C. (i) The mean curvature of test samples A, B, and C.

Supplementary Table 2. RMSE and R² of selected multi-variable linear regression models.					
Notation: D Relative density SA Specific surface area. ϕ Strut diameter τ Average tortuosity G Gaussian curvature M Mean curvature α Bruggeman exponent β $M / (G \cdot \phi)$					
Model	Features	Cross-Validation		Testing	
		RMSE	R ²	RMSE	R ²
A	τ, G	17.1	0.956	25.1	0.851
B	SA, τ	22.0	0.933	35.5	0.701
C	SA, τ, G	16.8	0.961	35.9	0.695
D	τ, G, M	14.7	0.970	30.6	0.778
E	ϕ, τ, G	16.6	0.962	37.1	0.674
F	D, τ, G	17.1	0.960	35.9	0.694
G	D, SA, M	12.0	0.980	33.8	0.730
H	SA, τ, M	13.5	0.975	29.5	0.793
I	SA, ϕ, τ, G	16.6	0.962	36.9	0.678
J	SA, α	11.4	0.982	25.1	0.851
K	α, β	25.3	0.911	15.4	0.944
L	SA, β	23.7	0.922	35.2	0.706
M	SA, ϕ, α	10.7	0.984	24.7	0.856
N	SA, G, α	11.4	0.982	25.1	0.851
O	SA, M, α	11.2	0.982	23.7	0.867
P	τ, SA, β	11.3	0.982	28.4	0.809
Q	SA, α, β	9.7	0.986	21.9	0.886
R	SA, M, ϕ, α	10.5	0.985	23.4	0.870
S	SA, G, ϕ, α	10.6	0.984	24.8	0.855
The calculation of RMSE and R ² of multi-variable linear regression models are shown according to cross validation and testing. The features column displays the features for each model.					

Supplementary Note 9: CGAN architecture and its mode collapse

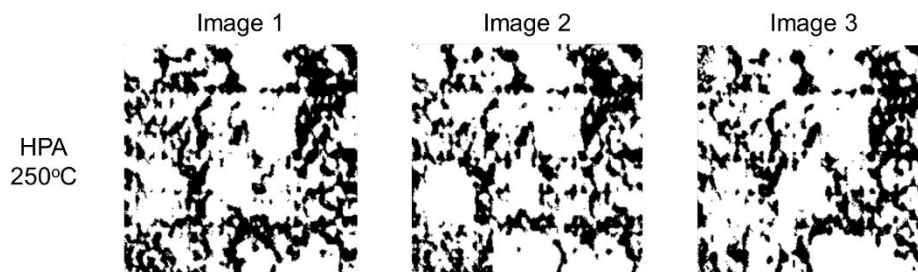
We resize the segmented 2D images to 224 x 224 pixels². Supplementary Table 3 and Supplementary Figure 9 show the architecture of generator and discriminator of the CGAN.

Supplementary Table 3. Generator and discriminator architecture					
Notation:					
k	Kernel size				
s	Strides				
p	Padding				
i	Number of classes				
Generator			Discriminator		
Layer	k, s, p	Output shape	Layer	k, s, p	Output shape
Input z	-	(200+i) x 7 x 7	Input	-	(1+i) x 224 x 224
1	4, 2, 1	256 x 14 x 14	1	5,2,1	128 x 112 x 112
2	4, 2, 1	256 x 28 x 28	2	5,2,1	128 x 56 x 56
3	4, 2, 1	256 x 56 x 56	3	5,2,1	128 x 28 x 28
4	4, 2, 1	256 x 112 x 112	4	5,2,1	128 x 14 x 14
5	4, 2, 1	256 x 224 x 224	5	5,2,1	128 x 7 x 7
6	7,1,1	1 x 224 x 224	Global Max pooling	-	1
The noise vectors of z have the size of 200. The number of classes are depending on the bin size selection. We use LeakyReLU activation with alpha = 0.2 in both generator and discriminator except for the generator output, which uses Tanh. Batch Norm is also used in the generator and the discriminator. The optimizers for the generator and the discriminator are Adam with learning rate 0.0004 and 0.0001, respectively. The loss is binary cross-entropy.					



Supplementary Figure 9. CGANs generated images with temperature labels. CGANs with 6 labels produce inaccurate microstructure and lower quality image. On the other side, the images of CGANs with σ bins are smoother and more accurate (see the main text).

The generator sometimes produces similar microstructure with different noise vector input⁷. It happens after extended training steps for all materials. Supplementary Figure 10 depicts one example of the mode collapse.



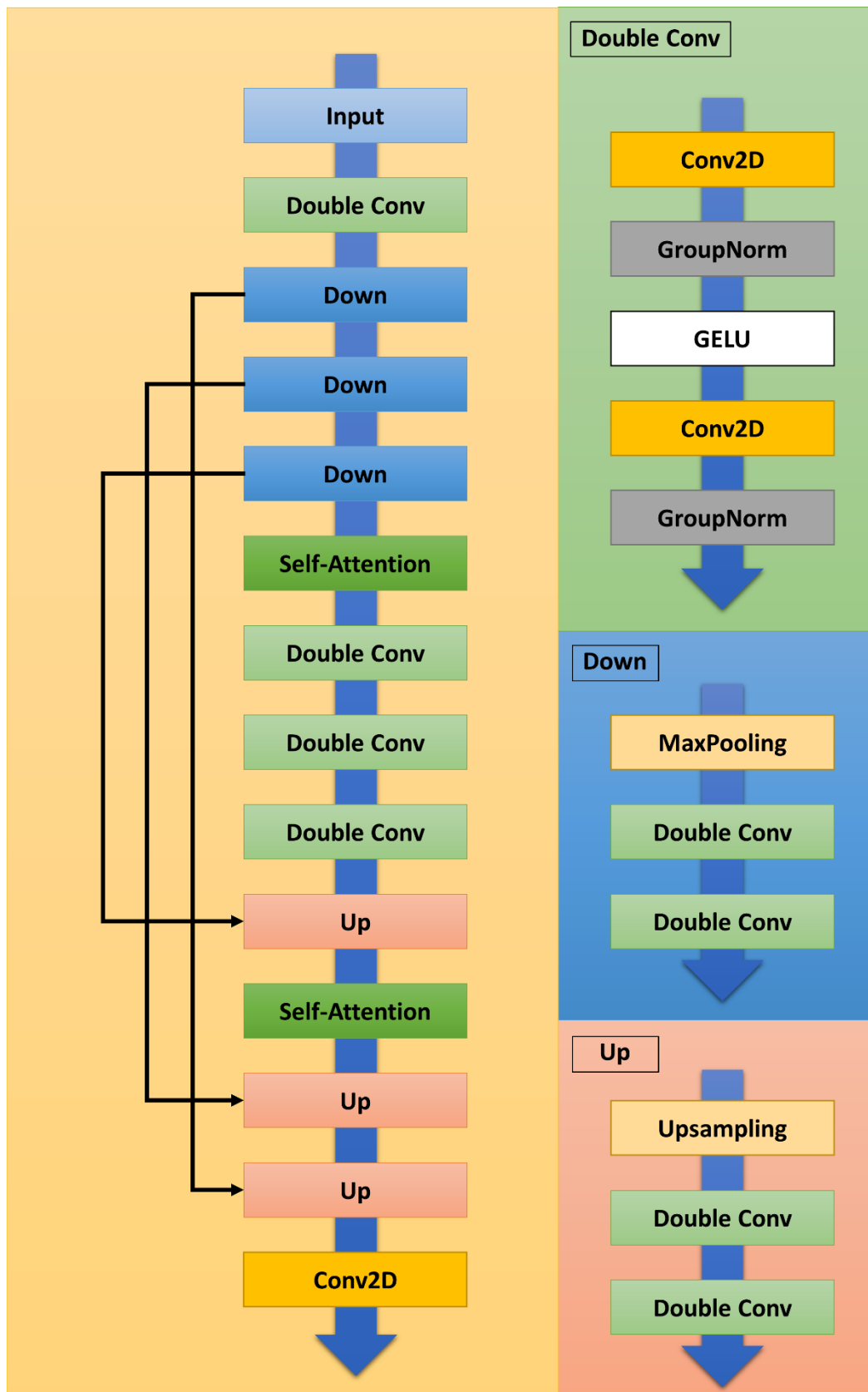
Supplementary Figure 10. Mode collapse example. HPA's CGAN fails to produce different microstructure.

Supplementary Note 10: DDPM architecture

The implemented DDPM generative model^{8,9} is the modification of this diffusion models GitHub page¹⁰. The time step t for noise is set to 600, while the variance scheduler β starts at 0.0001 and ends at 0.02. The batch size is 32. The U-Net architecture is used to determine the noise distribution. Supplementary Figure 11 provides a complete overview of this U-Net.

The encoding part comprises of three blocks of down-sampling blocks. Each block consists of a max-pooling layer followed by two double convolutional blocks. Each double convolutional block includes a convolutional layer, a group normalization layer, a Gaussian Error Linear Unit (GELU) activation function, another convolutional layer, and another group normalization layer. After this, three double convolution blocks are applied, followed by an up-sampling block. An up-sampling block consists of a bi-linear up-sampling and two double convolutional blocks. The self-attention mechanism with four heads is used after the final down-sampling layer and the first up-sampling block.

The decoder is almost identical to the encoding architecture, except that after each up-sampling layer, the data is concatenated with skip connections corresponding to channels with the same resolution as the encoding part. The model is conditioned on the time step by the sinusoidal positional embedding¹¹, which is added to each down-sampling and up-sampling layer. Class labelling is included in the same way as the time step by adding a class embedding to the time step embedding.



Supplementary Figure 11. The U-Net architecture of the denoising diffusion probabilistic model to find the noise distribution.

Supplementary References

1. Ronneberger, O., Fischer, P. & Brox, T. U-Net: Convolutional Networks for Biomedical Image Segmentation. *Med. Image Comput. Comput. Interv.* **9351**, 234–241 (2015).
2. Chollet, F. Image segmentation with a U-Net-like architecture.
https://keras.io/examples/vision/oxford_pets_image_segmentation/ (2019).
3. Chollet, F. Xception: Deep learning with depthwise separable convolutions. *Proc. - 30th IEEE Conf. Comput. Vis. Pattern Recognition, CVPR 2017* **2017-Janua**, 1800–1807 (2017).
4. Clennell, M. Ben. Tortuosity: a guide through the maze. *Geol. Soc. Spec. Publ.* **122**, 299–344 (1997).
5. Taillon, J. A., Pellegrinelli, C., Huang, Y. L., Wachsman, E. D. & Salamanca-Riba, L. G. Improving microstructural quantification in FIB/SEM nanotomography. *Ultramicroscopy* **184**, 24–38 (2018).
6. Ebner, M., Chung, D. W., García, R. E. & Wood, V. Tortuosity anisotropy in lithium-ion battery electrodes. *Adv. Energy Mater.* **4**, 1–6 (2014).
7. Amiri, H., Vasconcelos, I., Jiao, Y., Chen, P. E. & Plümper, O. Quantifying microstructures of earth materials using higher-order spatial correlations and deep generative adversarial networks. *Sci. Rep.* **13**, 1–19 (2023).
8. Ho, J., Jain, A. & Abbeel, P. Denoising diffusion probabilistic models. *Adv. Neural Inf. Process. Syst.* **2020-Decem**, 1–25 (2020).
9. Azqadan, E., Jahed, H. & Arami, A. Predictive microstructure image generation using denoising diffusion probabilistic models. *Acta Mater.* **261**, 119406 (2023).
10. Rampas, D. Diffusion Models. <https://github.com/dome272/Diffusion-Models-pytorch>.
11. Vaswani, A. *et al.* Attention is all you need. *Adv. Neural Inf. Process. Syst.* **2017-December**, 5999–6009 (2017).