Super-resolution of multiphase materials by combining complementary 2D and 3D image data using generative adversarial networks

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Abstract

Modelling the impact of a material’s mesostructure on device level performance typically requires access to 3D image data containing all the relevant information to define the geometry of the simulation domain. This image data must include sufficient contrast between phases to distinguish each material, be of high enough resolution to capture the key details, but also have a large enough field-of-view to be representative of the material in general. It is rarely possible to obtain data with all of these properties from a single imaging technique. In this paper, we present a method for combining information from pairs of distinct but complementary imaging techniques in order to accurately reconstruct the desired multi-phase, high resolution, representative, 3D images. Specifically, we use deep convolutional generative adversarial networks to implement super-resolution, style transfer and dimensionality expansion. To demonstrate the widespread applicability of this tool, two pairs of datasets are used to validate the quality of the volumes generated by fusing the information from paired imaging techniques. Three key mesostructural metrics are calculated in each case to show the accuracy of this method. Having confidence in the accuracy of our method, we then demonstrate its power by applying to a real data pair from a lithium ion battery electrode, where the required 3D high resolution image data is not available anywhere in the literature. We believe this approach is superior to previously reported statistical material reconstruction methods both in terms of its fidelity and ease of use. Furthermore, much of the data required to train this algorithm already exists in the literature, waiting to be combined. As such, our open-access code could precipitate a step change in the computational materials sciences by generating the hard to obtain high quality image volumes necessary to simulate behaviour at the mesoscale.

1 Introduction

The geometrical arrangement of porous or composite materials at the mesoscale can significantly impact the performance of the devices they constitute. In order to accurately model the physical processes that mediate this relationship, it is necessary to have geometric information about the distribution of the various phases. This is typically derived from imaging data, which must have the following four properties: Firstly, it must be three-dimensional (3D), since many percolation networks are not reducible to two dimensions. Secondly, the image data must be of high enough resolution to capture the key details. Thirdly, it must have a large enough field-of-view to be representative of the material in general. Finally, it must be possible to confidently differentiate between each of the material phases present. It is rarely possible to obtain data with all four of these properties from a single imaging technique, meaning that the resulting simulation domains may be lacking some vital information, which can undermine the value of any conclusions drawn from these numerical analyses.

Lithium-ion battery cathodes exemplify this kind of challenging imaging scenario. They are usually porous composites where the solid phase is comprised of a

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ceramic “active material” (AM) that is a lithium intercalation compound (e.g. nickel manganese cobalt oxide or NMC), as well as a polymeric binder material containing carbon black called the “carbon binder domain” (CBD) which provides both mechanical integrity and electronic conductivity. The pores are filled with a liquid electrolyte and must be well percolated to facilitate transport of the dissolved lithium ions. These electrodes are typically around 100 μm thick with particle size distributions varying along their thickness; whereas the CBD may have relevant features below 100 nm that can critically impact performance.

Imaging these cathode materials in 3D using X-ray computed tomography (XCT) enables a large, representative field-of-view to be observed; however, current XCT technology does not provide sufficient contrast for the CBD phase to be confidently differentiated from the pores\(^\text{7}\). Furthermore, even if the contrast could be enhanced, the resolution would be insufficient (c. 100 nm) to observe the CBD’s nanoscale features\(^\text{19}\). It is also common to image cathode materials with focused ion beam scanning electron microscopy (FIB-SEM), which can provide much higher resolution data (c. 10 nm), but the imaged regions are usually quite small as a result. SEM is better suited than XCT at distinguishing between the solid phases (i.e. AM vs. CBD); however, since SEM is based on reflected electrons, it is difficult to distinguish between material that is on the desired imaging plane compared to the inner surface of pores. This “pore-backs” problem has several proposed solutions, including infiltration\(^\text{16}\) and advanced image tracking\(^\text{20}\), but remains a significant challenge. An additional common limitation of FIB-SEM generated volumes is that uniform spacing in the “slicing” direction is hard to guarantee\(^\text{11}\) and so any observed anisotropy may be artificial.

The 2D SEM imaging technique is, however, far better at differentiating the CBD phase through the use of various contrast enhancement techniques\(^\text{15}\). However, as highlighted in\(^\text{7}\), for isotropic materials, although the statistical information present in a representative two-dimensional (2D) slice is sufficient to fully define the material, 3D domains are still required to extract certain key performance metrics, such as percolation and the tortuosity factor. Therefore, the successful fusion of the valuable information found in pairs of complementary 2D and 3D images of a material is highly desirable.

To close this gap, our proposed method achieves an accurate reconstruction of high resolution (high-res), large field-of-view and multi-phase 3D volumes by fusion together information from complementary 2D and 3D imaging techniques. For this, we use a machine learning framework called generative adversarial networks (GANs)\(^\text{6}\). GANs have previously been used to 1) enhance the resolution of (“super-resolve”) colour photographs\(^\text{17}\) and 2) transfer style between images (e.g. converting a photograph to a painting)\(^\text{13}\). Furthermore, Kench and Cooper recently published a GAN architecture able to 3) generate 3D volumes from a single representative 2D slice\(^\text{14}\). We propose a GAN architecture that combines these 3 capabilities, with an architecture designed specifically for the generation of materials mesostructure phase maps.

Unlike the state-of-the-art in this field\(^\text{9}\), our approach does not require the user to select and extract characteristic metrics in order to generate or modify their images, but instead it learns the key features and relationships directly from the data. Also, unlike a recent paper\(^\text{12}\) that presents a 2D to 2D super-resolution (super-res) mesostructure technique, our approach not only super-resolves 3D mesostructure, but also is able to add missing phases, change existing ones and capture anisotropy.

To illustrate the capability and accuracy of the proposed method, we present four case studies, each based on open-access imaging data. In each study, the paired datasets used for training consist of high-res 2D images and a low-resolution (low-res) 3D volume, with the aim of producing a super resolved (super-res) data with all the best properties of each. The first two studies serve to validate the accuracy of the method in two distinct scenarios, the third explores the performance across a range of user demands, and the fourth study demonstrates the application of the approach on real paired datasets from the literature. Since the first three case studies require access to the ‘ground truth’ in order to compare the calculated metrics, the low-res volumes were derived directly from down-sampling the original high-res versions and in some cases removing phases or features to add realism. More details about the training data and how it was prepared for analysis can be found in the Supplementary Information Table 1.

1. Battery cathode (validation on an isotropic material with abundant training data): A low-res, large field-of-view volume showing only two of the three phases, is combined with a high-res three-phase 2D image to produce a large, 3D, three-phase super-res volume. The material is isotropic and training data is abundant. The ground truth is known as the low-res is created from the high-res and detailed statistical comparisons are made.

2. Battery separator (validation on anisotropic material with limited training data): A low-res, large field-of-view, two-phase volume is combined with a set of
Figure 1: The SuperRes model inputs and output evaluated on the isotropic NMC cathode XCT dataset (Case study 1)\footnote{22}. The different phases are pore (black), active material (grey) and binder (white). The top row shows the inputs and output of the model. For training, the model requires a high-res multi-phase 2D image and a low-res binary 3D volume, and the output is a super-res volume of the low-res volume with the same multi-phase high-res 2D fine characteristics. The bottom row shows a comparison between a random sample of a cross section in the same position of the low-res and super-res volumes, and how the super-res cross section fine details are compared with the high-res slice subsection.

orthogonal high-res 2D images containing fine features to produce a large, 3D, super-res volume. The material is anisotropic and training data is limited. The ground truth is known as the low-res is created from the high-res and detailed statistical comparisons are made.

3. Fuel cell anode (exploration): To explore different scale-ratios between low-res and high-res, several LR, large field-of-view volumes showing three phases each combined with a high-res three-phase 2D image to produce a large, 3D, three-phase, super-res volume. The material is isotropic and training data is abundant. The ground truth is known as the low-res is created from the high-res and visual comparisons are made.

4. Battery cathode (demonstration): A low-res, large field-of-view XCT volume showing only two phases is combined with a high-res three-phase 2D SEM image to produce a large, 3D, three-phase, super-res volume. The material is isotropic and training data is limited. The ground truth is not known. To the authors knowledge, this is the highest resolution Li-ion cathode mesostructure volume dataset that contains the binder phase.

The mesostructures that are examined in this paper are all battery or fuel cell related, hence the relevance of the mesoscale transport properties that are extracted. However, our model is material and lengthscale agnostic and can be applied to diverse scenarios including catalyst beds, rock formations, or snow and soil packing.

2 Results

2.1 Validation using an isotropic, three-phase cathode dataset

To illustrate the method’s capabilities for isotropic, three-phase materials with abundant training data, an open-access dataset derived from XCT imaging of a lithium-ion battery cathode material was used\footnote{22}. This dataset was produced by XCT imaging which then has the binder phase added stochastically based on statistical information from an SEM image. Figure 1 illustrate the workflow and effectiveness of the model. The different phases of the cathode shown are pore (black), active material (grey) and binder (white). In addition to demonstrating the generated super-res of the low-res
volume, this scenario also exhibits the in-painting of a phase present only in the high fidelity 2D slice.

For this experiment, in order to have a very large volume to showcase the power of the approach, a 3D high-res three-phase volume of $1024^3$ voxels was generated using a SliceGAN\textsuperscript{14} trained on the three-phase volume from\textsuperscript{22}. The low-res volume was then created from this high-res volume by first merging the pore and binder phases together and then down-sampling the volume by a factor of 4 to $256^3$ voxels. The same SliceGAN model was also used to independently generate a 2D high-res three-phase slice containing $2048^2$ pixels. As explained in the introduction, the decision to merge the pore and binder phases in the low-res volume stems from the fact that they are very difficult to distinguish using XCT due to the very low X-ray attenuation of the binder compared to the active material.

Figure 2 shows a quantitative comparison between the original high-res volume and the super-res volume. In the metrics comparison, the ultimate objective is for the super-res volume to be in good agreement with the 3D high-res ground truth volume, but since it only sees the 2D image as the high-res input, the agreement of the metrics depends on the representativeness of the 2D image to the ground truth volume. As can be seen, except for the pore/binder interphase surface area, the 2D high-res image is representative of the properties of the 3D volume, thus the super-res volume is in good agreement with the ground truth volume on all of these metrics. Interestingly, since the 2D high-res image is the only information for the high-res binder characteristics for the reconstruction, the super-res volume has good agreement with it for the Pore/Binder interphase surface area, slightly more than the 3D original volume. This good agreement with the 2D high-res image highlights the accuracy of the proposed method and the need of a representative 2D slice for an accurate 3D reconstruction.

2.2 Validation using an anisotropic, two-phase separator dataset

To present the flexibility and diverse capabilities of the method, an anisotropic material was also investigated. For this, an open-access dataset of a porous polymer battery separator material was chosen\textsuperscript{24}. In the previous example, a new phase was introduced in the 2D high-res slice that was not present in the 3D low-res volume. However, an alternative scenario is that the same phase displays new features when imaged at higher resolution. The separator material consists of thick lamellae connected by thin fibrils.
Figure 3: The model results for an anisotropic material, here producing a super-res 3D battery separator material. The different model inputs and outputs are the same as in Figure 1, with the additional input of high-res 2D slices from all three perpendicular directions. Since two perpendicular directions have the same properties (upper slice in the top left corner), for simplicity only two high-res 2D slices are shown out of the 6 facets used for training data.

Although the XCT data collected by Finegan et al. was able to capture the lamellae, it did not have sufficient resolution to observe the fibrils. However, Xu et al. used a stochastic method to add fibrils based on statistical information taken from an SEM image. In this experiment, the high-res 3D separator data is 624×300×300 voxels of the lamellae and stochastically added fibrils. The low-res volume is a down-sampling of only the lamellae with a scale-factor of 4. Together with the low-res volume, the input also consists of high-res 2D slices, which is the set of all 6 outer facets of the high-res 3D volume (analogous to capturing 2D images of the outer surfaces of a volume). These slices together sum to less than 1 megapixel of data, which is a very limited amount of image training data compared to the amount generally required for deep learning imaging applications.

As can be seen in Figure 3 and the corresponding metrics comparison Figure 4, although the limited training high-res 2D data may have slightly harmed the agreement of the metrics, the facets of the original high-res volume had enough high-res information for the successful anisotropic super-res reconstruction. As in case study 1, the surface area of the high-res 2D slice was offset from the global 3D high-res value. The surface area of the 3D super-res volume more closely matches the high-res 2D value, demonstrating the ability of the generated volume to accurately reproduce the statistics of the training data.

2.3 Scale-Factors Exploration using fuel cell anode data

As described in more detail in Section 3, the model is able to support a variety of different scale-ratios between the resolutions of the two input images. Specifically, the model supports all rational scale-factors in the range between 1 to 8 that divide 64 without a remainder, which are all scale-factors $\frac{64}{d}$ for any integer $d$ in the range $8 \leq d \leq 64$. This set of scale-factors can be expanded and is only limited by current implementation.

For the exploration case study, an open-access FIB-SEM image dataset of a solid oxide fuel cell anode material was chosen. As can be seen in Figure 5, this material has three phases, pore (black), metal (grey) and ceramic (white). In this case, all three phases are preserved when down-sampling the original data to generate the low-res volume and are therefore also present.
Figure 4: Statistical comparison of key mesostructural metrics between the original high-res 3D volume, the high-res 2D slices and the reconstructed super-res 3D volume for the anisotropic separator material in Figure 3. The metrics description is the same as in Figure 2. Since the material is anisotropic, transport efficiency measurements were taken along all different axes.

| Scale factor | 1.6 | 2 | 4 | 8 |
|--------------|-----|---|---|---|
| Low res.     | ![Image](image1.png) | ![Image](image2.png) | ![Image](image3.png) | ![Image](image4.png) |
| Super res.   | ![Image](image5.png) | ![Image](image6.png) | ![Image](image7.png) | ![Image](image8.png) |
| Original     | ![Image](image9.png) | ![Image](image10.png) | ![Image](image11.png) | ![Image](image12.png) |

Figure 5: The results from exploring the effect of different scale-factors. For each scale-factor, the inputs for the model were a high-res 2D slice and a low-res volume which was down-sampled accordingly from the high-res volume. For ease of visualisation, the center of the 2D high-res image can be seen on the right, and the same position sub-sample of the low-res volumes can be seen on the top row. In each experiment, these inputs were fed into the architecture, and the super-res output volumes are presented in the middle row, with the same sub-sample positions of the low-res volumes. The bottom row shows the same location of the sub-samples from the original high-res volumes for comparison.

It is clear from Figure 5 that as the scale-factor increases, it is harder to reconstruct the exact original high-res volume before it was down-sampled, due to the loss of information and the large number of possible realistic reconstructions. Importantly, however, both objectives of the generated image were met: the super-res volume has a similar phase map to the low-res volume and the properties of the super-res volume are...
Figure 6: The model results for a demonstration isotropic material, combining XCT low-res volume and SEM high-res 2D slice, resulting in a super-res volume with nanoscale resolution that is hard to impossible to achieve with current imaging techniques.

similar to the high-res volume. This can be seen in all different scale-factor experiments, and the similarity of the high-res features is evident in comparison with the high-res 2D image training data that appears on the right side of the figure.

2.4 Demonstration using a pair of complimentary datasets from battery cathode

The previous case studies were designed to validate the accuracy of the method using experiments that possessed the ground truth high-res 3D volume alongside the input images. This way the generated super-res volumes can then be quantitatively compared to the ground truth. Once the performance has been validated, a natural next step is to test the method on a real-world problem, where the desired ground truth high-res 3D volume does not exist, but all of the necessary information is contained in a pair of complimentary datasets.

For the demonstration case study, the same battery cathode dataset is used as in the first case study. As mentioned in the first case study, the binder phase in this dataset was added stochastically to the XCT imaged volume based on statistical information from an SEM image of the same material. In this case study, we will use the XCT dataset without the added binder phase as the low-res input, leaving only the confidently distinguished active material phase as can be seen in middle top of Figure 6.

The SEM image of the same cathode sample used to extract the binder statistics in will be used as the high-res 2D input. The high-res SEM image offered high enough contrast to distinguish between the CBD phase from the pores, allowing it to be segmented in three-phase image using Weka segmentation software accompanied by some manual finishes, to try and account for the ‘pore-backs’ problem described in the introduction. The entire three-phase segmentation result can be seen in the upper left corner of Figure 6, while the original SEM image with segmentation overlay can be seen in the Supplementary Information Figure 9.

Other than a real representation of the CBD phase, another difference to the first case study is the resolution and the scale-factors used. While in the first case study the low-res volume had a voxel lengthscale of 1.6 µm and the high-res 2D slice had a pixel lengthscale of 400 nm (scale-factor of 4), in this case study the low-res volume has a voxel lengthscale of 400 nm and the high-res 2D slice has a pixel lengthscale of 50 nm (scale-factor
of 8). To the authors knowledge, this is the highest resolution Li-ion cathode mesostructure volume dataset that contains a realistic representation of all three phases.

We mention that although the SEM image had sufficient information to observe the CBD’s nanoscale features, it had many unwanted artifacts that damaged the true distribution of the phases in the image. As well as the ‘pore-backs’ issue, which may cause the binder to appear over represented in the SEM image, an additional artefact referred to as ‘curtaining’ can occur when FIB is used to prepare cross-section for SEM imaging. Vertical streaks are formed by the ion beam milling through phases with different densities, which can make segmentation more challenging. This was not totally accounted for during the manual correction of the segmentation and can be most clearly seen in the bottom right of the 2D input image in Figure 6. However, we find that this anisotropic artifact is hardly found in the output super-res volume. This positive finding can be linked to the isotropic data augmentation we performed as a preprocessing step, which will be described in the next section.

3 Methods

The SuperRes model was built upon the development of SliceGAN, a generative adversarial network architecture that generates 3D mesostructure from a 2D slice. In SliceGAN, the input of the Generator (G) is random noise and the output is a one-hot encoded 3D mesostructure which is then sliced orthogonal to x, y and z axes to feed the Discriminator (D) with 2D images. Here, as well as feeding G with random noise, the input of G also contains a low-res 3D mesostructure which we wish to upsample and augment. The architecture of G is changed accordingly, whilst D is kept the same. Although it is a reasonable assumption that G would learn to use the low resolution input structure to generate a high resolution reconstruction with similar features, this is not guaranteed to be the case. This is because the standard GAN loss function does not include a measure of the similarity between the input and output image. It is thus possible for the generated 3D images to look very different from the low-res image.

To constrain G to generate images that will have the same features as the input image, we add a loss term given by the voxel-wise mean squared error (MSE) between the low-res image and a down-sample of the super-res image, as outlined in Algorithm 1 and in the Supplementary Information Figure 8. Since the low-res input to G is a segmented volume, after down-sampling the generated volume a sigmoid function was applied instead of a threshold (step) function to result in a near one-hot encoded phase map and keep the pixel-wise loss differentiable. Other than the addition of the voxel-wise loss, the Wasserstein loss function remains similar to SliceGAN.

Data augmentation can increase the apparent diversity of the training set and therefore improve the quality of the trained model by reducing overfitting. This is particularly important when training data is hard to acquire and scarce. For example, if the material is assumed to be isotropic then a single 2D slice can be transformed into 8 slices of the same size, constituting of all the different mirrors and 90° rotations of the original image. The overfitting of D on the high-res training data can disrupt the learning process of G. This failure mode will enforce G to generate volumes with high voxel-wise loss to the input volumes, in search of D’s structural space, harming the super-res property. It should also be noted that the 2D high-res training data does not need to come from a single large image, but can be from a collection of smaller images, which may be much easier to obtain.

The voxel-wise loss between the low-res and super-res volumes is only introduced after a threshold of 0.5% difference has been met. This threshold is introduced at a later stage to provide flexibility to the super-res output to distance from the blocky corners of the phases present in the low-res volume. The concatenated input noise layer gives flexibility to G and is necessary for a successful reconstruction, as shown in the Supplementary Information Figure 10.

The algorithm presented above is for isotropic materials, for anisotropic materials, a minor change in the algorithm is needed with 3 different discriminators for the x-y, y-z and x-z slices as described in SliceGAN. Note that the last step of the algorithm output is possible due to the convolutional design of the generator that is detailed in Supplementary Information Figure 7, that allows for any size of input volume.

4 Discussion

The results of the four case studies make a strong case for the utility and potential impact of this GAN based super-resolution method. The first and second case study demonstrated excellent agreement between the metrics calculated on the 3D high-res ground truth and super-resolution output. Notably, there was strong agreement of transport efficiency which is an inherently 3D property, highlighting the power of this method to perform both dimensionality expansion as well as data fusion, as this property could not be accurately calcu-
Algorithm 1 SuperRes algorithm for isotropic materials

**Require:** $G$, the generator function; $D$, the discriminator function; $LR$, the low-resolution volume; $HR$, the high-resolution 2D slice; $sf$, the scale-factor between $LR$ and $HR$; $b$, threshold for the voxel-wise loss (default 0.01); $c$, the coefficient of the voxel-wise loss multiplication (default 10); $\sigma$, a trilinear-downsample with a scale-factor of $sf$ and a sigmoid function; All batch operations and optimization and gradient penalty parameters are not shown for simplicity.

**Preprocessing:** $HR \leftarrow$ Data augmentation step, expanding $HR$ into all 8 different possible mirrors and rotations of $HR$ by 90° multiples.

1: while weights of $G$ did not converge do
2: $lr \leftarrow$ Sample a $(\frac{64}{3})^3$ voxels cube uniformly from $LR$.
3: $z \leftarrow$ Sample a $(\frac{64}{3})^3$ voxels cube of noise from normal distribution with mean 0 and standard deviation 1.
4: $lr \leftarrow$ concat($lr, z$) Concatenate the low-res and noise cubes along the phase dimension.
5: $sr \leftarrow$ $G(lr)$ Generate a 64³ super-res volume.
6: $sr_{slices} \leftarrow$ slice($sr$) Slice the volume output $sr$ in all perpendicular planes into 64 · 3 2D slices.
7: if Training of $G$ (Only every 5 iterations) then
8: $l_w \leftarrow$ MS $E(lr, \sigma(sr))$ Voxel-wise loss between low-res and a downsample of super-res volumes.
9: if $l_w < b$ then
10: $l_G \leftarrow -D(sr_{slices})$
11: else
12: $l_G \leftarrow -D(sr_{slices}) + c \cdot l_w$
13: Backpropagate and update the weights of $G$ from the loss $l_G$.
14: else Training of $D$
15: $hr \leftarrow$ Sample a 64² pixels square uniformly from $HR$.
16: $lp \leftarrow$ GP($sr_{slices}, hr, D$) Calculate the gradient penalty regularization based on real and fake outputs.
17: $l_D \leftarrow$ $D(sr_{slices}) - D(hr) + l_p$
18: Backpropagate and update the weights of $D$ from the loss $l_D$.

return The super-resolution volume of the full low-resolution volume $G(LR)$.

There is a small discrepancy between some of the 3D high-res and 3D super-res metrics, with the super-res metrics more closely aligned to the 2D high-res metrics rather than 3D high-res metrics, especially for properties that are only contained in the high-res 2D training data, such as the binder phase or the thin fibrils. These results illustrate how the model is learning the distribution of the high-res training data whilst constrained by the super-resolution process, as the bulk distribution is enforced by the low-res input, but the high fidelity details are purely learned from the 2D high-res slice.

In the last demonstration case study it is clearly visible that the binder phase in the super-res volume matches the binder phase in the SEM image shown in Figure 6, showing some particularly intricate geometry. Furthermore, the contrast to geometry of the binder phase that was added statistically (based on metrics from the same SEM image) in the open-access data as shown in Figure 1 is stark.

It is interesting to note that it is possible to implement this super-res method using just a low-res volume as the input to $G$, without concatenating a layer of noise. In this scenario, the model is deterministic and if it is well trained, its output should represent a likely underlying mesostructure associated with each neighbourhood of the input. Yet, the addition of noise offers three advantages over the deterministic approach:

- Limiting $G$ to a single output for a given low-res input could lead to unwanted homogeneity of the resulting mesostructure, especially when the low-res input contains many repeated patterns. This is exemplified for an input featuring only one phase, as shown in Supplementary Information Figure 10. Unlike the heterogeneity of features generated with the stochastic input, without added noise $G$ is only able to output a single pattern across the pore volume, resulting in a failure mode. The figure also shows the scenario with large patches of pore and a similar but more complex failure mode is observed.

- There are two ways in which overfitting of $D$ can occur, either $D$ learns the true dataset as described in the Architecture section, or $D$ can learn the dataset which comprise of the output distribution of $G$. In this scenario, after $D$ learns $G$’s outputs, it can simply classify this dataset as fake, and all other data as true. By adding noise, $G$ can learn to use it to generate greater diversity in its generated outputs to overcome this kind of overfitting. This is especially relevant...
when only a small amount of low-res volume data is available. There are many possible super-res reconstructions for a single low-res volume, enlarging G’s output space thus helps avoiding overfitting of D on G’s outputs.

• By varying the noise one can generate different mesostructures and explore the distribution of potential features and properties, allowing the user to quantify the uncertainty and richness of the super-resolution process. Additionally, the noise is spatially distributed as the input volume, allows for the creation of periodic boundaries of the properties that are governed by the noise input. Therefore, latent space exploration of the noise input and its use in mesostructure optimization is promising as a subject of future research.

It is possible to try and replicate the capability of the proposed method using only the SliceGAN presented by Kench and Cooper. This can be attempted by searching the latent space of a pretrained generator for a specific high-res 3D mesostructure whose down-sampled version and the input 3D volume have a low voxel-wise loss. However, this process is often unsuccessful, landing in unrealistic local minima far from the desired mesostructure. This is likely because the latent space of a typical SliceGAN generator contains a broad distribution of realistic mesostructures, however it does not contain every possible arrangement of features. Thus, it is highly likely that a specific 3D image is not contained within G. SliceGAN is only able to reconstruct mesostructures with the same distribution of features, as opposed to the exact high resolution volume associated with the available low-res dataset.

During training, G generates $64^3$ voxel cubes in such a way that, after convergence, every 2D perpendicular $64^2$ slice of the cube is indistinguishable by the discriminator from the real training data samples. After training, the final output is then generated using an arbitrarily larger input to the generator. For example, a low-res input volume of $128^3$ with a scale-factor of 4 will output a super-res volume size of $512^3$, while during training generating $64^3$ volumes required only $16^3$ low-res inputs. A possible limitation of the SuperRes method is that the large volume will fail to capture very long-range interactions, since it was only trained and measured by generating small volumes.

The transport efficiency (the ratio of the phase fraction to its tortuosity factor) can be used to explore the longer range interactions in a large volume, because it is calculated by simulating a flow through the entire volume between two parallel faces. A comparison was made between the transport efficiencies across the different phases of the super-res large volume and the mean of the small $64^3$ cubes in both the validation case studies, and no apparent difference was found, with $< 0.01$ mean absolute difference. This positive finding can be linked to the nature of the super-resolution process, as the bulk structure of the material is already present in the low-res volume, retaining the long-range structure of the material. For example, the active material arrangement in the first case study, and the lamellae arrangement in the second case study.

Our proposed model does not implement the classic style transfer algorithm by Gatys et al., in part because of the 2D to 3D dimensionality mismatch of the style representation. However, our approach is not too dissimilar to the framework in, as the loss of G consists of a linear combination of the content representation (the voxel-wise loss) and the style representation (D’s score for fake images).

Any super resolution approach will contain a degree of uncertainty in the generated result. One advantage of the SuperRes approach is that the expected variation in the generated volume can be explored by varying the noise vector. Furthermore, around the edges of the super-res volume the uncertainty is particularly acute. This is because the low-res volume does not contain any information about the regions adjacent to its boundaries. So, for example, in the case of the cathode, a pore just inside the boundary may in reality be next to a particle of active material that was just outside the imaged region. This would affect the morphology of the binder phase nearby. This concept is reflected in the non-uniform information density near the edges of the volume, as described in. As such, it is advisable for users of the SuperRes method to crop their volume by the equivalent of 1 voxel of the input volume from the generated volume (e.g. in $4\times$ resolution increase, remove 4 layers from each face), as advised in the github repository manual of this project.

5 Conclusion

The results presented in this paper show the ability of the SuperRes algorithm to faithfully synthesize a high-res, multi-phase and large field-of-view volume by fusing complementary 2D and 3D image data. This was shown across a range of lengthscales and phase sensitivities. Although this study focused on energy storage materials, the application of SuperRes is not limited to this domain, and could be extended to other materials and imaging techniques. For example, cata-
lyst beds, rock formations, or snow and soil packing, as well as medical imaging.

By the complementary fusion of 2D and 3D image data, this method can overcome the physical and experimental constraints of contemporary 3D imaging such as contrast, resolution and field-of-view. With realistic, super-resolved, style-transferred 3D data, more accurate simulations and characterisations of materials can be performed. Finally, even when possible, imaging high-res 3D volumes can be expensive and time-consuming. SuperRes offers an alternative, exchanging expensive machinery and complex techniques with open-source software.

Future developments of this method might include adjusting the architecture such that it can more easily capture longer range relationships as the network can be deeper and the information density could be shared among a larger region. Other future work might include a combination with other common GAN approaches such as conditional GANs and transfer learning. The former will enable interpolation between mesostructures with differing properties, while the latter aims to speed up the training process. Additionally, this model could be used in an algorithm that applies to an inpainting problem, whereby missing or erroneous data can be in-painted, removing the need to repeat long imaging processes containing small errors and avoiding throwing away potentially useful datasets. Thus, there is broad potential for expansion of this approach as a tool for material characterization and optimization.

Data Availability

The study used open-access training data available from the following sources as described in the Supplementary Information Table 1. All generated data used are available from the authors on request.

Code Availability

The codes used in this manuscript are available at https://github.com/amirDahari1/SuperRes.

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Authorship

AD designed and developed the code for SuperRes, trained the models, performed the statistical analysis and drafted the manuscript. SK, IS and SJC contributed to the development of the concepts presented in all sections of this work, helped with data interpretation and made substantial revisions and edits to all sections of the draft manuscript.

Competing interests

The authors declare no competing interests.

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Supplementary Information
| Case | Name             | Isotropy | Data                     | Size (voxels number) | Shown in figure | Phases visible | Resolution | Field of view | Source                        |
|------|------------------|----------|--------------------------|----------------------|-----------------|----------------|------------|--------------|-------------------------------|
| 1    | Battery cathode  | Isotropic| High-res input           | 2048                 | 200             | 3 Medium      | Large      | Both inputs were generated using from dataset. | Both inputs were generated using from dataset. |
|      |                  |          | Low-res input            | 256³                | 60³             | 2 Low        | Large      | Low-res input High-res input | Low-res input. |
|      |                  |          | Super-res output         | 1024¹               | 240³            | 3 Medium     | Large      | High-res output | High-res input High-res input. |
| 2    | Battery separator| Anisotropic| High-res input          | 624 × 300 (×2)      | 624 × 300 (×2)  | 2 High       | Small      | Both inputs were obtained from dataset.                        | Both inputs were obtained from dataset.                        |
|      |                  |          | Low-res input            | 306 × 153 (×2)      | 306 × 153 (×2)  | 2 Low        | Large      | Low-res input | Low-res input.                        |
|      |                  |          | Super-res output         | 1024²               | 240³            | 3 Medium     | Large      | High-res input | High-res input.                        |
| 3    | Fuel cell anode  | Isotropic| High-res input           | 1024²               | 256³            | 3 High       | Medium     | Both inputs were obtained from dataset.                        | Both inputs were obtained from dataset.                        |
|      |                  |          | Low-res input 1.6        | 160³                 | 80³             | 3 Medium     | Large      | Both inputs were obtained from dataset.                        | Both inputs were obtained from dataset.                        |
|      |                  |          | Low-res input 2          | 128³                 | 64³             | 3 Medium     | Large      | Low-res input | Low-res input.                        |
|      |                  |          | Low-res input 4          | 64³                  | 32³             | 3 Low        | Large      | Low-res input | Low-res input.                        |
|      |                  |          | Low-res input 8          | 32³                  | 16³             | 3 Low        | Large      | Low-res input | Low-res input.                        |
|      |                  |          | All super-res outputs    | 256³                 | 128³            | 3 High       | Large      | High-res input | High-res input.                        |
| 4    | Battery cathode  | Isotropic| High-res input          | 758 × 628            | 758 × 628       | 3 High       | Small      | High-res input (SEM)                        | High-res input (SEM)                        |
|      |                  |          | Low-res input            | 240³                 | 64³             | 2 Medium     | Large      | Low-res input (XCT)                        | Low-res input (XCT)                        |
|      |                  |          | Super-res output         | 193³                 | 96³             | 3 High       | Large      | High-res input | High-res input (SEM)                        |

Table 1: Summary of the open-access input data and generate output volumes for the four scenarios investigated. Further information about each dataset can be found in the Supporting information.
Figure 7: The structure of the generator (G) and the discriminator (D) in the GAN architecture. G’s structure is dependent on the scale factor used, and can have 3 different structures. This design enables the same output of $64^3$ cubes from different inputs, that are dependent on the scale factor. For example, low-res input with a scale factor of 2 is a $32^3$ cube and a low-res input with a scale factor of 4 is a $16^3$ cube and both will output a $64^3$ super-res cube.
Figure 8: Model training diagram.

Figure 9: Segmentation of the SEM 2D image. The segmentation to three-phase image was using Weka segmentation software\textsuperscript{1} accompanied by some manual finishes, to try and account for the ‘pore-backs’ problem described in the introduction. The different segmented phases are pore (black), active material (grey) and binder (white). The original image is taken from an open source dataset\textsuperscript{22}.
Figure 10: The importance of concatenating noise to the low-res input of G. A comparison is made between models that are trained with and without the concatenated input noise. Left column shows one slice of the low-res volume input to G, top left is from an all pore volume input and bottom left is from a volume that contains active material. The middle and right columns shows slices of the outputs from models trained without (middle) and with (right) concatenated noise inputs. Respectably, the top row are slices of outputs from the all-pore volume (top left) and the bottom row are slices of outputs from the volume that contains active material (bottom left).