

Matthias Neumann

How Many Parameters Describe Nanostructures in Battery Materials?

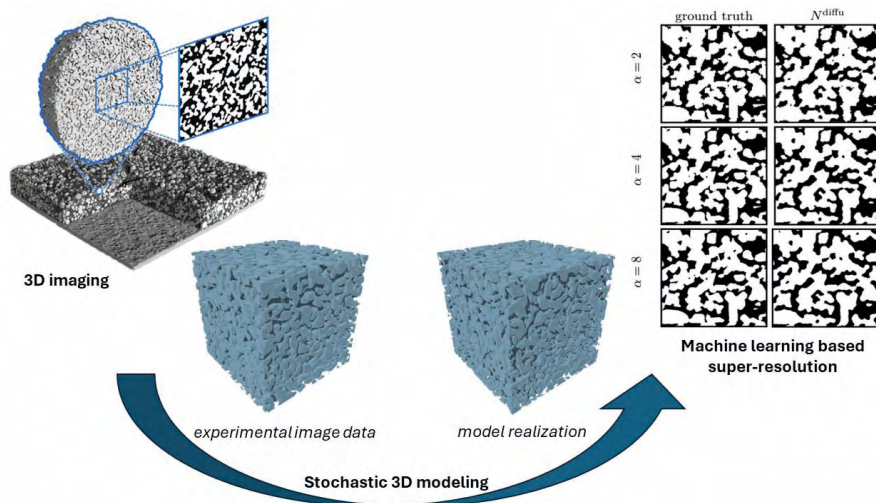
Pathways on the nanoscale influence how your battery performs when driving long distances in your car. Parametric stochastic 3D modelling is a powerful tool to reduce the experimental effort for elucidating quantitative relationships between electrode morphology at the nanoscale and effective physical properties.

In many functional materials, the morphology of disordered micro- and nanostructures influences the effective physical properties at the macro-scale. Battery electrodes, as depicted in Figure 1, consist of active material for the storage of ions, a binder-additive phase for electron transport and pores for ion transport. When speaking about morphology at the micro-scale, we mean the spatial arrangement of these three phases in the electrode. One way to increase the performance of battery electrodes is to use nanostructured active materials for creating new pathways

for ionic diffusion. This means that the active material particles are nanoporous themselves. On the nano-scale again, an irregular morphology arises from the spatial arrangement of active material and nanopores. This morphology influences effective properties of the active material particles such as their effective ionic diffusivity. For improving the performance of the battery, a detailed understanding of the interplay between morphology and properties on the nano-scale is crucial. This requires methods for capturing these irregular morphologies that are

resolved by modern techniques from 3D imaging and visualized in the centre of Figure 1. The latter can be approached by parametric stochastic 3D modelling of the morphology.

In general, a parametric stochastic model is a model that allows for generating random output, where the randomness depends on the underlying parameters. A prominent example is the normal distribution, the parameters of which are its mean and standard deviation. Drawing realizations from this model gives us random numbers with the predefined mean and standard deviation. Stochastic 3D modelling of the morphology means that each realization of our model is a 3D image representing the morphology of the electrode material, where the model parameters control morphological descriptors such as porosity, pore size distribution or the length of shortest transportation paths for the ions. Figure 1 shows a realization of a stochastic 3D model with only two parameters, that has been calibrated experimental image data. Here it is particularly fascinating that this low-parametric model has been calibrated to six data sets each manufactured with different process parameters, i.e. particle size and calcination temperature have been varied. While the parameters are calibrated based on morphological descriptors, the model is validated by showing that the numerically simulated effective ionic diffusivity is nearly identical when comparing experimental image data with model simulations [1].



Workflow from 3D imaging via stochastic 3D modelling to an artificial, yet realistic increase of resolution by machine learning. The latter is also referred to as super-resolution.

Source: Figure 1 is compiled from figures from <https://doi.org/10.1016/j.jisolsr.2023.112394> (open access under CC BY-NC-ND 4.0, Ref [1] in the text) and [10.1088/2632-2153/ae0c55](https://doi.org/10.1088/2632-2153/ae0c55) (open access under CC BY-NC-ND 4.0, Ref [2] in the text).

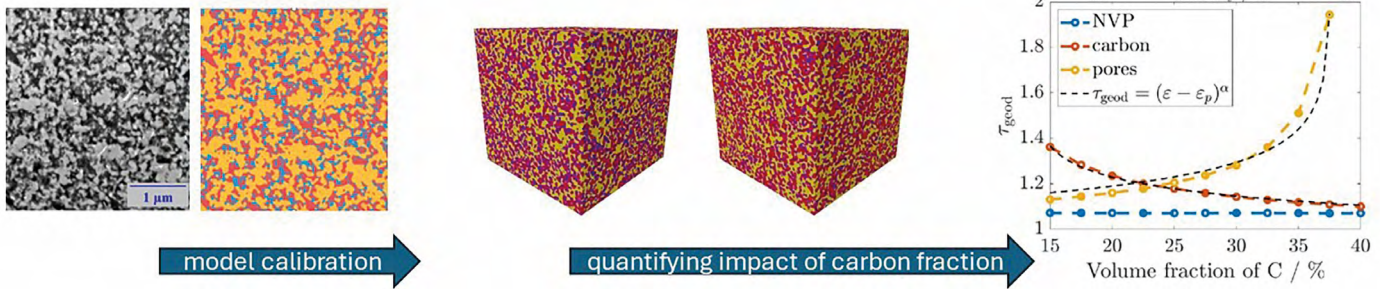


Figure 2: 2D image data serves as a basis for calibrating a stochastic model for generating 3D morphologies of nanostructures in active materials consisting of sodium vanadium phosphate with carbon. The virtually generated 3D data is used to elucidate the impact of the volume fraction of carbon on the normalized lengths of shortest transportation paths. Source: Figure 2 (NVPC.jpg) is a compilation of figures from a publication that appeared in "Batteries & Supercaps 2024" as open access under the CC BY-NC-ND 4.0 licence; see <https://doi.org/10.1002/batt.202300409> (Ref [3] in the text).

By means of stochastic 3D modelling large data sets of virtual morphologies can be generated in order to investigate quantitative relationship between morphology and effective physical properties with a reduced experimental effort. Moreover, an additional advantage of this type of models is that they are defined in continuum space. This means that we can use the model to generate 3D images of electrode morphologies with a higher resolution than the 3D images representing real electrode material. Thereby, we have generated data for training machine learning models that artificially increase the resolution of experimental image data [2], which leads to so-called super-resolved images. Here we use the property that our stochastic 3D model generates morphologies that are statistically similar than the observed ones. Reliable algorithms for super-resolution are an important ingredient for the digitalization of materials science as this accelerates the process of generating highly-resolved image data representing the morphology of micro- and nanostructures in functional materials.

Another possibility to virtually improve the experimentally acquired data-base is to calibrate the parameters of the 3D model based on 2D image data representing a few cross-sections of the material, which is possible under certain assumptions on the stochastic model. We have applied this to generate virtual, but realistic 3D morphologies of nanostructured active material used in sodium-ion batteries. The active material in the considered materials consists of sodium-vanadium-phosphate with carbon. For this purpose, we have used a stochastic 3D model with five parameters, that is able to generate 3D morphologies, the 2D cross-sections of which are statistically similar to the 2D cross-sections observed by imaging techniques. An application of the model was to quantify the relationship between the amount of carbon and the so-called mean geodesic tortuosity which measures the normalized lengths of shortest transportation paths through the phases [3], see Figure 2.

Beyond battery materials, the interplay between the manufacturing process, the morphology of micro- and nanostructures and the corresponding effective physical properties is also highly relevant for other types of functional materials, such as, e.g. additively manufactured alloys or paper-based materials. Regarding paper-based materials, relationships between physics and morphology are investigated in a long-standing collaboration with the Institute of Solid State Physics at TU Graz. Recent results focus on the influence on morphology on air flow through paper sheets [4].

REFERENCES

- [1] Neumann, S.E. Wetterauer, M. Osenberg, A. Hilger, P. Gräfensteiner, A. Wagner, N. Bohn, J.R. Binder, I. Manke, T. Carraro and V. Schmidt. A data-driven modeling approach to quantify morphology effects on transport properties in nanostructured NMC particles. *International Journal of Solids and Structures* 280 (2023), 112394.
- [2] O. Furat, P. Gräfensteiner, R. Saxena, M. Osenberg, M. Neumann, I. Manke, T. Carraro and V. Schmidt, Super-resolving 3D nanostructures using artificially generated image data and spatial transport simulations. *Machine Learning: Science and Technology* 6 (2025), 045006.
- [3] M. Neumann, T. Philipp, M. Häringer, G. Neusser, J. R. Binder and C. Kranz. Stochastic 3D modeling of nanostructured NVP/C active material particles for sodium-ion batteries. *Batteries & Supercaps* 7 (2024), e202300409.
- [4] P. Gräfensteiner, A. Rodriguez, P. Leitl, E. Baikova, M. Fuchs, E. Machado Charry, U. Hirn, A. Hilger, I. Manke, R. Schennach, M. Neumann, V. Schmidt and K. Zojer, Predicting air flow in calendered paper sheets from micro-CT data: Combining physics with morphology. *Transport in Porous Media* 153 (2026), 15.



Source: Privat

Matthias Neumann is tenure-track assistant professor at the Institute of Statistics at TU Graz. The focus of his current research is on the development of statistical methods for analysing and modelling micro- and nanostructures in close collaborations with partners from mathematics, physics, chemistry, engineering and materials science.