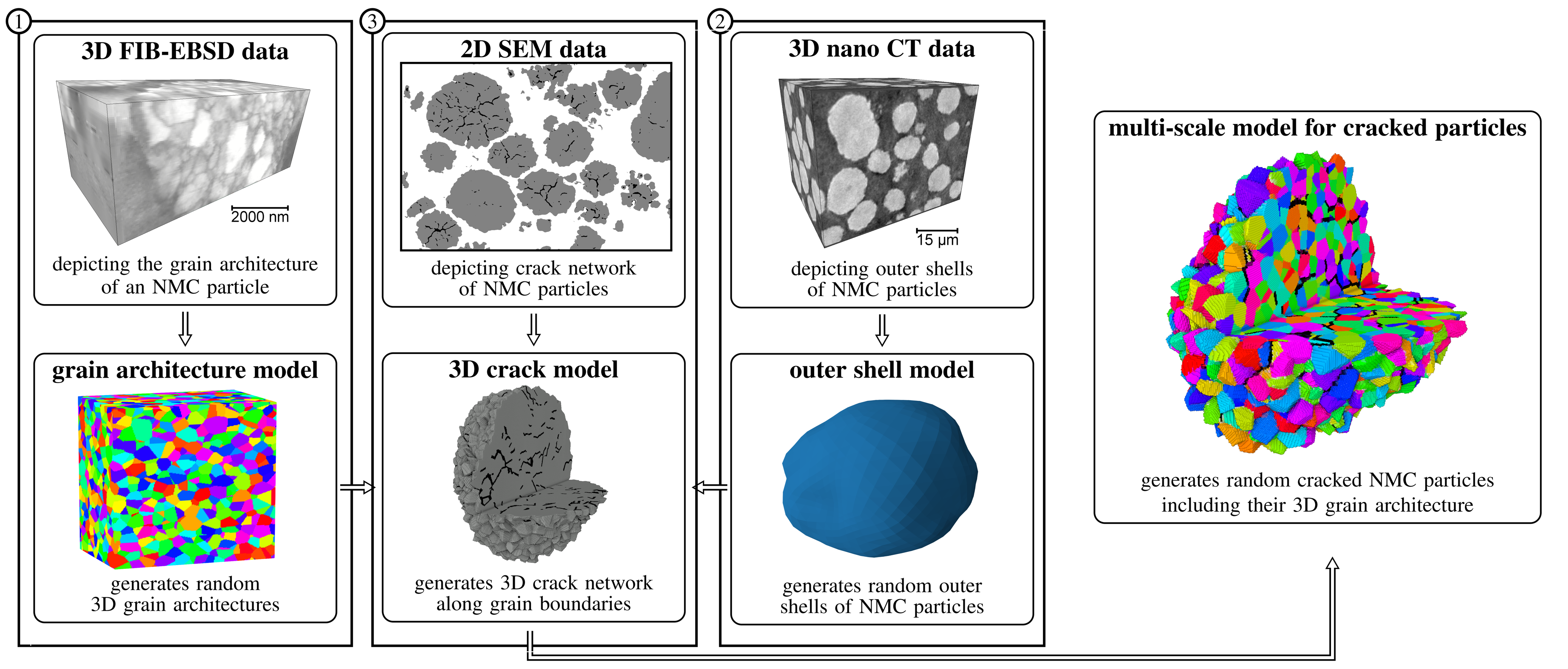
Stochastic 3D reconstruction of cracked polycrystalline NMC particles using 2D SEM data

P. Rieder[1], O. Furat[1], F.L.E. Usseglio-Viretta[2], J. Allen[2], P. Weddle[2], D.P. Finegan[2], K. Smith[2], V. Schmidt[1]

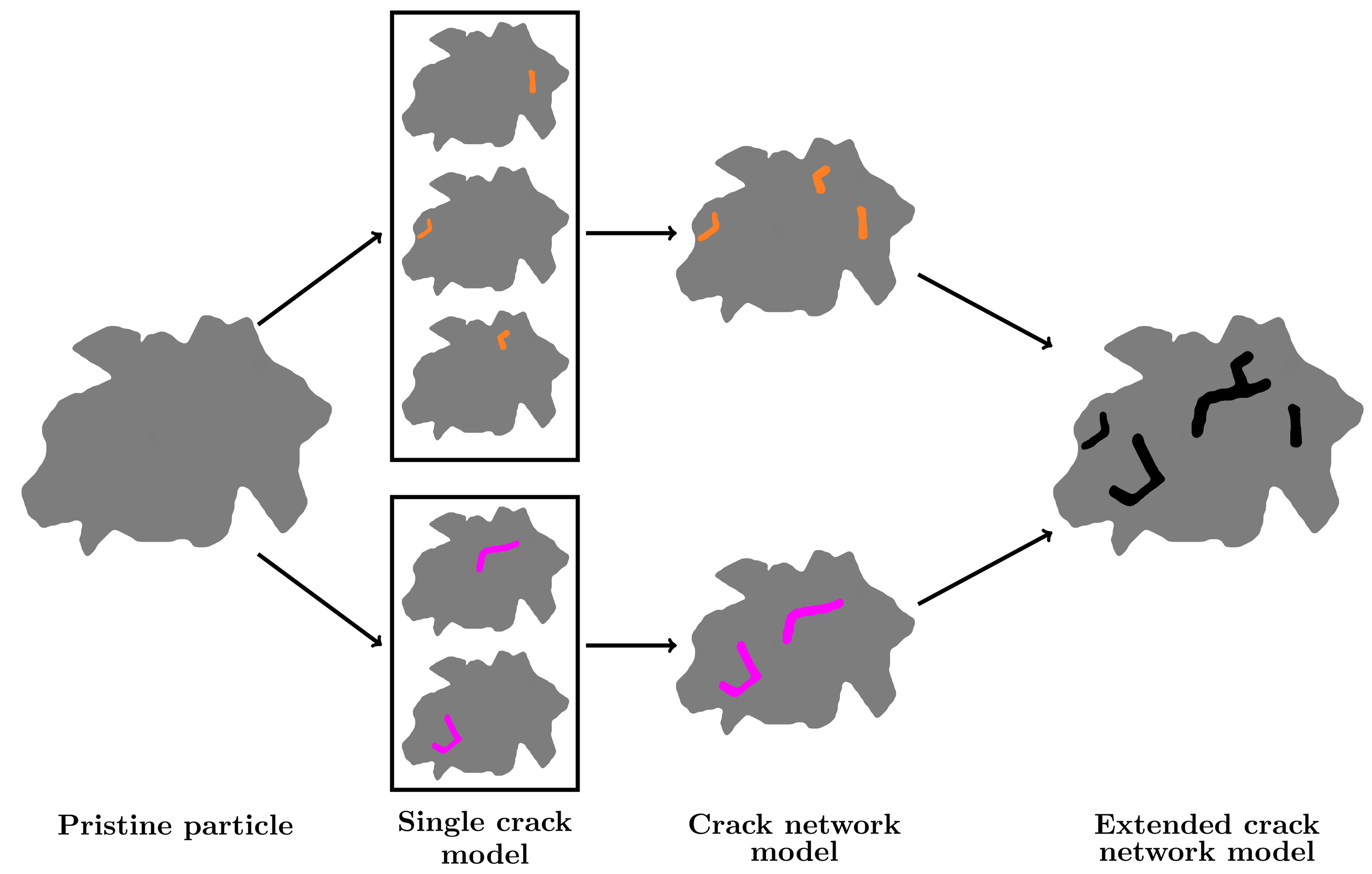
[1]Institute of Stochastics, Ulm University, Ulm, Germany, [2]National Renewable Energy Laboratory, Golden (Colorado), USA

MOTIVATION & WORKFLOW

- **SEM, nano CT** and **FIB-EBSD** provide detailed 2D and 3D image data of nano- and microstructures of functional materials
- Cost for 3D imaging and insufficient information provided by 2D imaging for investigating descriptors related to transport paths **motivates the development** of a stochastic 3D crack model to address this stereological challenge.
- Utilize **spatial stochastic modeling** for the **holistic structural characterization** of active material (AM) particles in Li-ion battery electrodes
  - Allows generation of arbitrarily many **virtual, but realist** cracked AM particles
  - Realizations can serve as **input** for several **numerical simulations**
- Investigation of **3D structure-property relationships**, i.e., how effective material properties are influenced by their nano- and microstructure
- Provide **structuring recommendations** for manufacturing processes of optimized battery materials

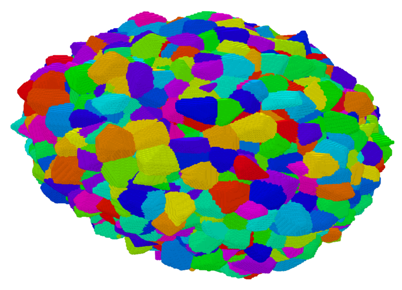


GRAPHICAL ABSTRACT



PRISTINE MODEL

Multi-scale pristine model



Multi-scale model for pristine particles [4]

(i) characterizes the shape of particles,

(ii) characterizes the grain architecture of particles,

(iii) can be used to generate input for numerical simulations [5].

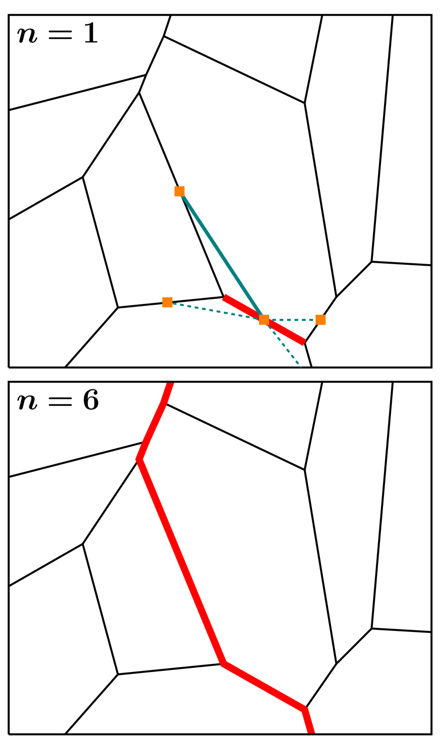
For generation see posters of Lukas Fuchs and Daniel XXX.

3. CRACK NETWORK

Single crack model

1. Initialize set of cracked facets $C = \emptyset$
2. Define $N_{\text{facets}} = \lceil \hat{N}_{\text{facets}} \rfloor$, as random number of cracked facets, where $\hat{N}_{\text{facets}} \sim \text{Wei}(\lambda_W, k_W)$
3. Choose a random initial facet f_1 and assign it to C.
4. Determine average normal v_C of facets in C.
5. Choose **facet f** adjacent to a $f_C \in C$, whose normal v **maximizes** $|\langle v_C, v\rangle|$ (e.g choose f, which is "aligned best" with C)
6. Repeat (iv) and (v) until $|C| = N_{\text{facets}}$
7. Dilate each $f \in C$ by $\delta \geq 0$, where $\delta \sim \Gamma(k_\Gamma, \theta_\Gamma)$

Depends on parameter $\theta_1 = (\lambda_W, k_W, k_\Gamma, \theta_\Gamma) \in \mathbb{R}^4_+$.



Crack network model

- $P_{\theta_1} = (\Xi^{(\theta_1)}_{\text{solid}}, \Xi^{(\theta_2)}_{\text{crack}})$ denotes a realization of single crack model, with $\Xi^{(\theta_1)}_{\text{solid}}, \Xi^{(\theta_2)}_{\text{crack}} \subset \mathbb{R}^3$ the solid and crack phase.
- $P_{\theta_1,1}, \ldots, P_{\theta_1,n_{\text{cracks}}}$ with $P_{\theta_1,i} = (\Xi^{(\theta_1,i)}_{\text{solid}}, \Xi^{(\theta_1,i)}_{\text{crack}})$ be i.i.d. copies of P_{θ_1}, where n_{cracks} sampled from Poisson distribution with parameter $\lambda_P > 0$.
- Define $P_{\theta_2} = (\Xi^{(\theta_2)}_{\text{crack}}, \Xi^{(\theta_2)}_{\text{solid}})$ with $\Xi^{(\theta_2)}_{\text{solid}} = \cap_{i=1}^{n_{\text{cracks}}} \Xi^{(\theta_1,i)}_{\text{solid}}$ and $\Xi^{(\theta_2)}_{\text{crack}} = \cup_{i=1}^{n_{\text{cracks}}} \Xi^{(\theta_1,i)}_{\text{crack}}$ as realization of the crack network model.
- Introduce technical scaling parameter c_{dim} and parametrize λ_W and λ_P.

Depends on parameter
$\theta_2 = (c_W, k_W, k_\Gamma, \theta_\Gamma, c_P, c_{\text{dim}}) \in \mathbb{R}^5_+ \times [0, 1]$.

Extended crack network model

- Draw two independent realizations $P_{\theta_2^{(1)}}, P_{\theta_2^{(2)}}$ of the crack network model.
- Define $P_\theta = (\Xi^{(\theta)}_{\text{solid}}, \Xi^{(\theta)}_{\text{crack}})$ with $\Xi^{(\theta)}_{\text{solid}} = \Xi^{(\theta_2^{(1)})}_{\text{solid}} \cap \Xi^{(\theta_2^{(2)})}_{\text{solid}}$ and $\Xi^{(\theta)}_{\text{crack}} = \Xi^{(\theta_2^{(1)})}_{\text{crack}} \cup \Xi^{(\theta_2^{(2)})}_{\text{crack}}$ as a realization of the extended crack model.

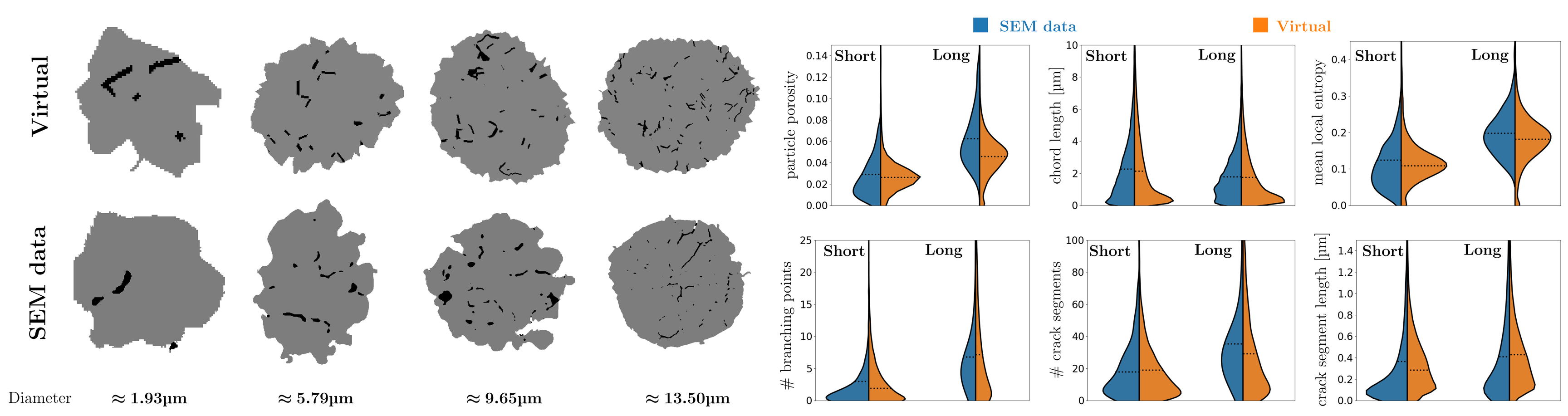
Depends on parameter
$\theta = (c^{(1)}_W, k^{(1)}_W, c^{(1)}_P, c^{(2)}_W, k^{(2)}_W, c^{(2)}_P, k_\Gamma, \theta_\Gamma, c_{\text{dim}}) \in \mathbb{R}^8_+ \times [0, 1]$.

CALIBRATION AND VALIDTAION

Calibration

The parameters of the 3D crack model are fitted by minimizing a loss function, which measures the discrepancy between virtual 2D cross sections with 2D SEM data [8]. The model was fittet to two data sets, exhibiting predominantly **short** and **long** cracks, respectively.

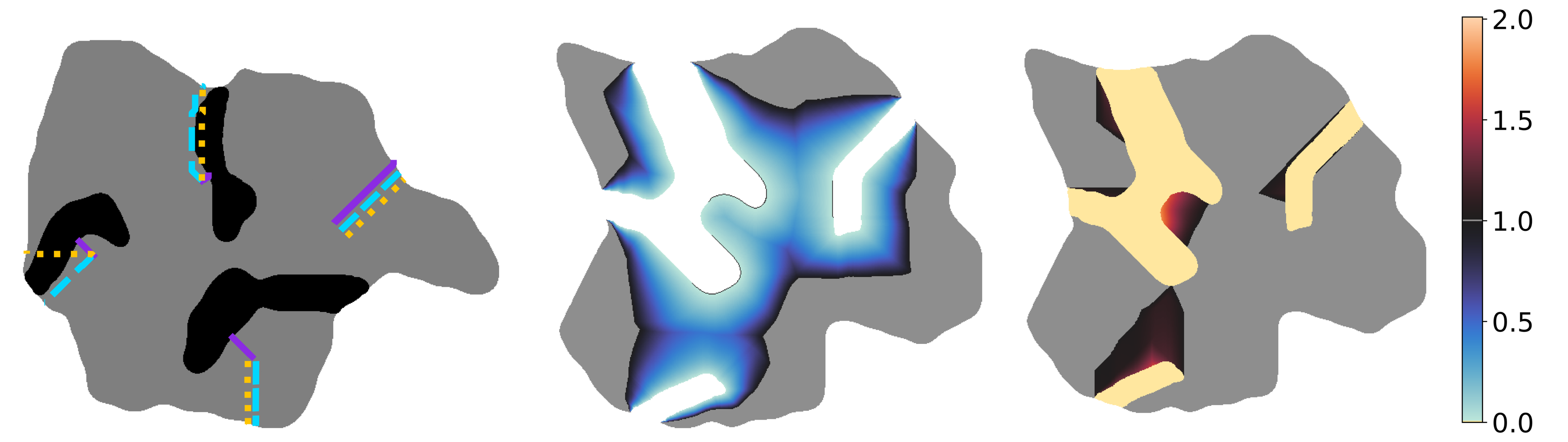
Validation



EFFECTIVE PROPERTIES

Transport path lenghts (Ion transport)

Realizations of the stochastic 3D for cracked particles can be utilized to investigate the change of path lengths before and after cracking. The change of path lengths for ion transport depends on the kind of battery. In wet cell batteries, cracks are flooded with electrolyte, resulting in shorter paths (middle), compared to the pristine particle. Conversely, in dry cell batteries, cracks typically serve as obstacles, leading to an increase in path lengths (right).



REFERENCES

[1] O. Çiçek, A. Abdulkadir, S. S. Lienkamp, T. Brox, O. Ronneberger. 3D U-Net: Learning dense volumetric segmentation from sparse annotation. Lecture Notes in Computer Science, vol 9901. Springer (2016), 424-432.

[2] O. Furat, D.P. Finegan, D. Diercks, F. Usseglio-Viretta, K. Smith, V. Schmidt, Mapping the architecture of single electrode particles in 3D, using electron backscatter diffraction and machine learning segmentation. J. Power Sources 483 (2021), 229148.

[3] L. Petrich, O. Furat, M.Y. Wan ... ng of 3D tessellations to curved polycrystalline grain boundaries. Frontiers in Materials 8 (2021), 60602.

[4] O. Furat, L. Petrich, D.P. Fine ... nith, V. S ... esentative single Li-ion electrode particle architectures from microscopy data. npj Comput. Mater. 7 (2021), 105.

[5] J. Allen, P.J. Weddle, A. Verm ... P. Finega ... t, O. Furat, D. Diercks, T. Tanim, K. Smith, Quantifying the influence of charge rate and cathode-particle architectures on degradation of Li-ion cells ... tinuum-level ... 021), 230...

[6 ... P. Fischer, T. Brox, U-Net: Convolutional networks for biomed ... ngs of Medical Image Computing and Computer-Assisted Intervention, (Cham, Switzerland, 2015), pp. 234–241.

[7 ... negan, Z. Yang, T. Kirstein, K. Smith, V. Schmidt, Super-resolving microscopy images of Li-ion electrodes for fine-feature quantification using generative adversarial networks. npj Comput. Mater. 8 (2022), 68.

[8 ... rat, F. Usseglio-Viretta, J. Allen, P. Weddle, D. P. Finegan, K. Smith, V. Schmidt, 2024. Stochastic 3D reconstruction of cracked polycrystalline NMC particles using 2D SEM data, working paper.

Download the poster

Contact: Philipp Rieder, philipp.rieder@uni-ulm.de, +49(0) 731 50 23525, Institute of Stochastics, Ulm University

universität ulm

NREL Transforming ENERGY