Modelling of Lithium-ion Battery Electrode Calendering by Discrete Element Method (DEM)

Ruihuan Ge, Denis Cumming & Rachel Smith
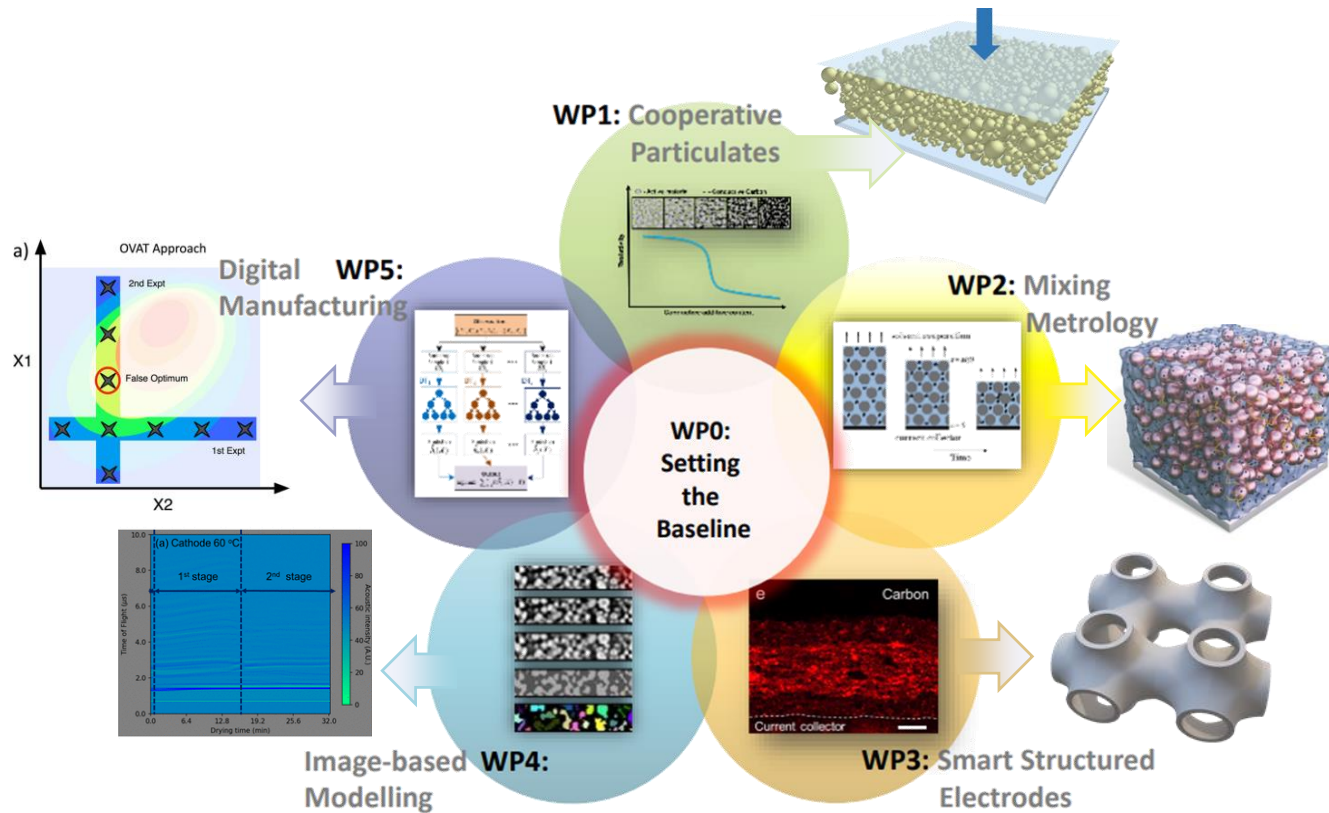
The University of Sheffield

THE FARADAY INSTITUTION

NEXTRODE

ELECTRODE MANUFACTURING

Nissan Leaf

Next Generation Electrodes (NEXTRODE)

➤ The Faraday Institution project- NEXTRODE


- **WP1: Denis Cumming (1st Nov. 15:55)**

  Ruihuan Ge (2nd Nov. 12:30)

- **WP2 and WP4: Yeshui Zhang (poster)**

  ACS Appl. Mater. Interfaces **2021, 13, 30, 36605–36620**

- **WP3: Yige Sun (1st Nov. 14:50)**
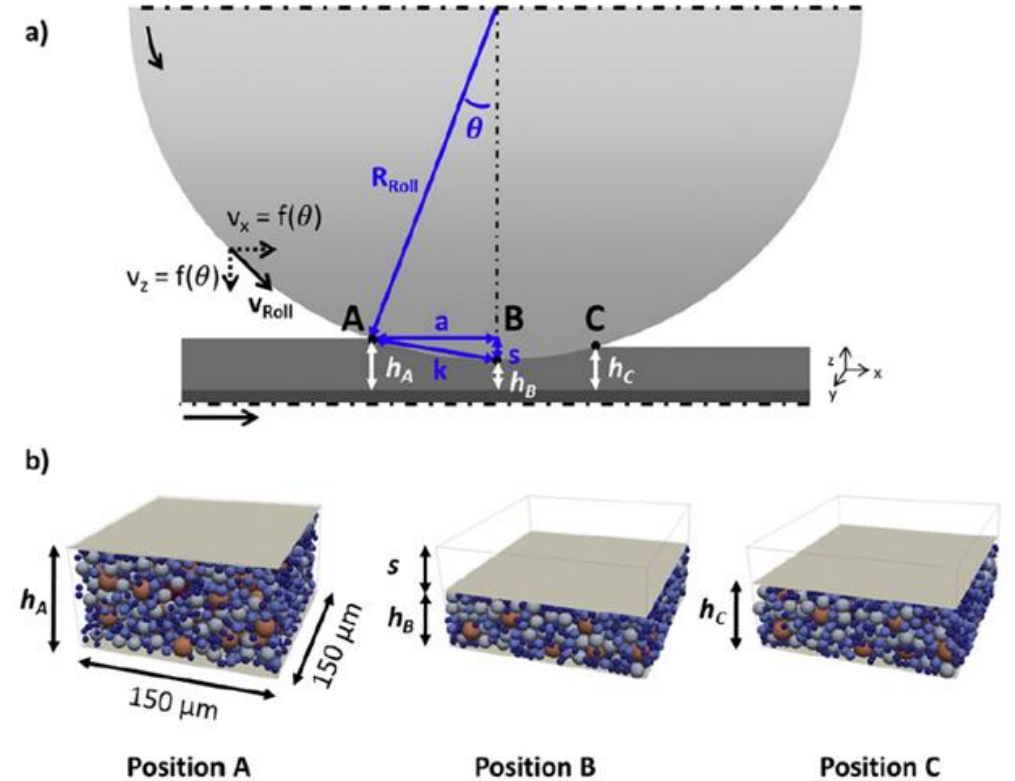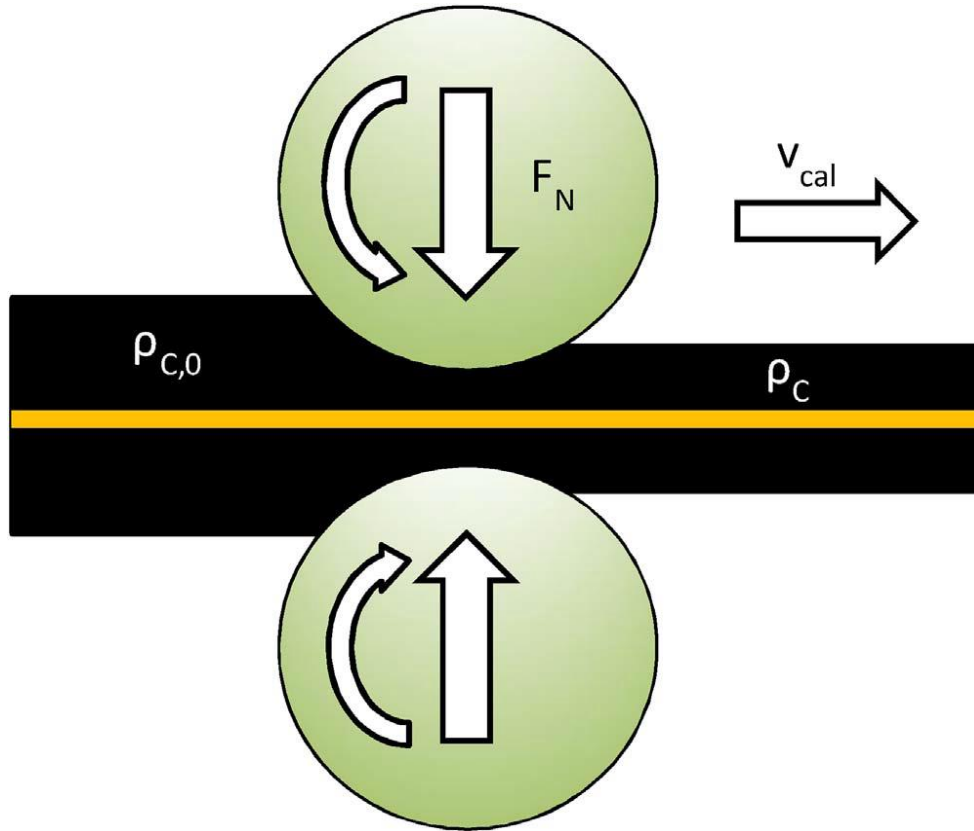
- **WP5: Geanina Apachitei (2nd Nov. 15:35)**

The WP1 research project

- The electrode manufacturing process can be understood and optimized by particulate modelling, e.g. Discrete Element Method (DEM).

- The electrode microstructure evolutions can be modelled via DEM.
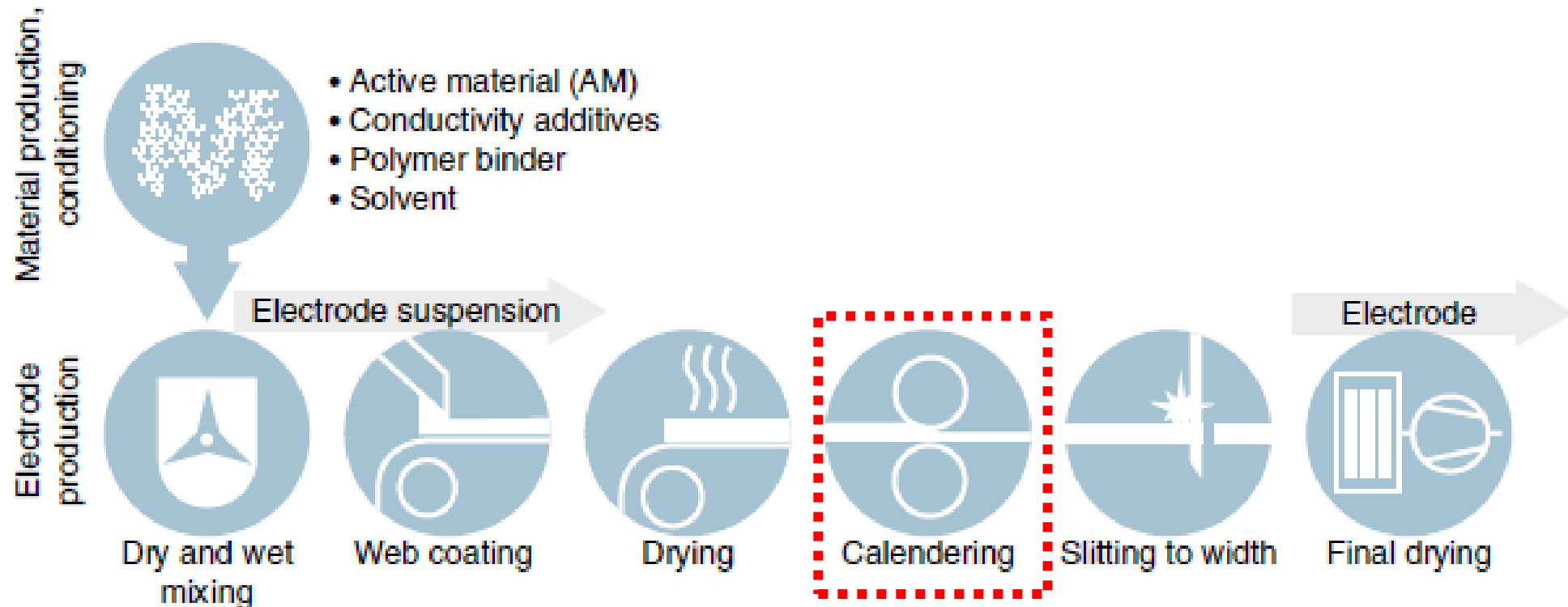
Kwade et al. *Nature Energy*

Calendering process

High calendering levels can improve the volumetric energy density, but will increase the diffusive resistance. How does the microstructure evolve during calendering?
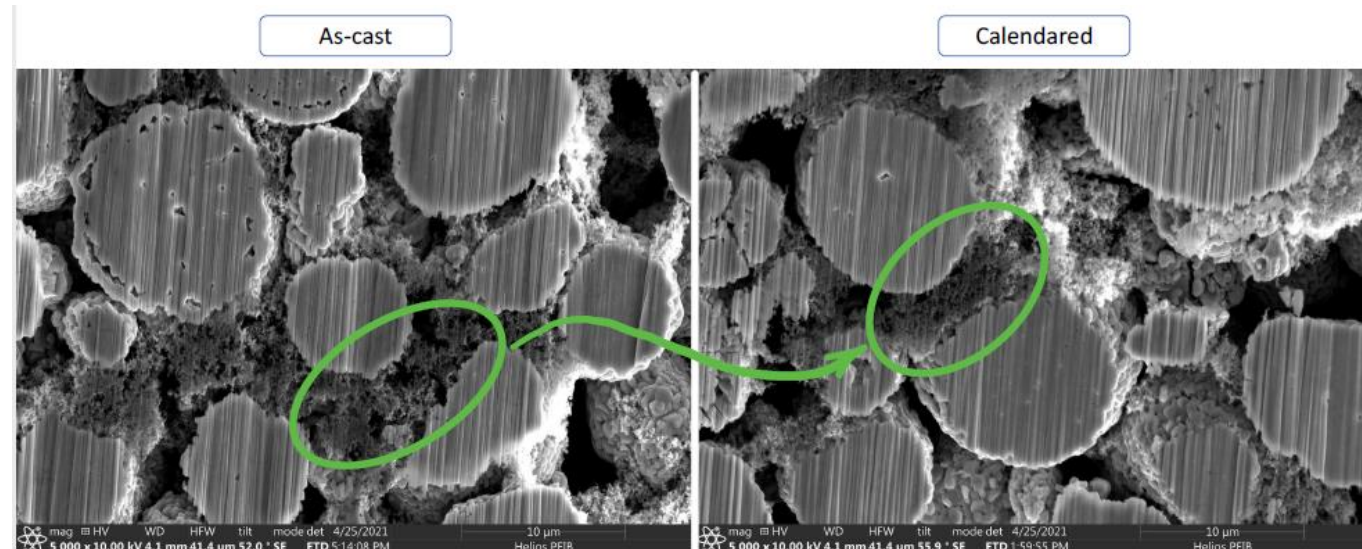
The battery electrode

- The battery electrode consists of active material (AM) and carbon binder domain (CBD).
- Calendering will improve the energy density, but increase the diffusive resistance.

I. SEM micrograph at different scales
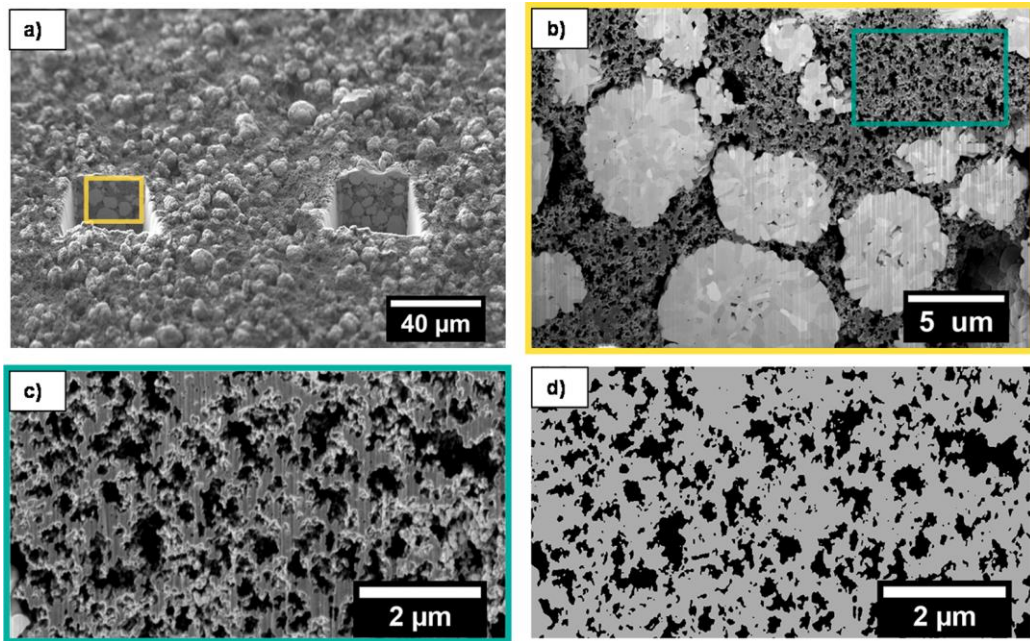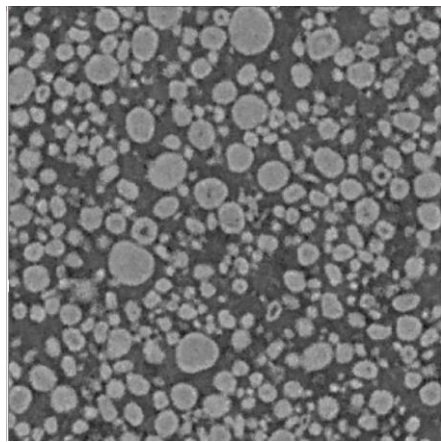
II. The effect of calendering

Daemi et al. *ACS Advanced Energy Materials*
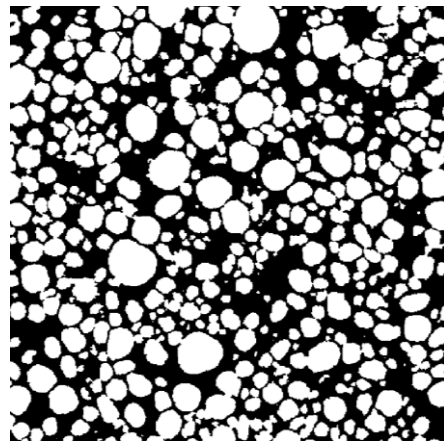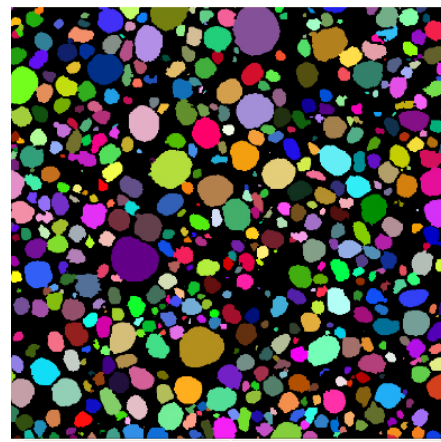
From: Yige Sun, *Oxford University*

X-ray tomography

- The tomography scan (NMC 622 active material particle) is converted to spherical particle packing.
- The uncalendered structure based on spherical particle approximations is used in DEM simulation.
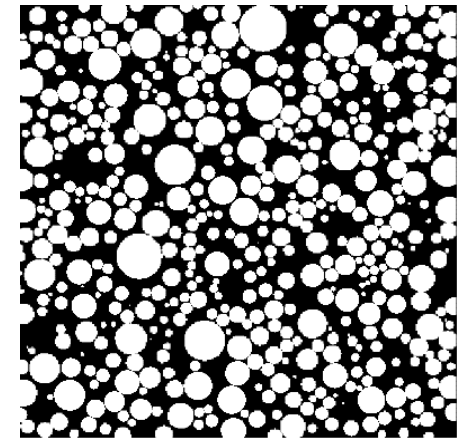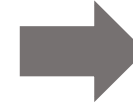

(a) Raw tomography scan
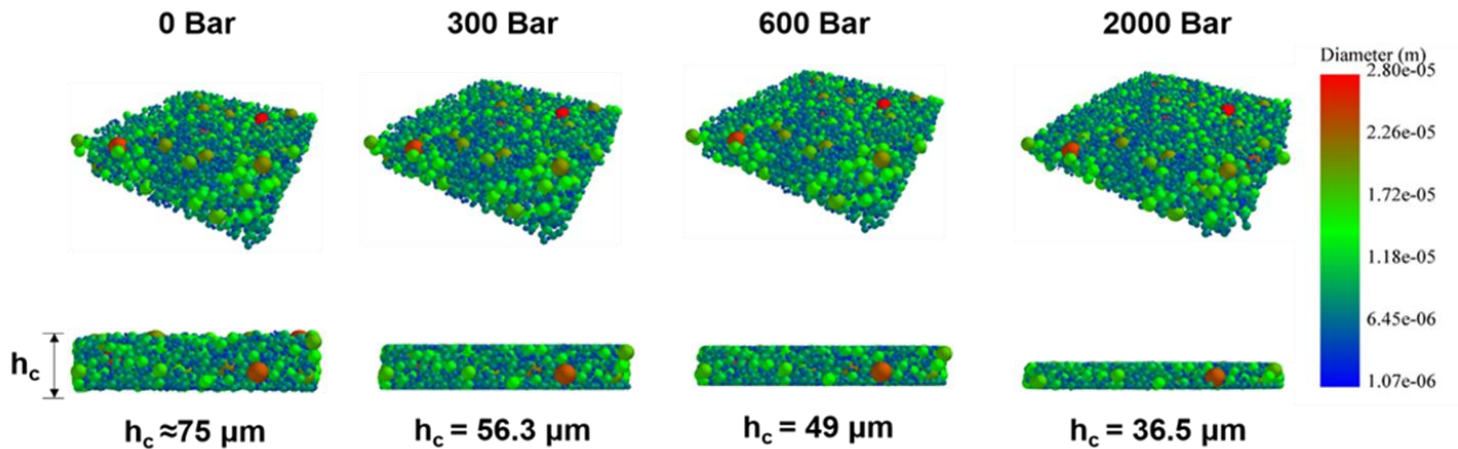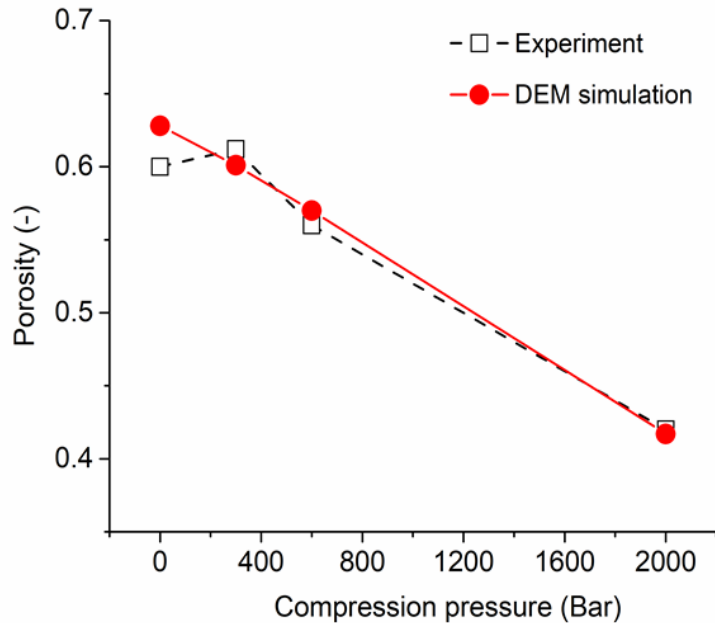
(b) Binary image

(c) Segmentation

(d) Spherical particle approximation

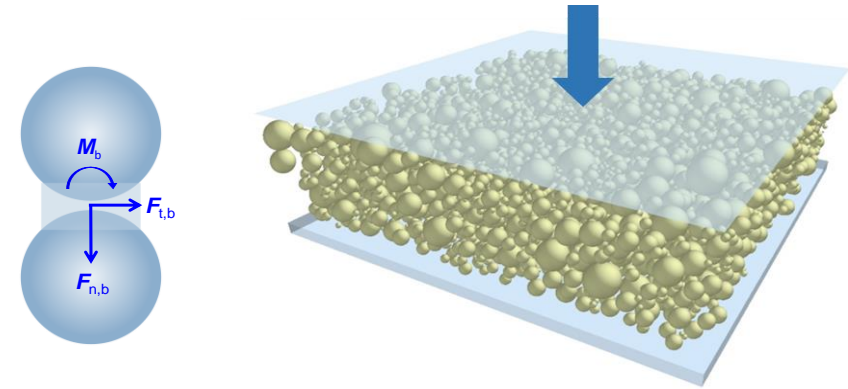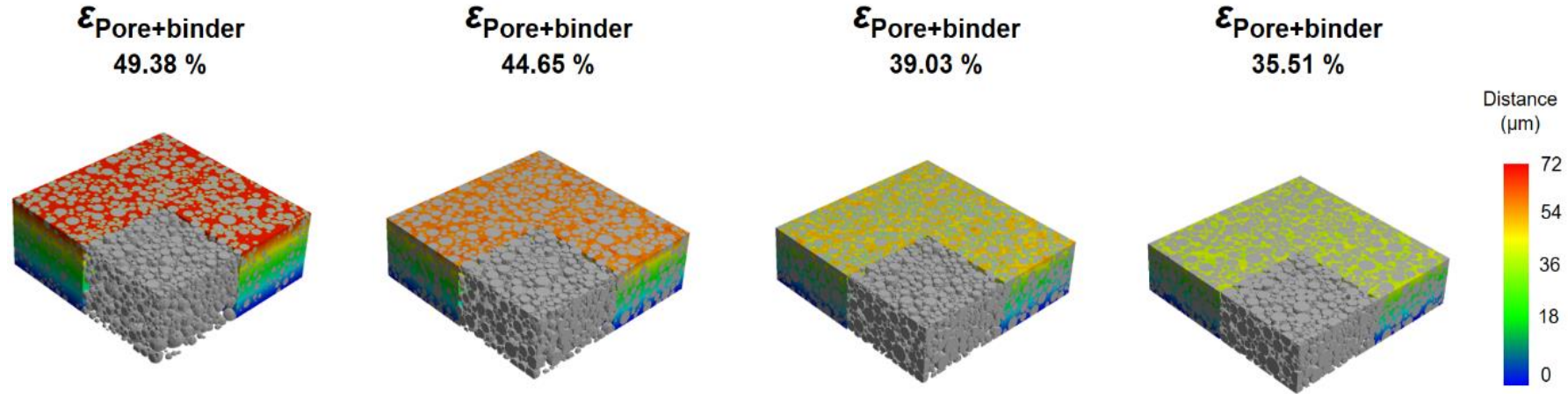# DEM simulation

- The DEM simulation predictions agree well with experimental results.

- The structures are obtained for varying calendering pressures, and the mechanical behaviour and tortuosity are analysed.
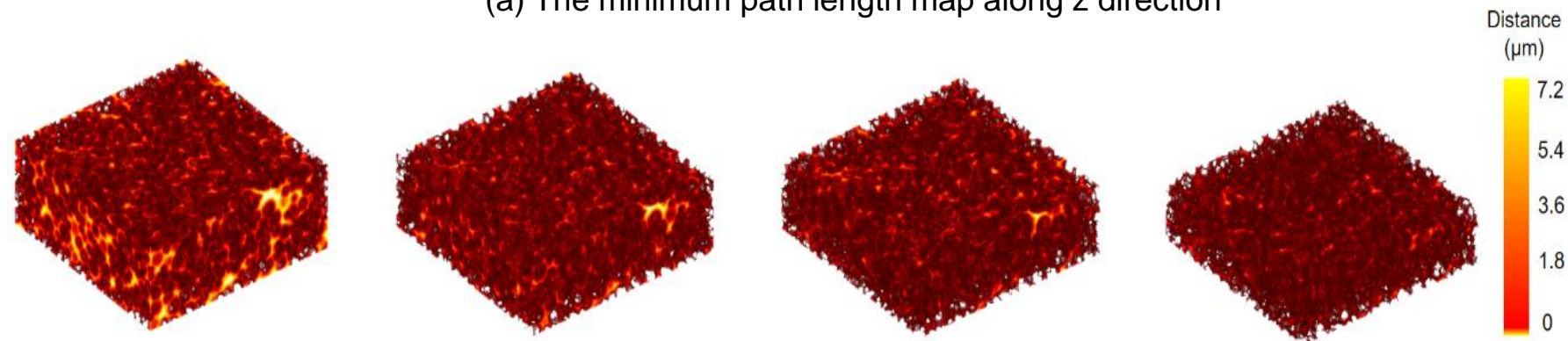
DEM simulation results

- The electrode structures under different calendering conditions.

(a) The minimum path length map along z direction

(b) The inter-particle gap distribution

Mechanical analysis

- The normalized force at each particle contact pair is calculated and plotted as contact force network.

- With increased calendering stress, the particle contact force becomes more obvious and homogeneous, and force chains are formed.

$$F_i = \sqrt{F_{x,i}^2 + F_{y,i}^2 + F_{z,i}^2} \quad i = 1, 2 \cdots n$$

$$F_{i,normalised} = F_i / \max(F_1 \cdots F_n) \quad i = 1, 2 \cdots n$$
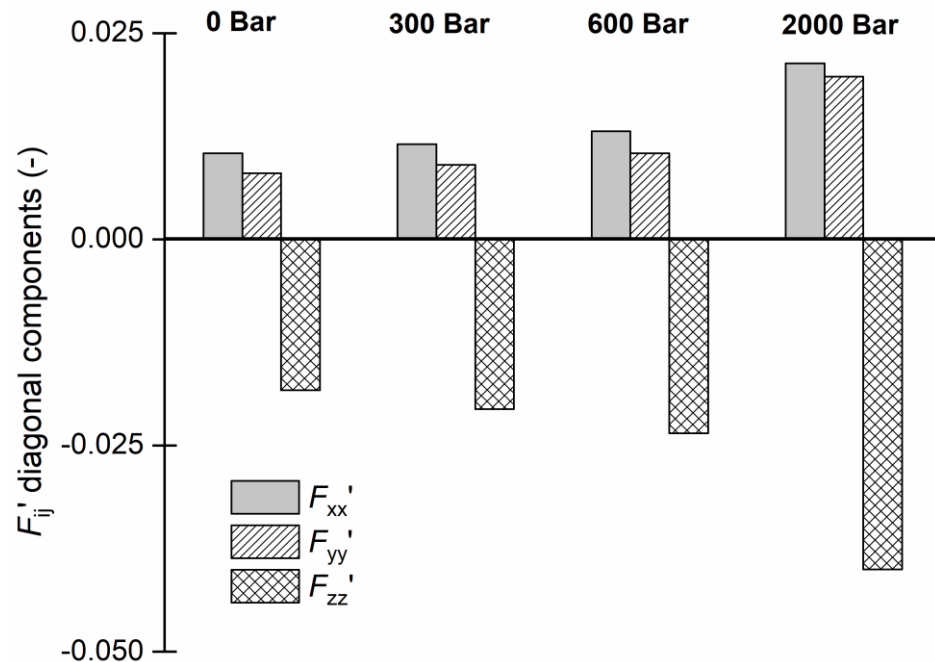

0 Bar

300 Bar

600 Bar

2000 Bar
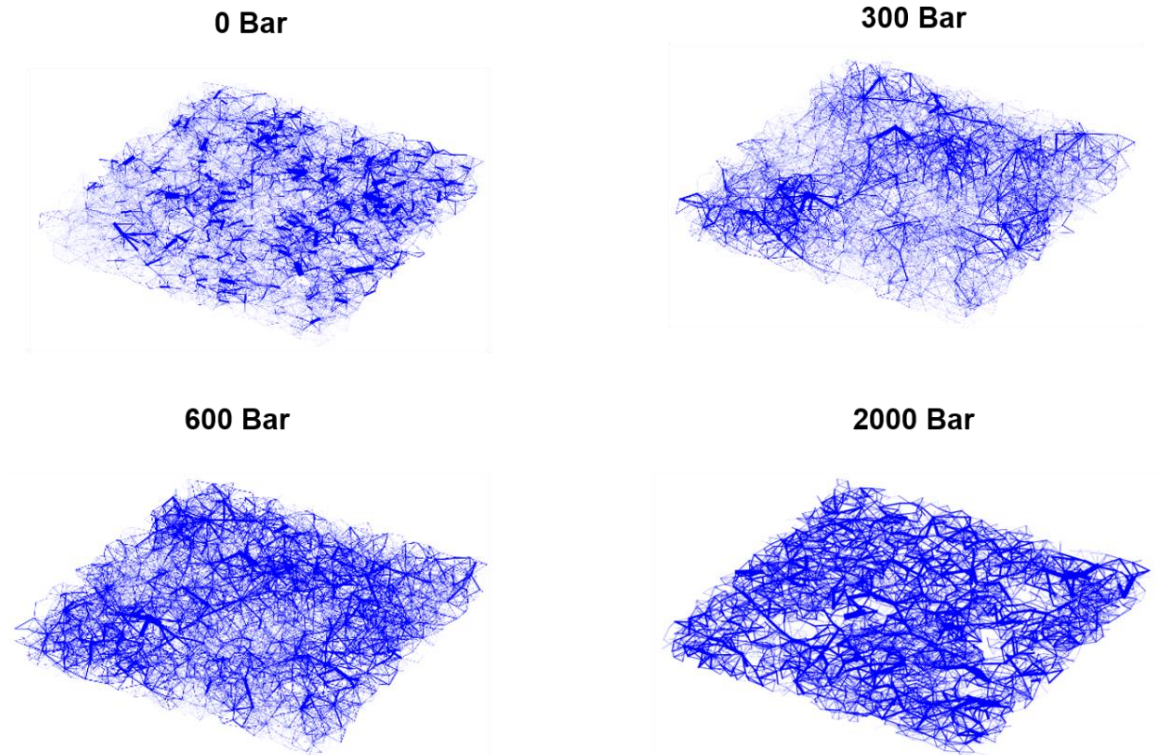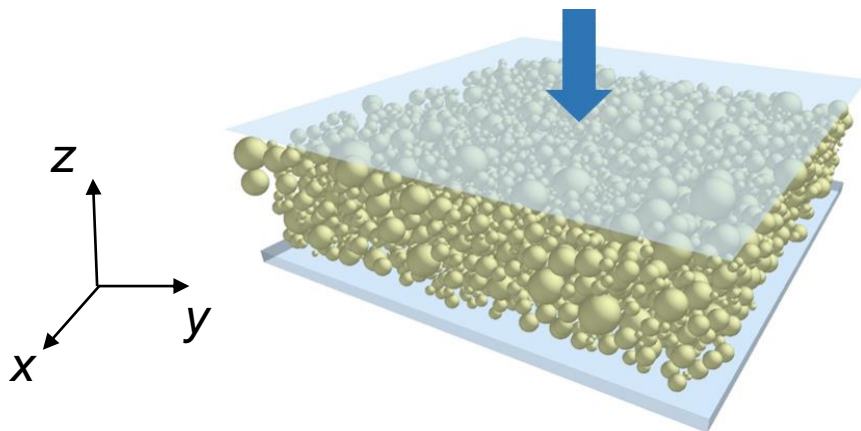
Mechanical analysis

- Two vectorial quantities: **fabric tensor** and **stress tensor** are defined to investigate the **directionality and anisotropy of particle contacts within electrode structures**.

➤ Fabric tensor: $\quad F_{ij} = \dfrac{1}{N} \sum_{\alpha=1}^{N} n_i^{\alpha} n_j^{\alpha}$
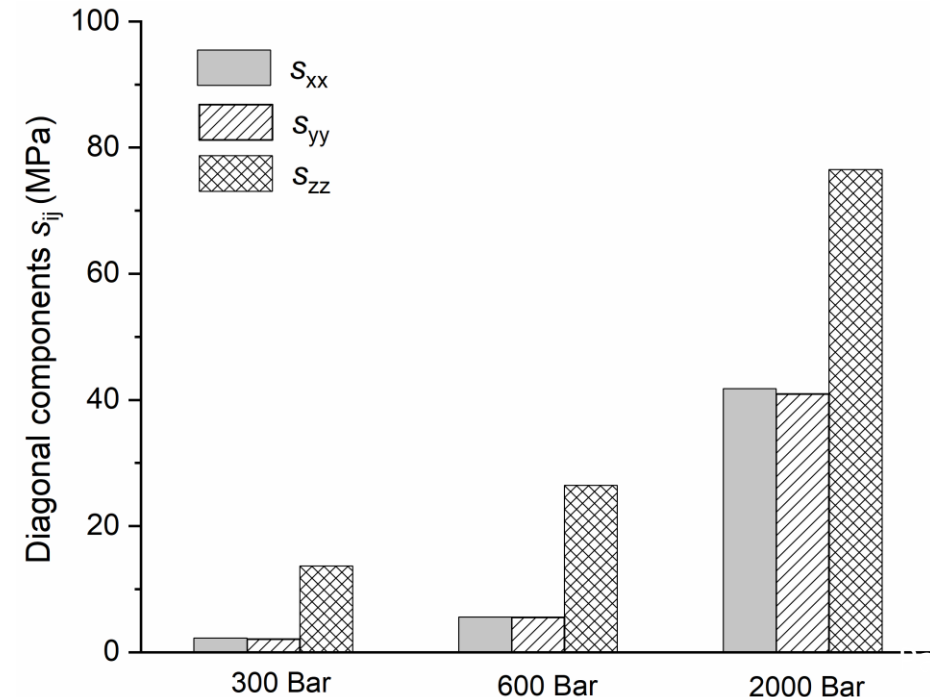
$n_i \, n_j$: contact normal between particle pairs.

➤ Stress tensor: $\quad \sigma_{ij} = \dfrac{1}{2V} \sum_{\alpha=1}^{N_c} \left(r_{1i} F_{1j} + r_{2i} F_{2j} \right)$

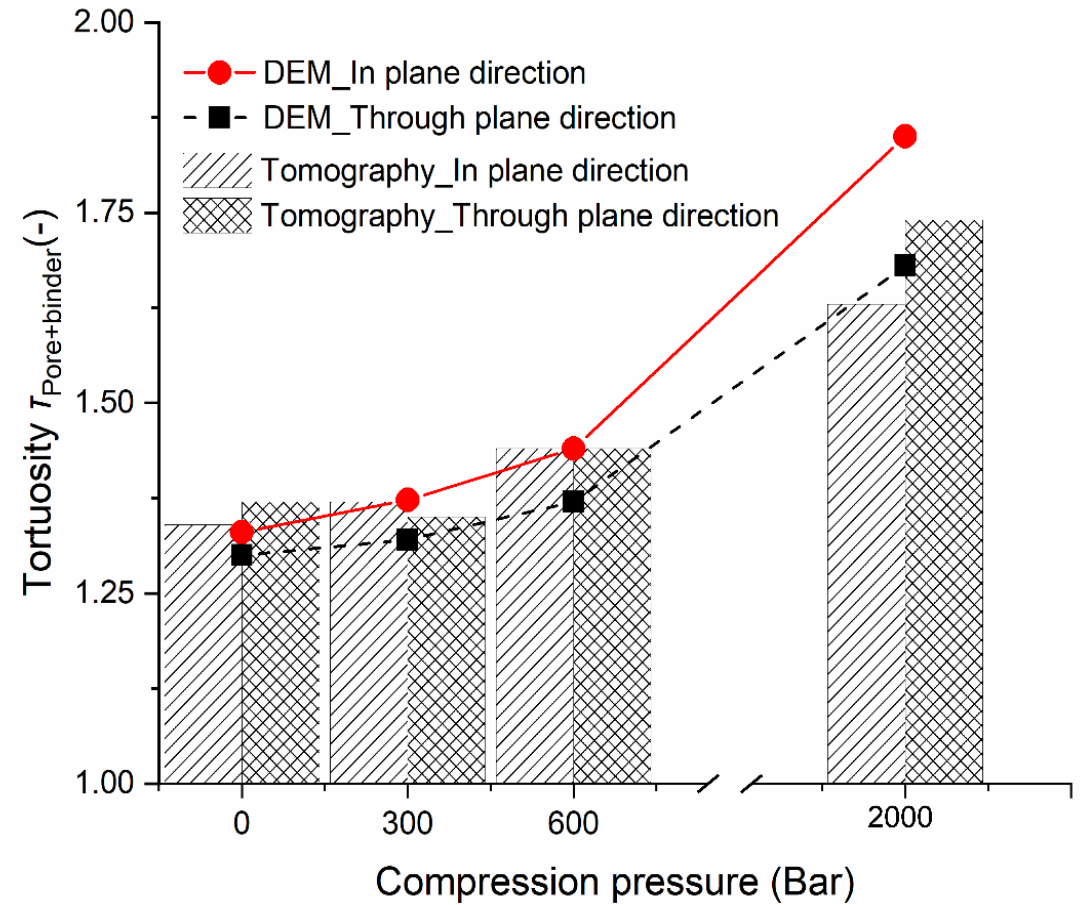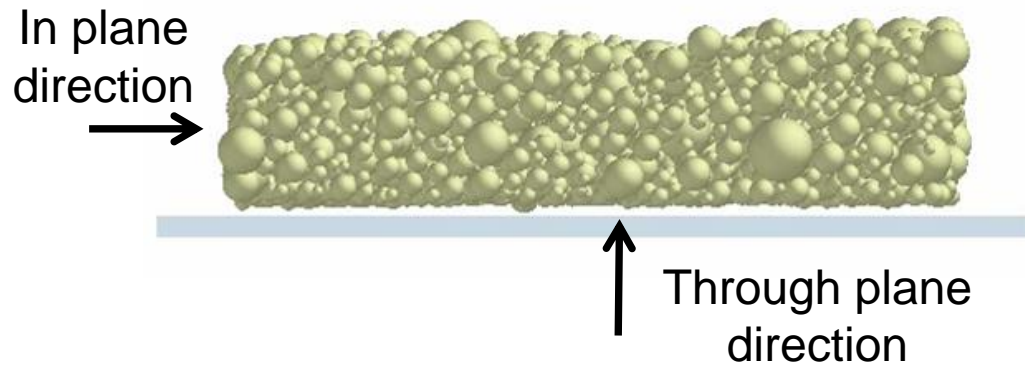$r_{1i} \, r_{2i}$: particle positions of a particle pair.

$F_{1j} \, F_{2j}$: contact force of a particle pair.

Tortuosity analysis

- The structures from DEM simulations show very similar variation tendency with the structures from tomography

- Tortuosity anisotropy is observed considering the in plane/through plane direction

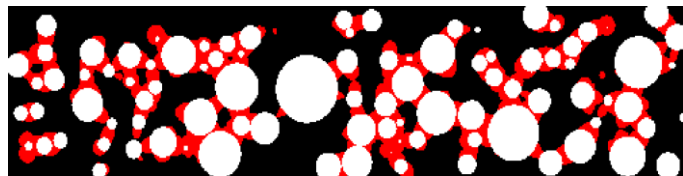In plane direction →

Through plane direction ↑

Tortuosity analysis

- The structures from DEM simulations show very similar tortuosity porosity relationships with experiment

(a) DEM generated structure without binder phase

(b) DEM generated structure with binder phase

Legend:
- × Experiments
- ■ Tortuosity $\tau_{Pore}=0.885\varepsilon_{Pore}^{-1.068}$
- □ Tortuosity $\tau_{Pore+binder}=1.02\varepsilon_{Pore+binder}^{-0.53}$
- ⋯ Bruggeman $\tau=\varepsilon^{-0.5}$

Structure with binder phase

Structure without binder phase

Axis labels: Tortuosity factor τ (-) vs Porosity ε (-)
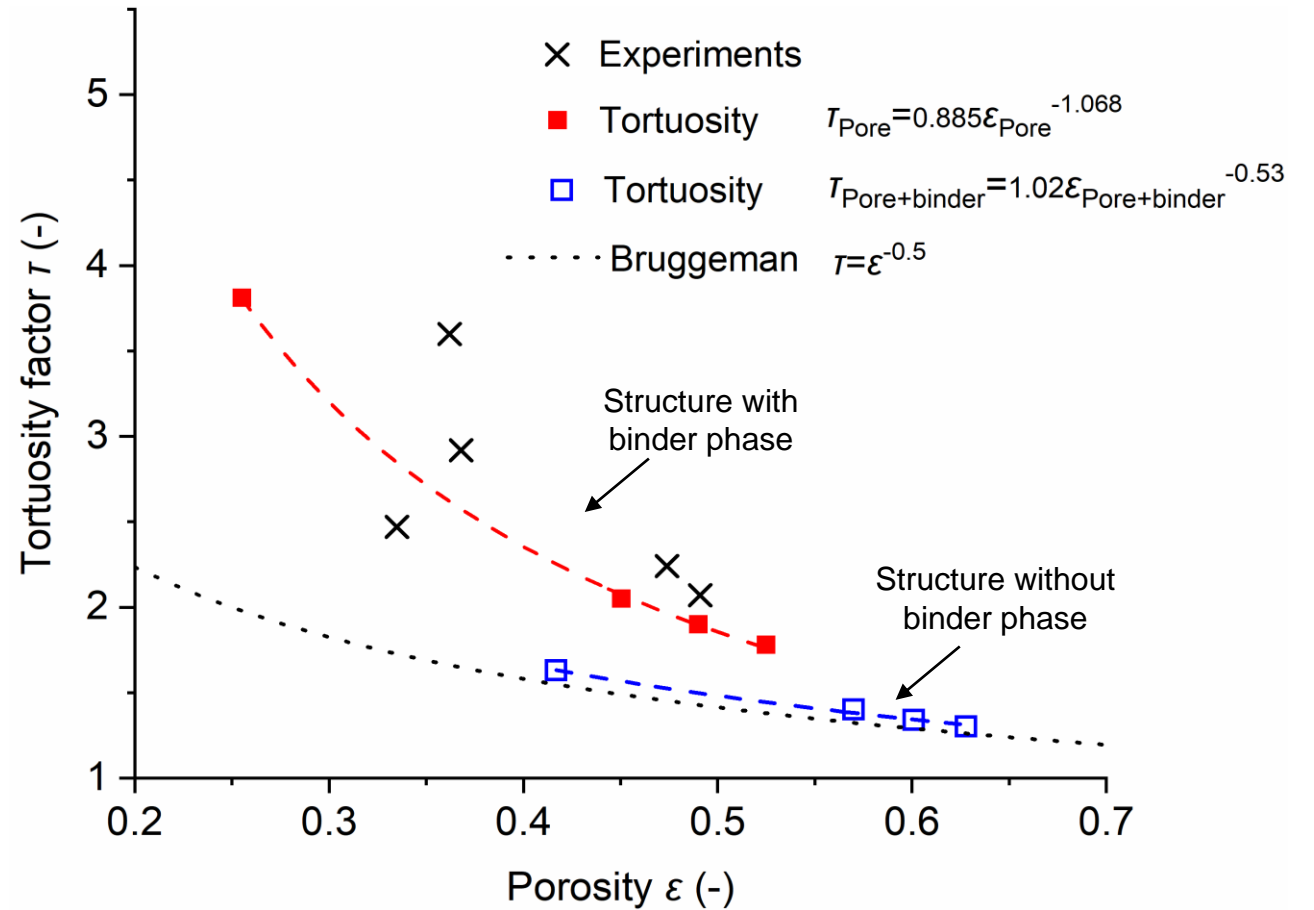
Electrochemical analysis
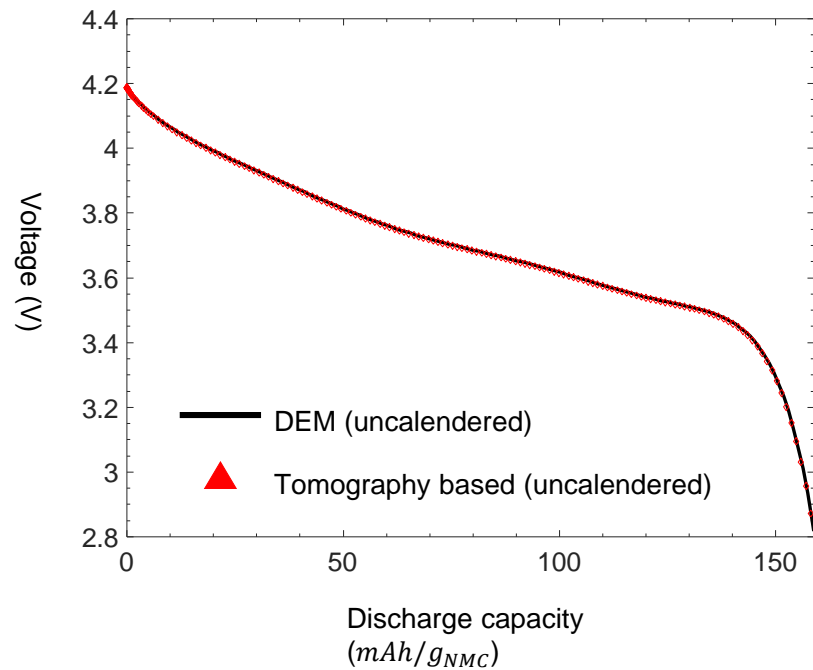
- The structure generated from DEM is compared with the corresponding structure from tomography. (**WP1-WP4 collaboration**)

- Further electrochemical analysis is underway.

➤ Discharge response of uncalendered DEM vs tomography (1C)

➤ An increase in current density at the surface of larger particles is observed in the tomography based simulation. The current density is also more heterogeneous for the tomography case.
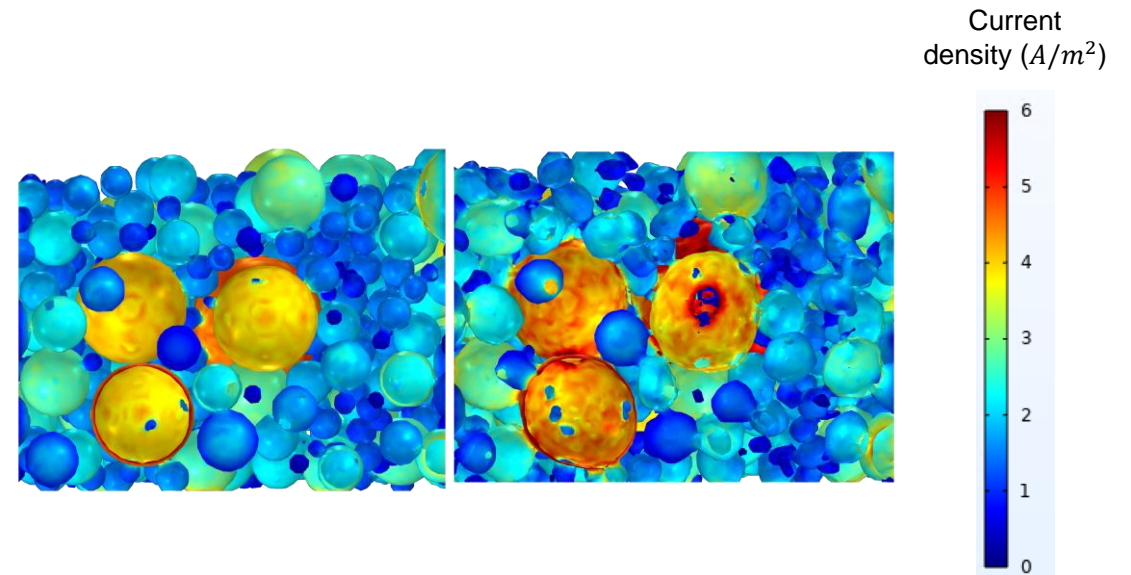
Uncalendered structure, Left: DEM Right: tomography

Summary/future work

- Discrete element method (DEM) is applied to model the electrode microstructure evolution under different calendering conditions.

- Further research is underway to investigate the effect of microstructure on the battery performance.

DEM simulation of calendering

- More accutate description of the mechanical properties of CBD phase.
- Develop a general numerical approach to predict the properties of electode microstructures
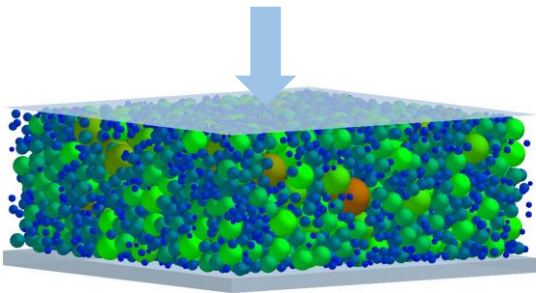

CBD phase algorithm & structure analysis

- Tune the particle shape, the porosity distribution via algorithm, e.g. non-spherical particle, graded structure.
- CBD phase morphology considering nano-pores and validated by tomography.
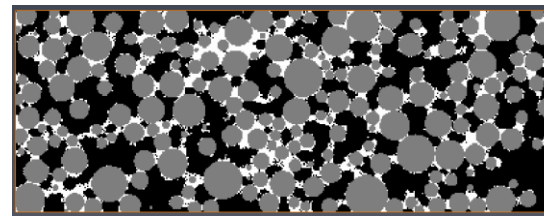
Electrochemical analysis

- Comparison of the performance of ternary DEM microstructures (incl. CBD) with image based ternary microstructures.
- Electrochemical simulations of DEM generated graded microstructures.

Current density (A/m^2)
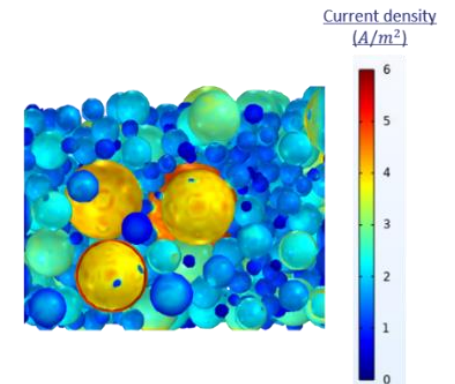
Thank you

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