eurofins
EAG Laboratories

Machine-Learning Assisted Analysis of Battery Electrode by PFIB-SEM Tomography

Ying Huang[1], Christopher Addiego[1], Xiuhong Han[1], Sarah Wang[2], Jiangtao Zhu[2], [1] Advanced Microscopy, Eurofins EAG Laboratories, Sunnyvale, CA [2] Advanced Microscopy, Eurofins Nanolab Technologies, Milpitas, CA
www.eag.com

INTRODUCTION

During battery cycling, particle morphological change, including cracking and disintegration, can lead to cell degradation [1]. These morphological degradations can range from micrometers to nanometers in scale and often require three-dimensional (3D) characterization. Plasma based focused ion beam-scanning electron microscopy (PFIB-SEM) tomography can access representative volume efficiently, while it is able to provide dataset with *nm* scale spatial resolution and hundreds of *μm* field of view [2].

Though the tomography techniques are well developed, gaining quantitative information from the representative 3D volume data is still demanding. Differentiating and extracting individual components of the battery from imaging dataset is one challenge, as the pixel value difference between the components can be minimal. On the other hand, statistical analysis of the active materials can boost the understanding of the electrode's structure-performance relationship, but it is challenging. Here, using a cycled commercial battery electrode, we present a machine-learning assisted imaging analysis approach to reveal its morphological properties with statistical significance. Our contributions are:

- Built a processing pipeline for a tomography dataset of battery cathode.
- Demonstrated how morphological information can be extracted from imaging dataset.
- Provided possibility of an end-to-end solution to facilitate electrode's development.

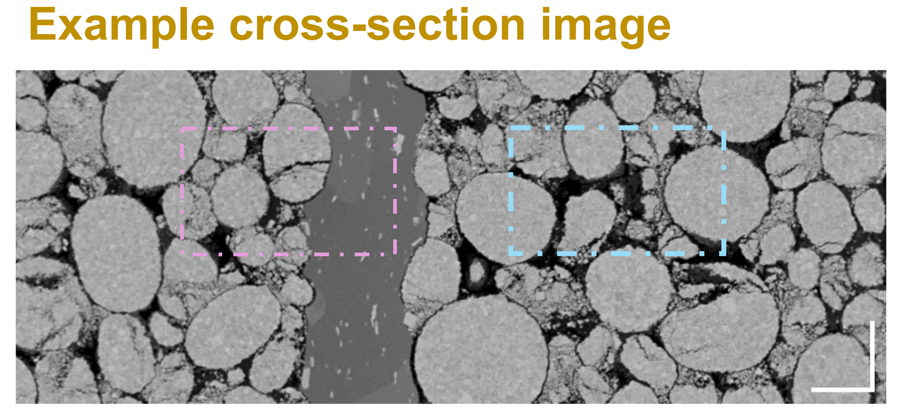
DATA COLLECTION

Sample is an electrode stack from a used commercial lithium-ion battery. The sample was mechanically cut and polished before imaging.

Imaging conditions

- Tool: ThermoFisher Helios 5 PFIB Uxe
- Backscattered electron image
- Physical Slice Thickness: 200 *nm*
- Voxel size of the collected dataset: 132 *nm* x 122 *nm* x 200 *nm*
- Processed volume: 136.6 *μm* x 47.0 *μm* x 45.6 *μm*

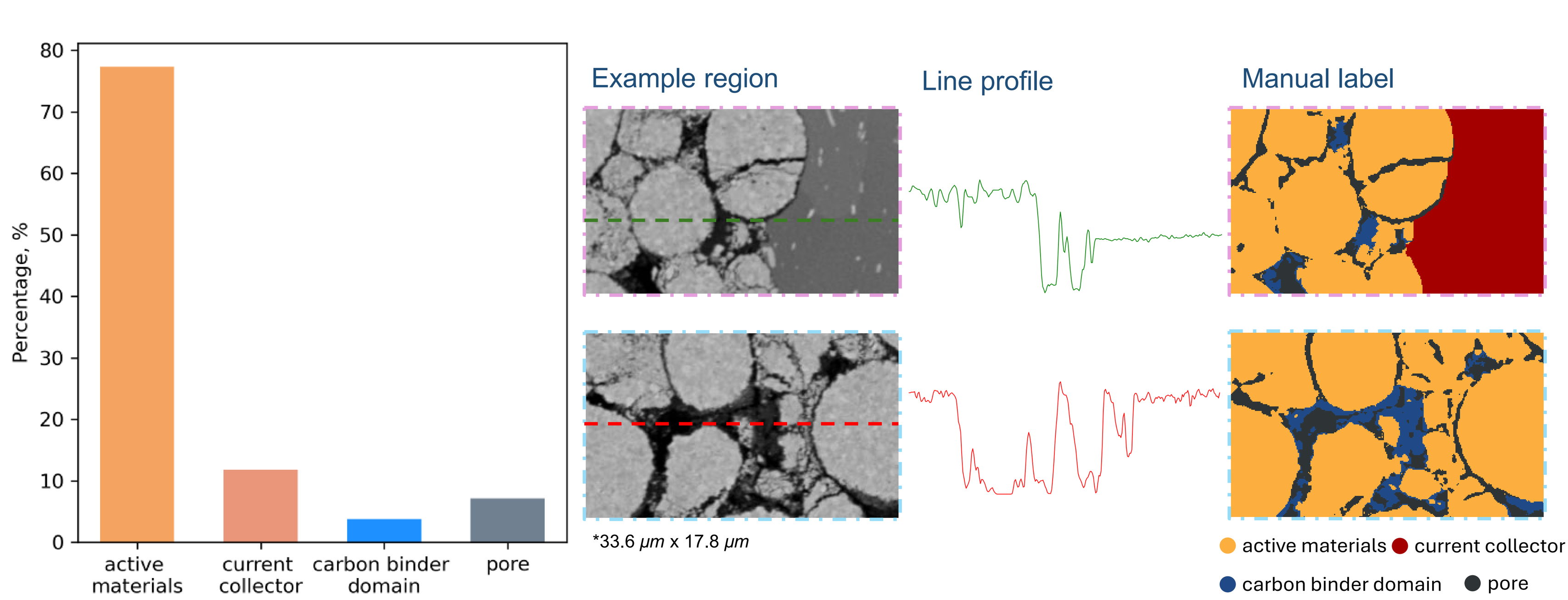
Example cross-section image



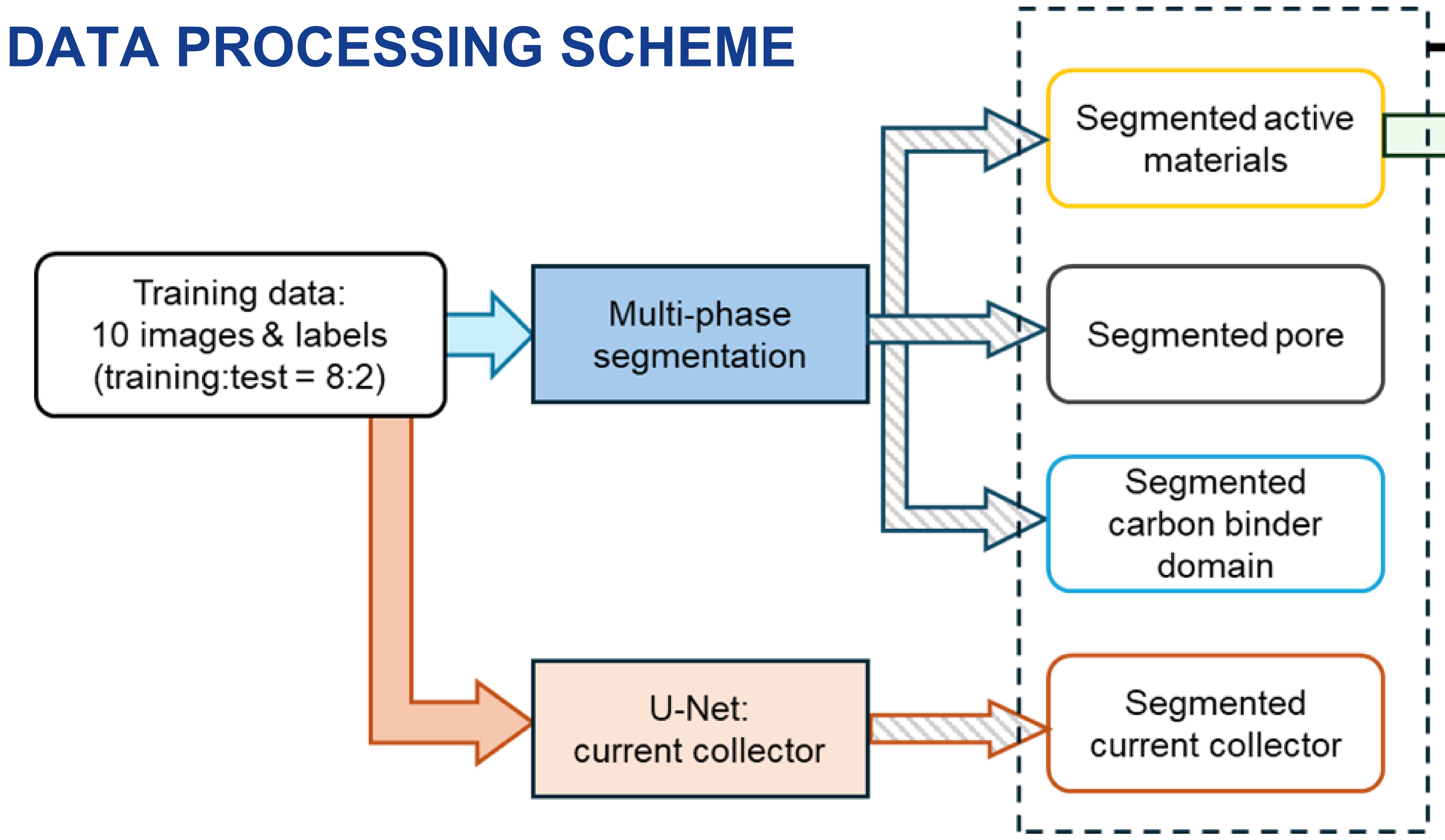
* *pre -processed (rotate and crop)* Scale bar: 10 *μm*

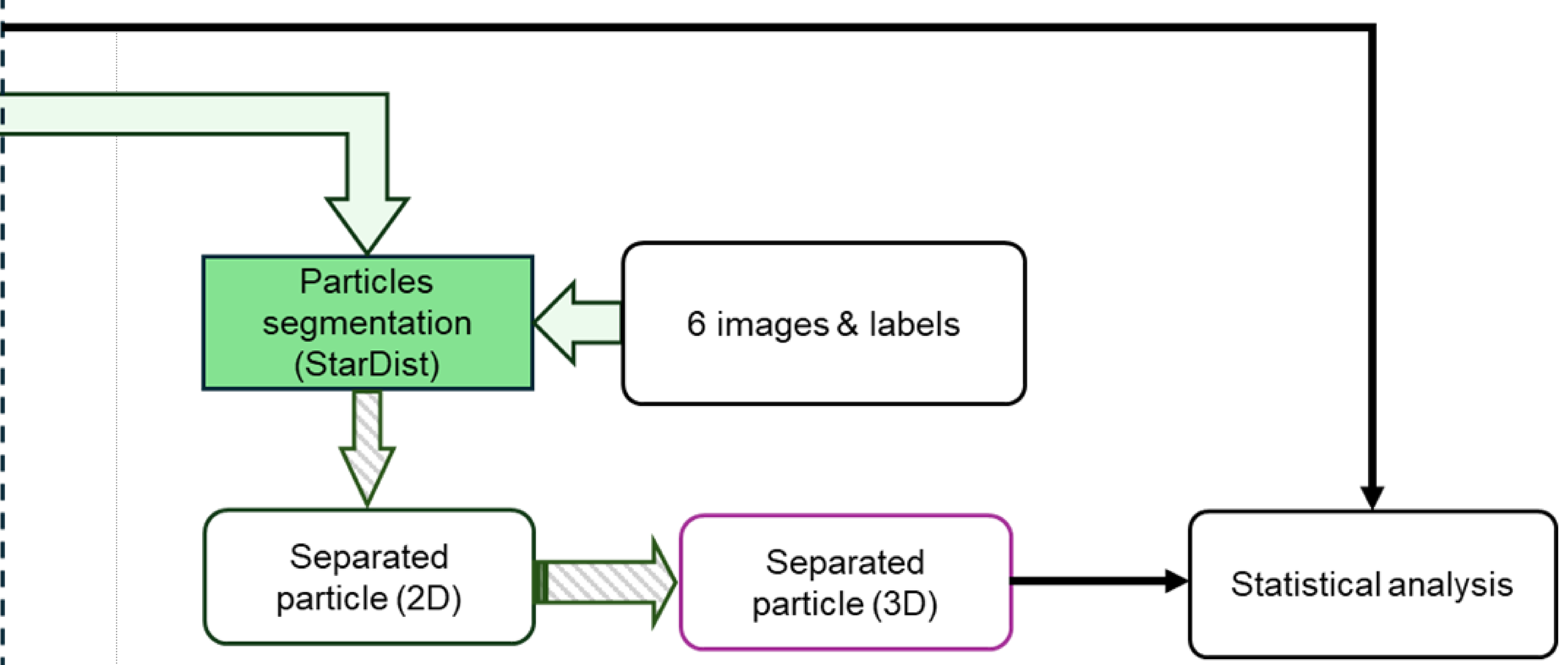
Imaging labeling

10 images were labeled for multi-phase segmentation. Data is imbalanced. Segmentation based on pixel values deference between components can be challenging.



DATA PROCESSING SCHEME



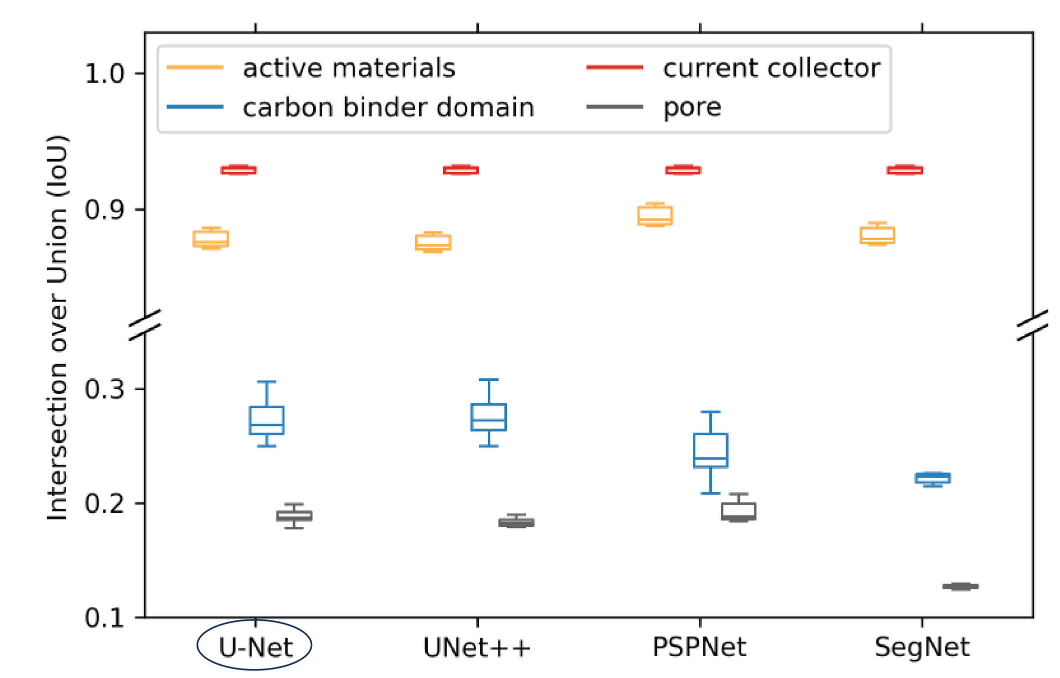


MULTI-PHASE SEGMENTATION

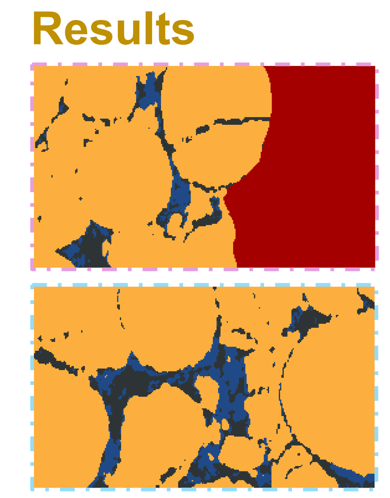
Four architectures were trained to achieve the multi-phase segmentation task. Both weighted loss function and unweighted loss function were tried to deal with the imbalanced data. Also, three different kinds of training dataset were compared: image patches, completed images and mixed patches and completed images. The ones trained with completed images provide the best results. The IoUs were calculated by comparing the prediction with the 10 labels. Eventually, the results of the best models in their own kinds were similar. The result of U-Net model was chosen for further analysis.

Best models:
**optimizer-Adam, learning rate-0.01, loss function – cross entropy*

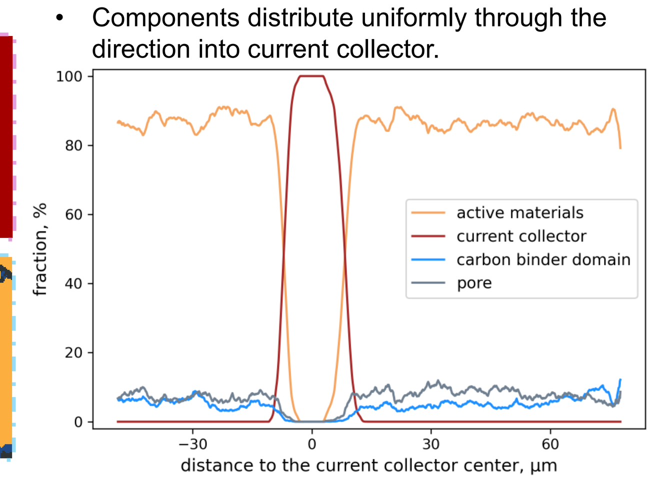
- U-Net: trained with unweighted loss function
- UNet++: trained with weighted loss function
- PSPNet: trained with unweighted loss function
- SegNet: trained with weighted loss function



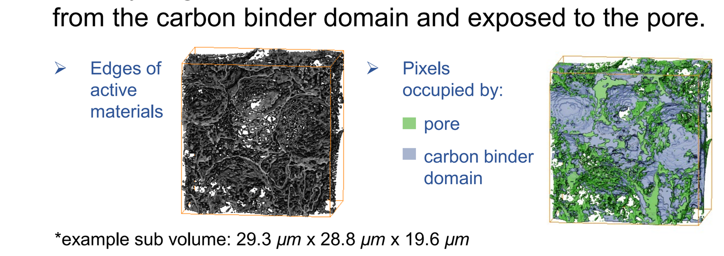
Results



- Components distribute uniformly through the direction into current collector.



- After cycling, most of the active materials are detached from the carbon binder domain and exposed to the pore.



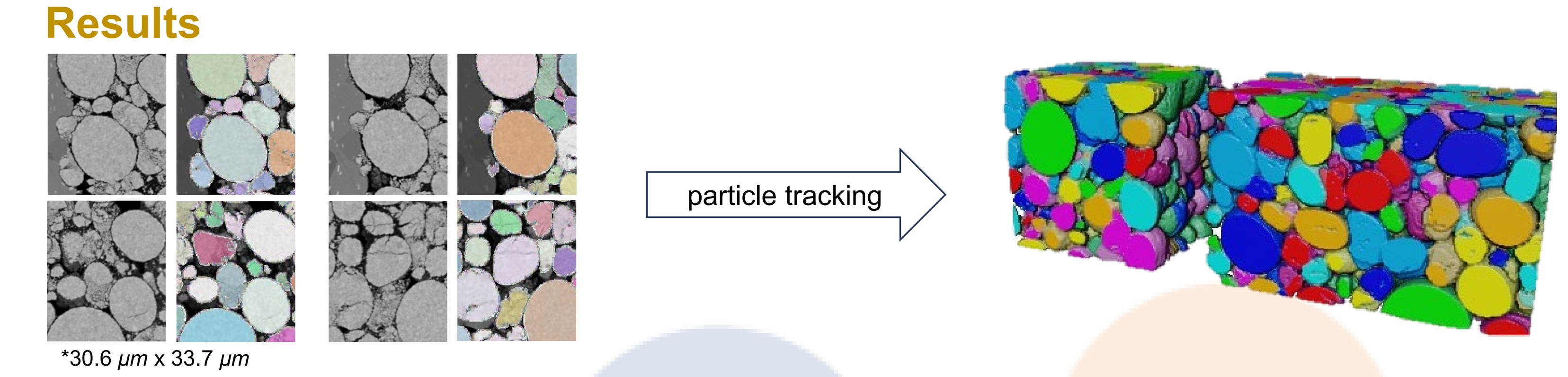
Component attached to active materials surface	Percentage
Pore	66.41%
Carbon binder domain	29.50%

Volume fraction occupied by different components:
•active materials – 76.61% •current collector – 11.95 % •carbon binder domain – 4.64% •pore - 6.80%

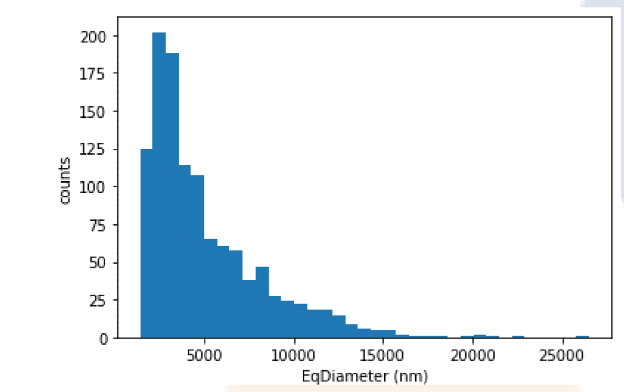
PARTICLE SEPARATION

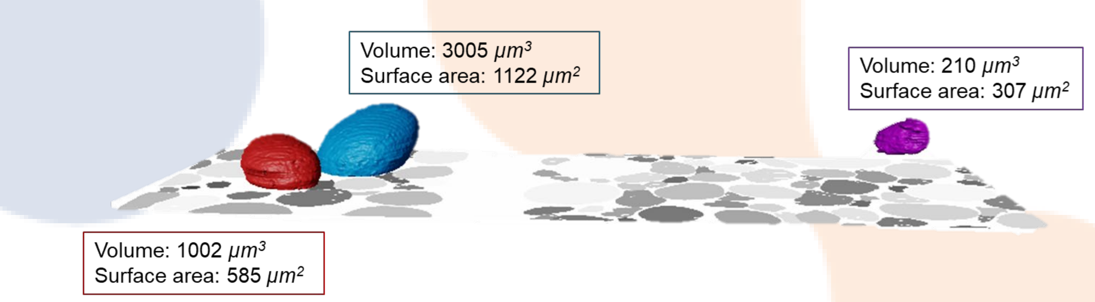
Instance segmentation was conducted with StarDist, a detection method for star-convex shapes. Six images were labeled and used as training set. Data augmentation, including random flipping and intensity changes, was applied. As results, more than 1,000 particles were extracted out as individual instances.

Results



- Active materials size distribution
- Properties of specific active material particles





CONCLUSIONS

A ML-aided pipeline was developed to reveal the 3D morphologic and structure information of individual particles within a thick battery cathode *via* PFIB-SEM. PFIB-SEM tomography combined with ML data processing is a versatile technique and can be applied to many other complicated materials systems where conventional segmentation methods fall short. Potentially, this method can be applied to understand local heterogeneity within one battery to dig out the cell degradation mechanism. Additionally, it can also be used to compare the particle morphology of the same type of batteries before and after cycling to find out the courses to capacity fade.

ACKNOWLEDGEMENT

We acknowledge Dr. Adam Stokes and Dr. Thomas F. Blum from Thermo Fisher Scientific for collecting the FIB-SEM data. We thank Tina Hsu and Christopher Kurtz for the help of battery breakdown.

[1] Xu, Rong, et al., *Journal of the Mechanics and Physics of Solids* **129** (2019): 160-183. [2] Burnett, T. L., et al., *Ultramicroscopy* **161** (2016): 119-129.