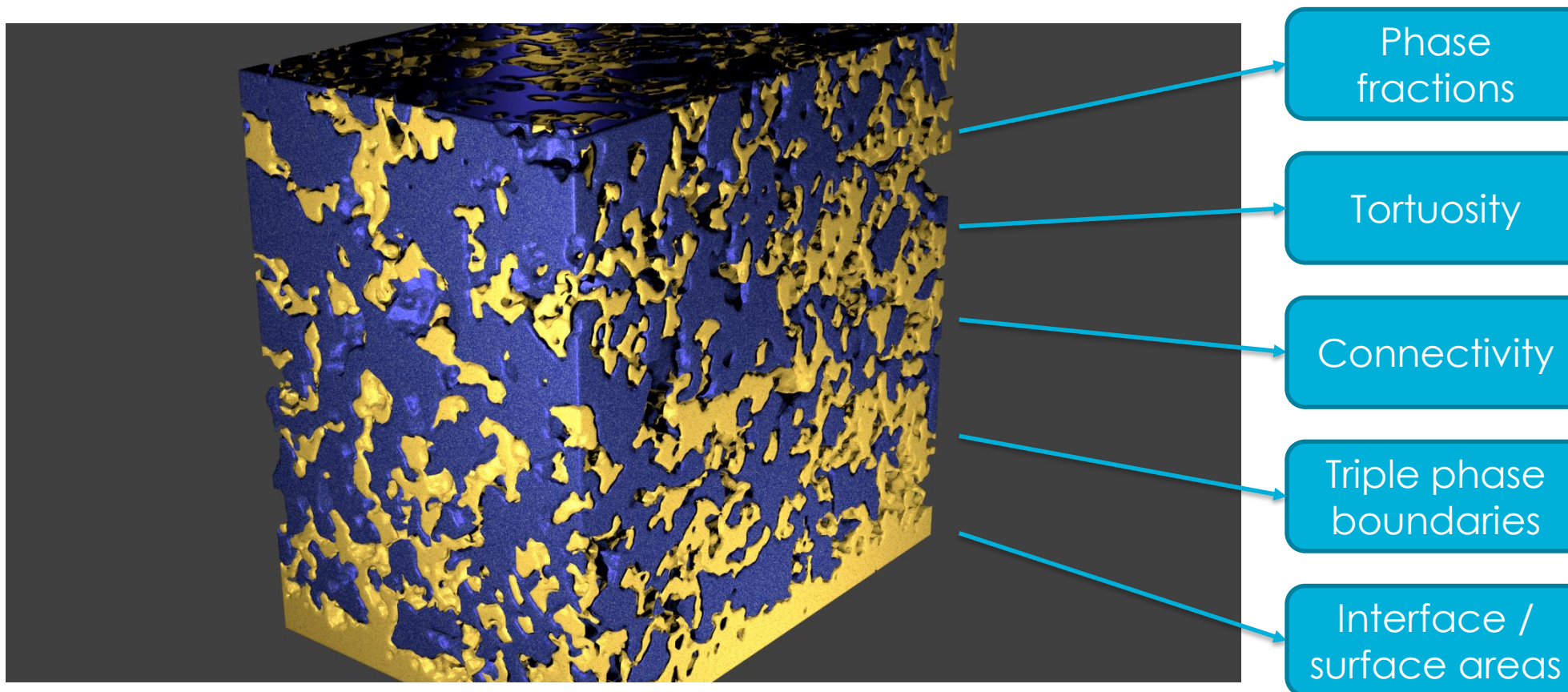


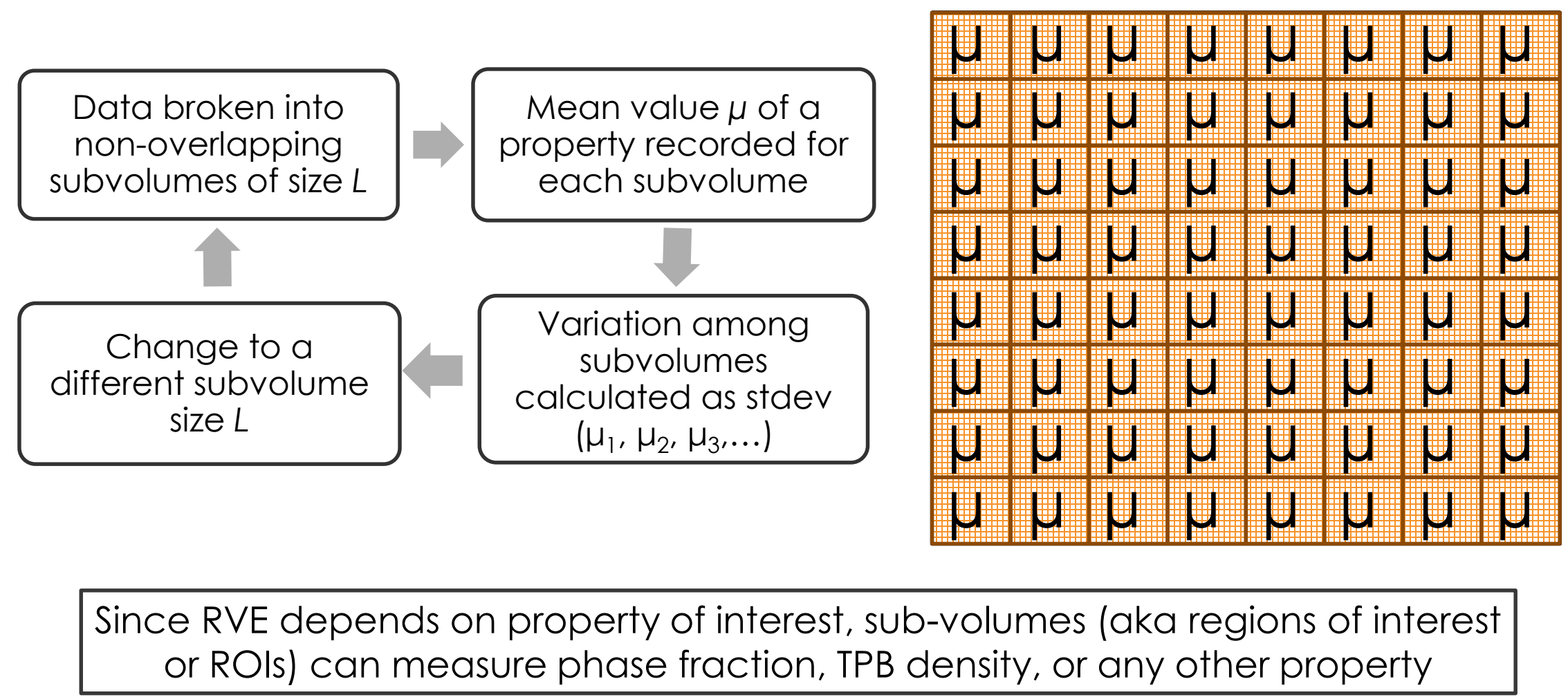
# Classifying Heterogeneity in SOFC Electrodes

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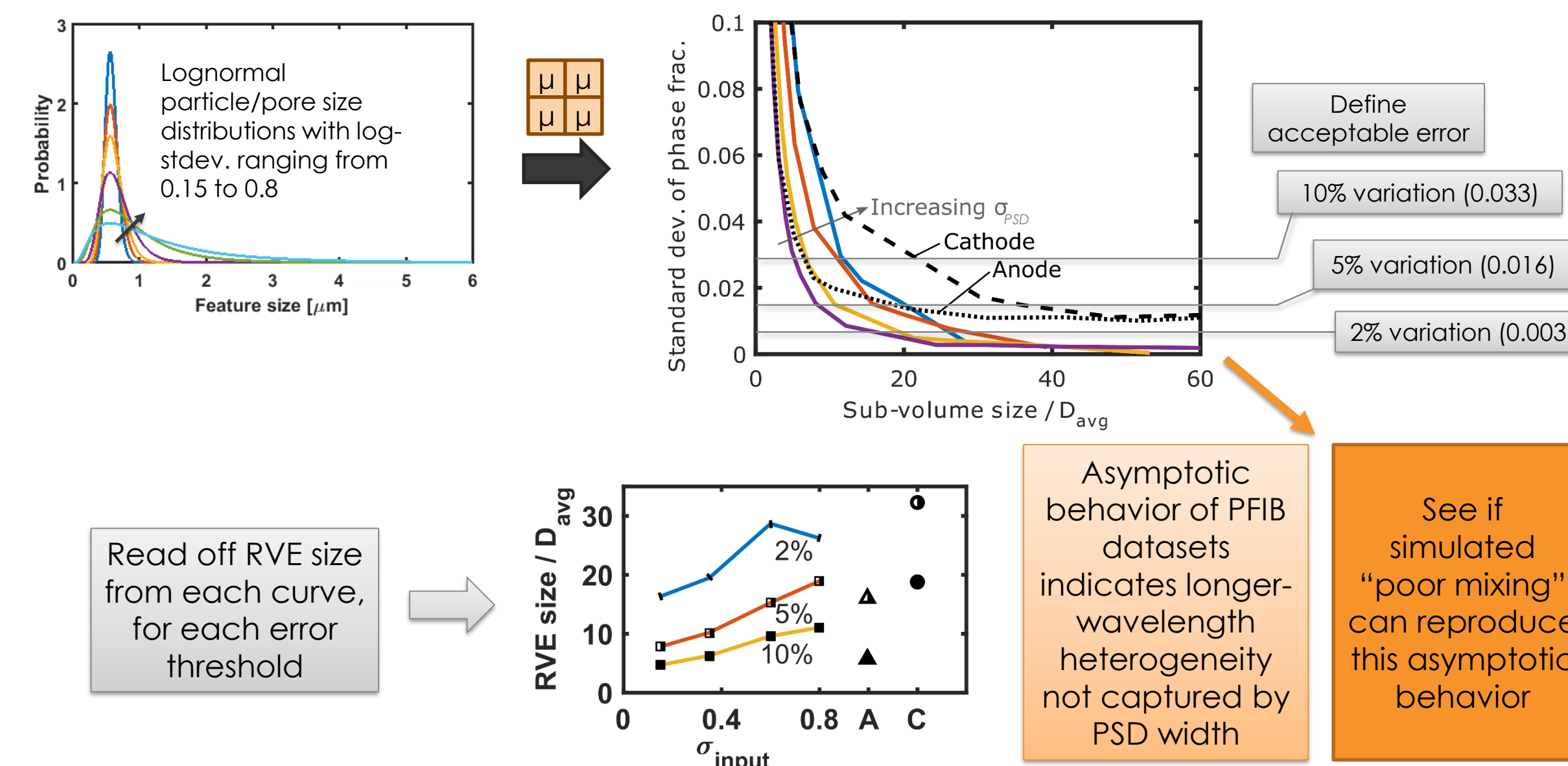
## Utility of Microstructure Imaging



## Quantifying Heterogeneity



## Heterogeneity from Size Distribution



## How Much to Measure?

Representative volume element (RVE)

- Reliably captures average of a property, within defined error
- Reliably captures entire distribution of a property, within defined error
- Reliably describes electrochemical processes, within defined error

**This work**

Prior work: Theoretical examination of RVE vs multiple properties. WM Harris & WKS Chiu, JPS v282 pg552 (2015)

Generally,  $L_{RVE} = 10-15 \times$  the size of one feature for lowest (1-2%) error

Two works\* in literature study RVE in 3D images of relatively homogeneous SOFC electrodes and agree:  $L_{RVE} = 10-15 \times D_{avg}$

\*J. Joos et al., Elect. Acta v81 pg 268 (2012)  
 J. Laurencin et al., JPS v198 pg 182 (2012)

**A clue:** "Certain features which may exist in real materials are not covered in this framework, including possible non-random agglomeration of particles of a certain phase, large disparities in structural/particle length scales, highly anisotropic features such as fibrous or planar structures, and graded structures." - Harris & Chiu, JPS 282 pg. 552 (2015)

Our imaged SOFC electrodes from several sources found larger RVEs required. Why? How to predict RVE size?

**Objective:** Investigate how different sources, scales of heterogeneity relate to RVE size

## Creating Heterogeneity in Synthetic Microstructures

- Objective is to span parameter space of different sources of heterogeneity and study the effect on e.g. RVE size.
- It would be difficult and costly to sweep across a parameter space with real electrodes and imaging studies.
- So, we will generate synthetic microstructures using DREAM.3D

**Different size distributions**

Lognormal particle / pore size distributions with same  $\mu$ , with log- $\sigma$  ranging from 0.15 to 0.8

Open-source software for generating synthetic multiphase microstructures: **DREAM.3D**

Realistic 3-phase microstructures with same average particle size, but narrower/broader size distributions

**"Well-mixedness" or aggregation of like particles**

Rep. volume vs Not a rep. volume

Ph. 1	Ph. 2	Ph. 3	N sampled
0.2	0.6	0.2	117
0.4	0.2	0.4	38
0.4	0.4	0.2	37
0.2	0.2	0.6	116
0.2	0.4	0.4	37
0.33	0.33	0.34	336
0.6	0.2	0.2	119

Generate library of "building blocks" of differing properties, e.g. phase fraction

Sample from blocks to achieve desired average and spread in property

**Experimental Data for Comparison**

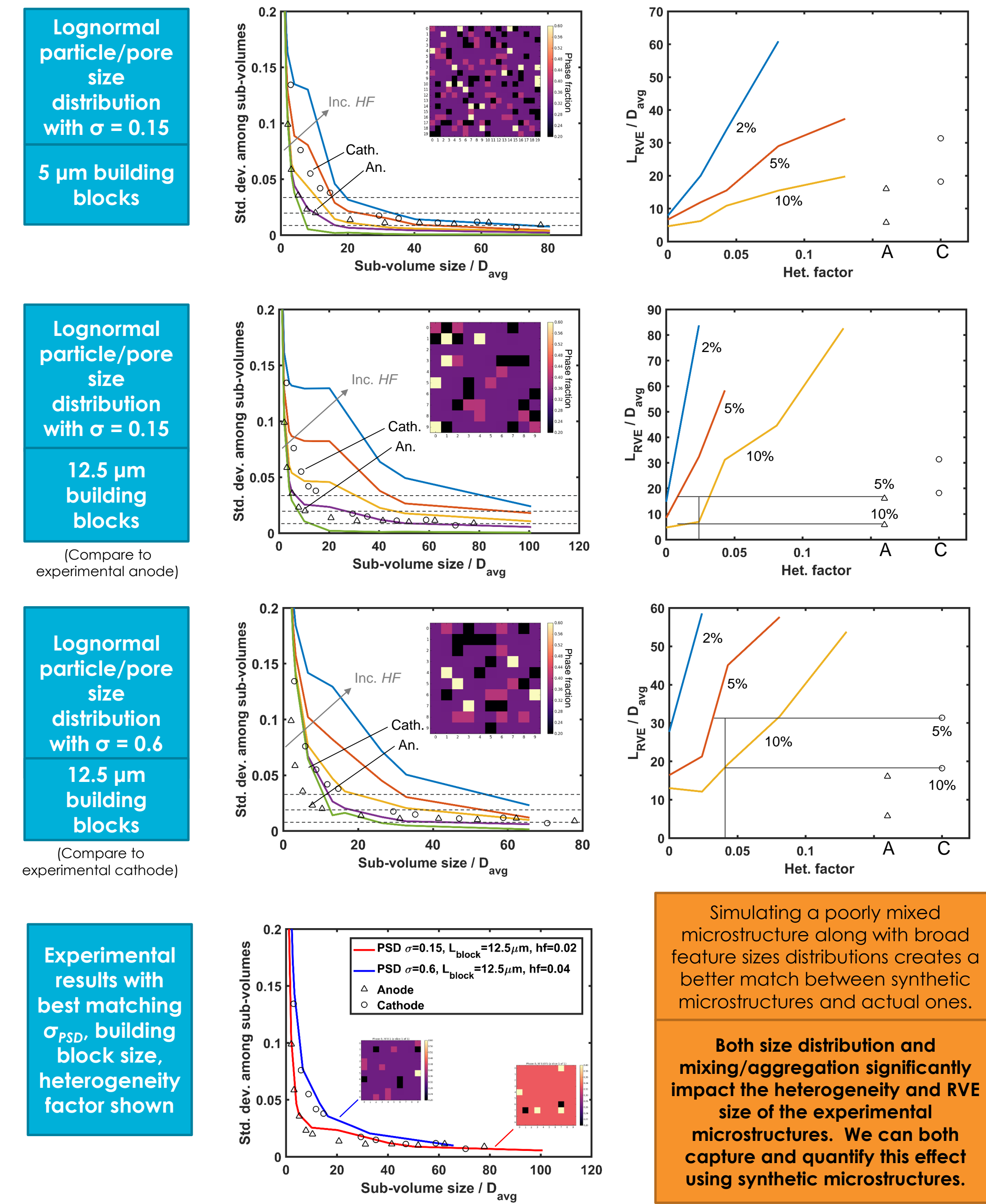
Large (ca. 100<sup>2</sup> μm<sup>2</sup>) areas of active cathode and anode regions from commercial planar cells collected using Xe-ion Plasma FIB-SEM at Carnegie Mellon University for comparison to synthetic microstructures

Population

Stdev ( $\mu_1, \mu_2, \mu_3, \dots$ )

## Heterogeneity from Poor Mixing

- Define "heterogeneity factor" as std. dev. among building blocks sampled. Vary HF across possible range.
- Initial parameters based on results from study above that best matched experimental data
- Building block size is important. In this study, **5 and 12.5 μm block sizes** considered
- Particle/pore size distributions important, as shown above. In this study, **lognormal-stdev. values of 0.15 and 0.6** considered



## Different Scales of Heterogeneity

**High-frequency heterogeneity**

- O(1 diam.)
- Mere existence of multiple phases
- All multi-phase systems have this, some are dominated by it

**Medium-frequency heterogeneity**

- O(10 diam.)
- Possible mechanisms:
  - Wide distribution of particle sizes
  - Non-random aggregation of particles

**Low-frequency heterogeneity**

- O(100 diam.)
- Possible mechanisms:
  - Rare but regular inclusion of a large particle
  - Gradient in sample

Domain of Harris & Chiu Model

Particularly large features