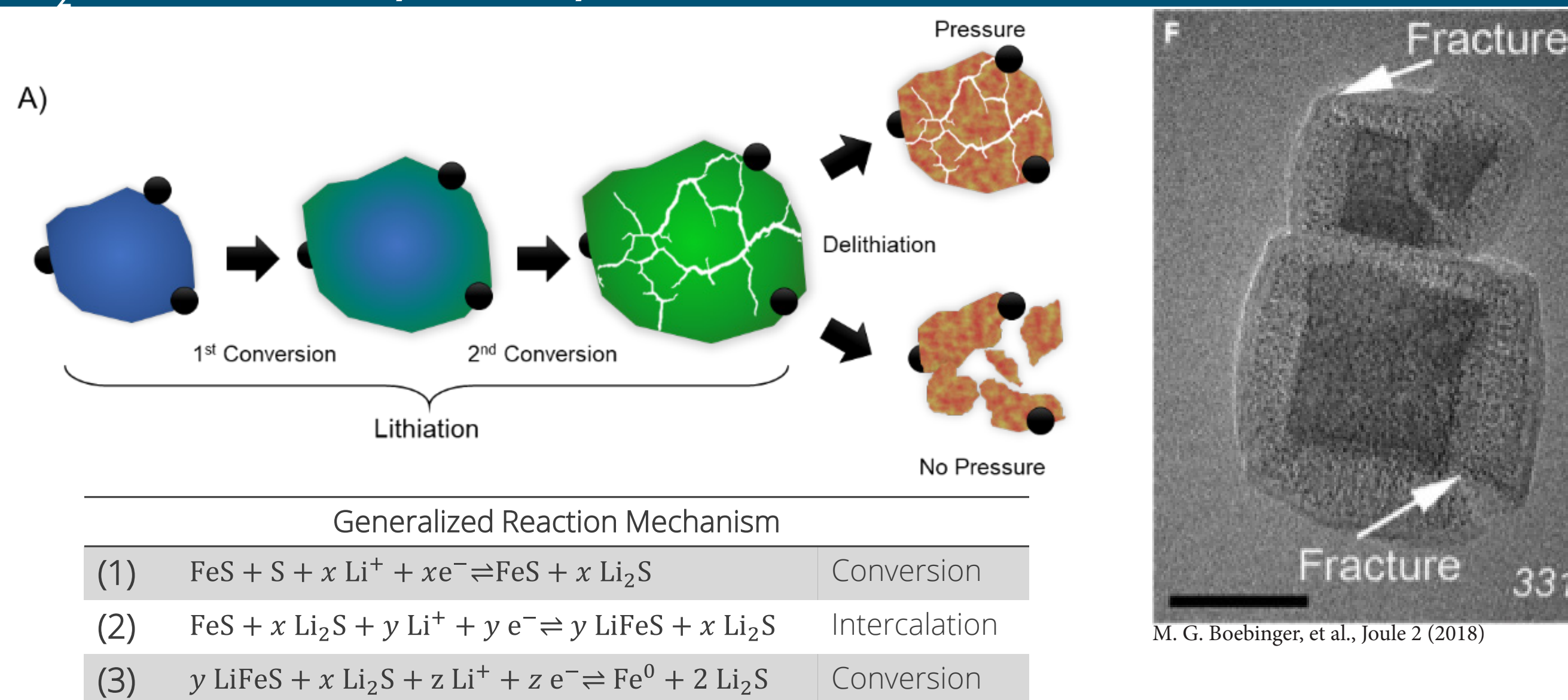


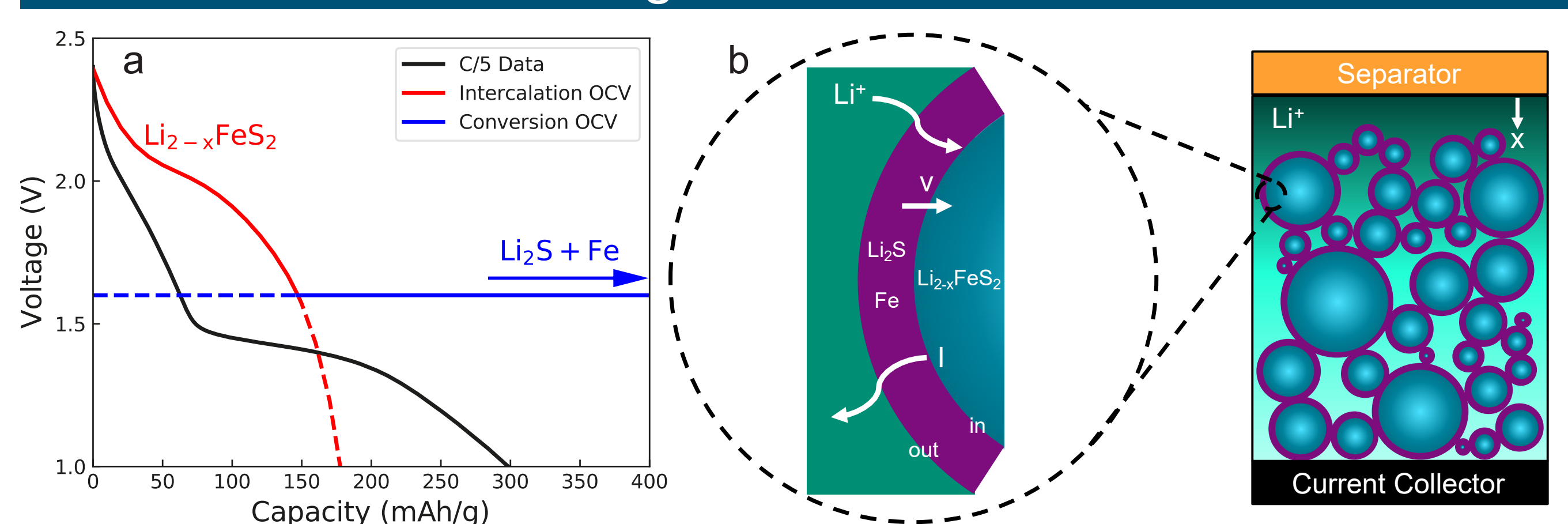
Modeling Coupled Electrochemistry and Mechanics for Conversion Cathodes

Scott A. Roberts (sarober@sandia.gov) and Jeffrey S. Horner

FeS₂ as a secondary battery cathode: Conversion reactions are brutal¹



P2D modeling for conversion cathodes²



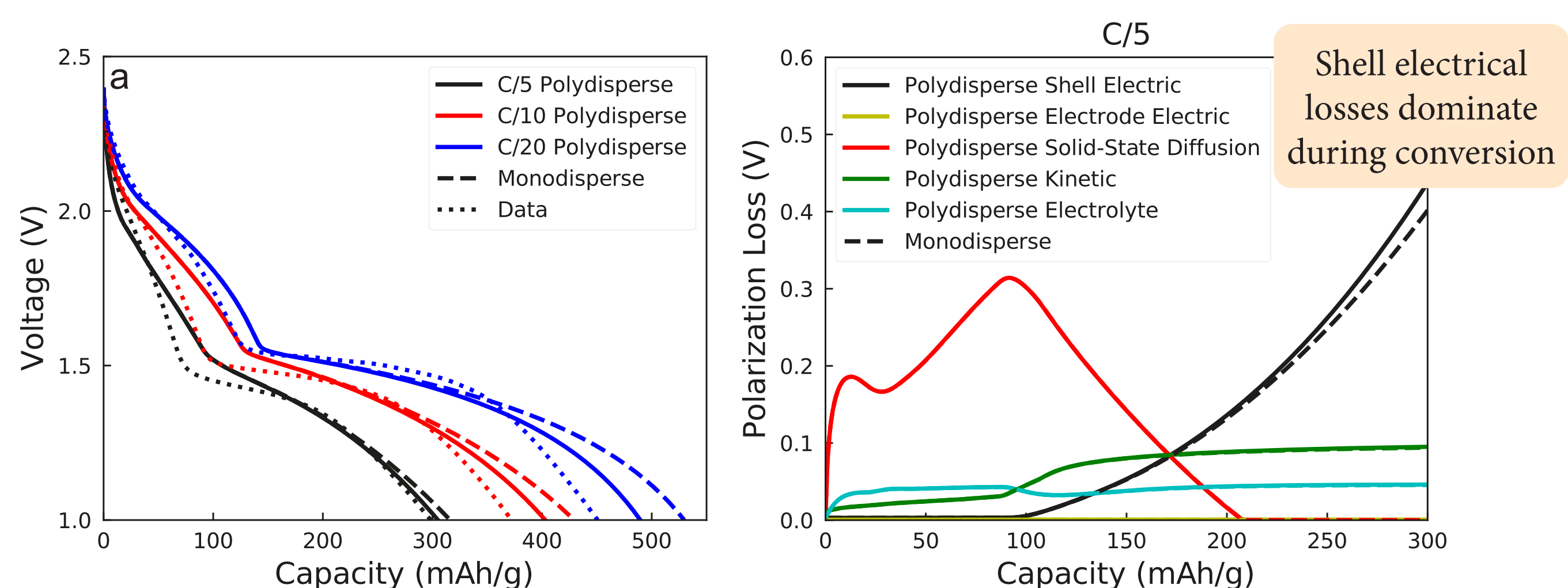
Additional equations specified for the converted particle shell region (purple Li₂S in above image)

$$\frac{\partial \delta}{\partial t} = -\frac{j_{\text{conv}} M_{\text{FeS}_2}}{\rho \left(4 - 2 \frac{C_{\text{Li}}}{C_{\text{max}}}\right)}$$

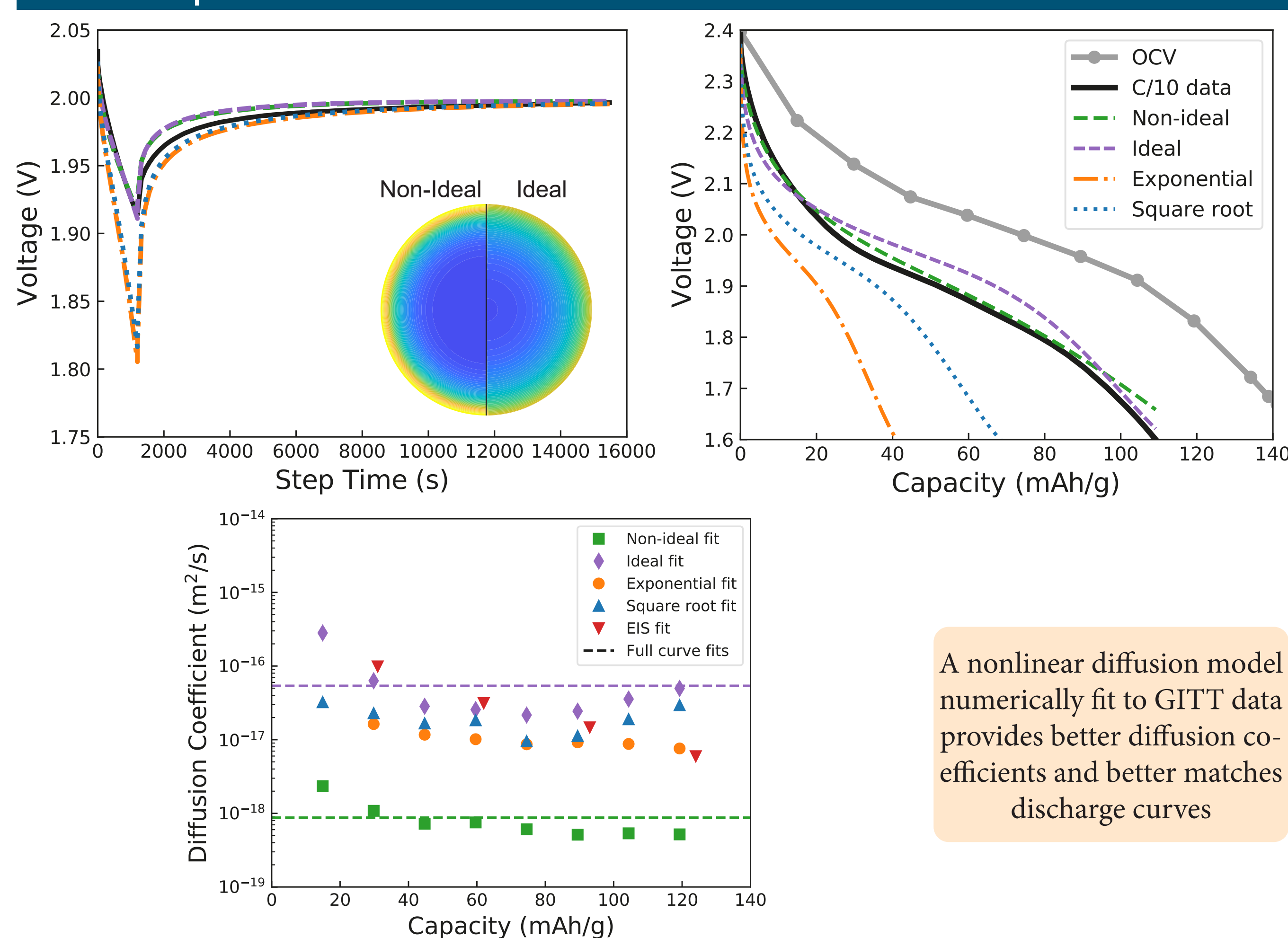
$$V_{s,\text{in}} = V_{s,\text{out}} - \frac{j F R_c^2}{\kappa_{\text{shell}}} \left(\frac{1}{R_c} - \frac{1}{R_p} \right)$$

$$V_{l,\text{in}} = V_{l,\text{out}} + \ln \left(\frac{C_{\text{Li}^+,\text{in}}}{C_{\text{Li}^+,\text{out}}} \right) \frac{RT}{F}$$

$$C_{\text{Li}^+,\text{in}} = C_{\text{Li}^+,\text{out}} + \frac{j R_c^2}{2 D_{\text{shell}}} \left(\frac{1}{R_c} - \frac{1}{R_p} \right)$$

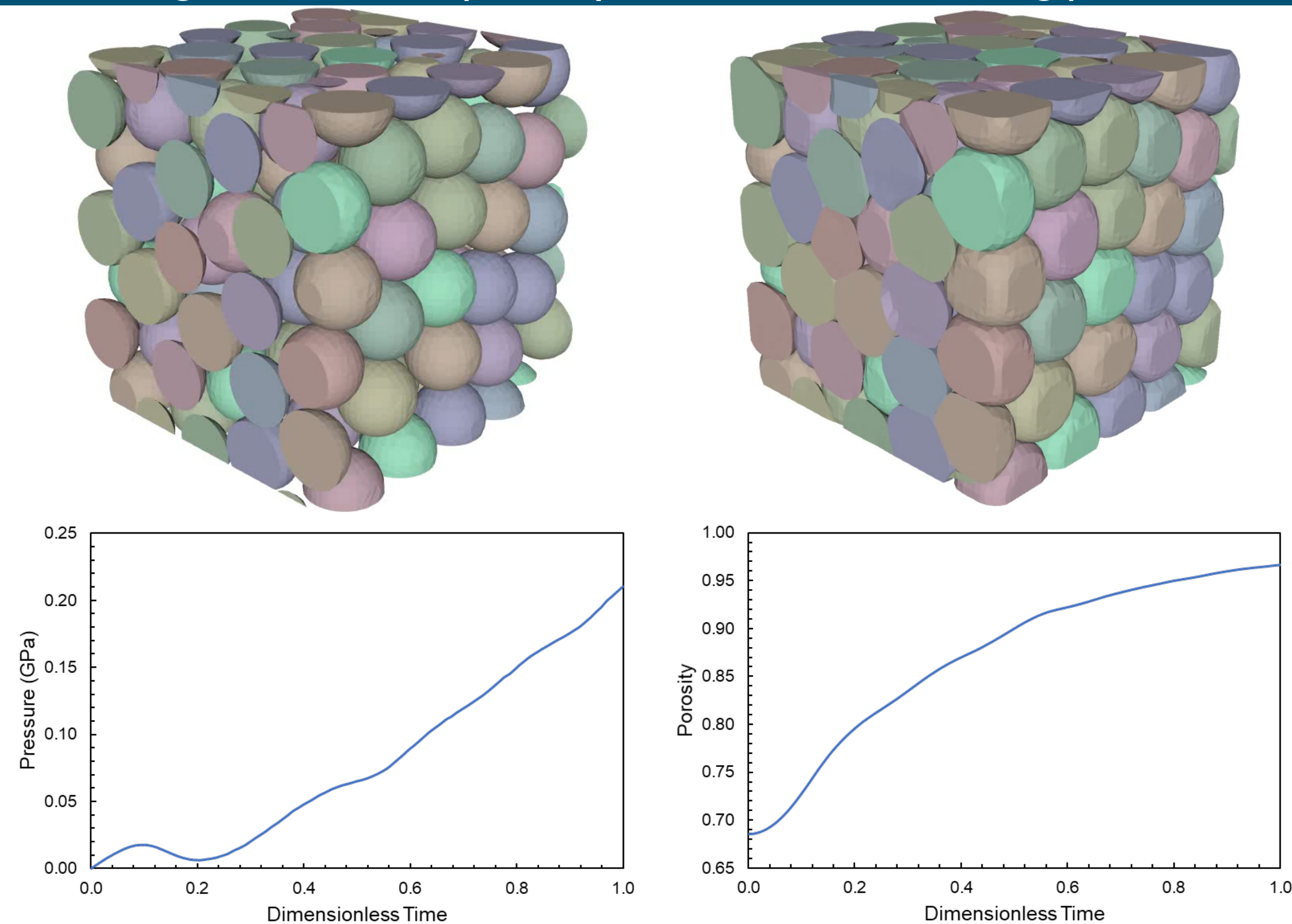


Improved extraction of diffusion coefficients from GITT³



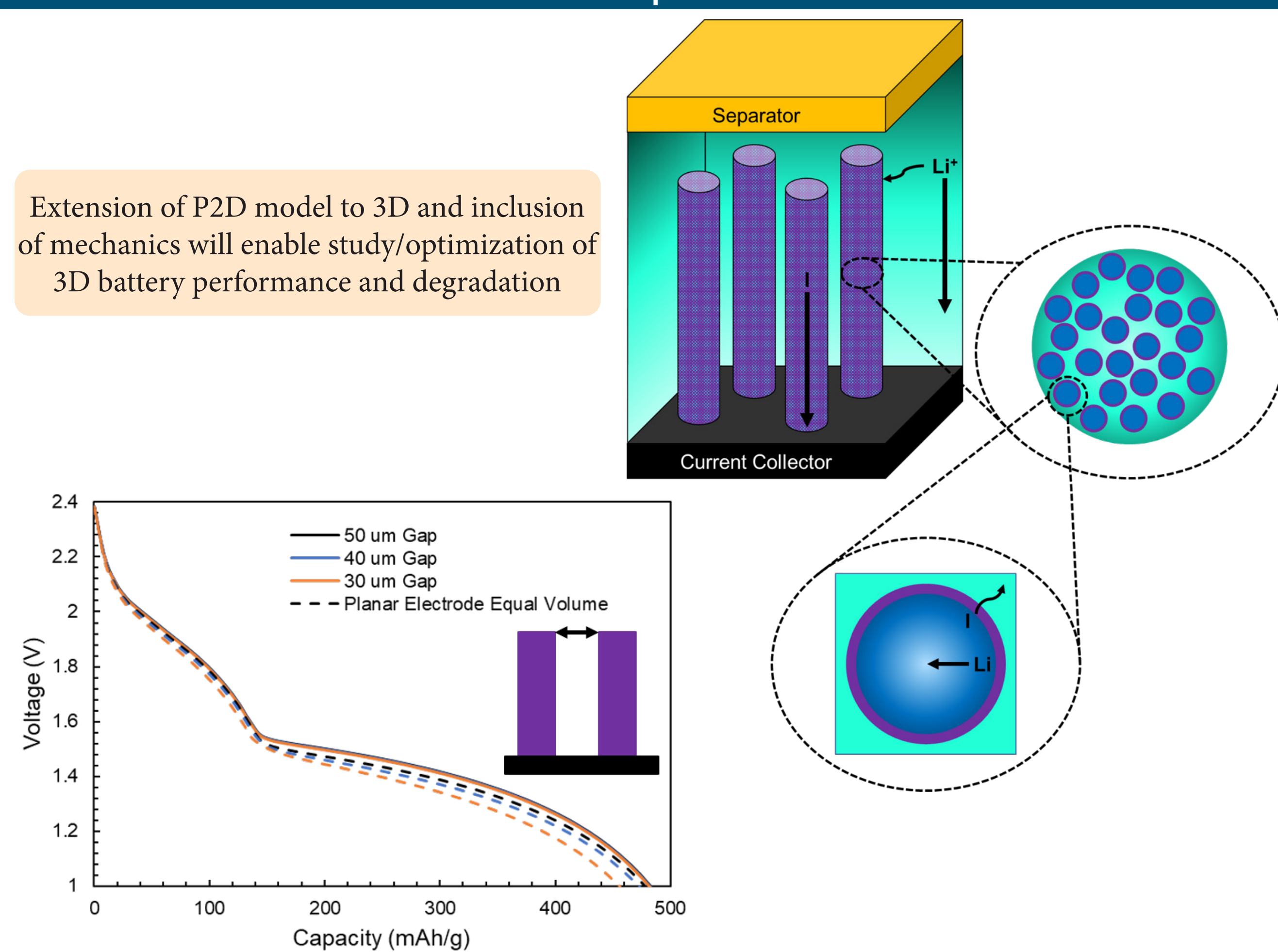
A nonlinear diffusion model numerically fit to GITT data provides better diffusion coefficients and better matches discharge curves

Stress generation and porosity evolution in converting particles^{1,4}

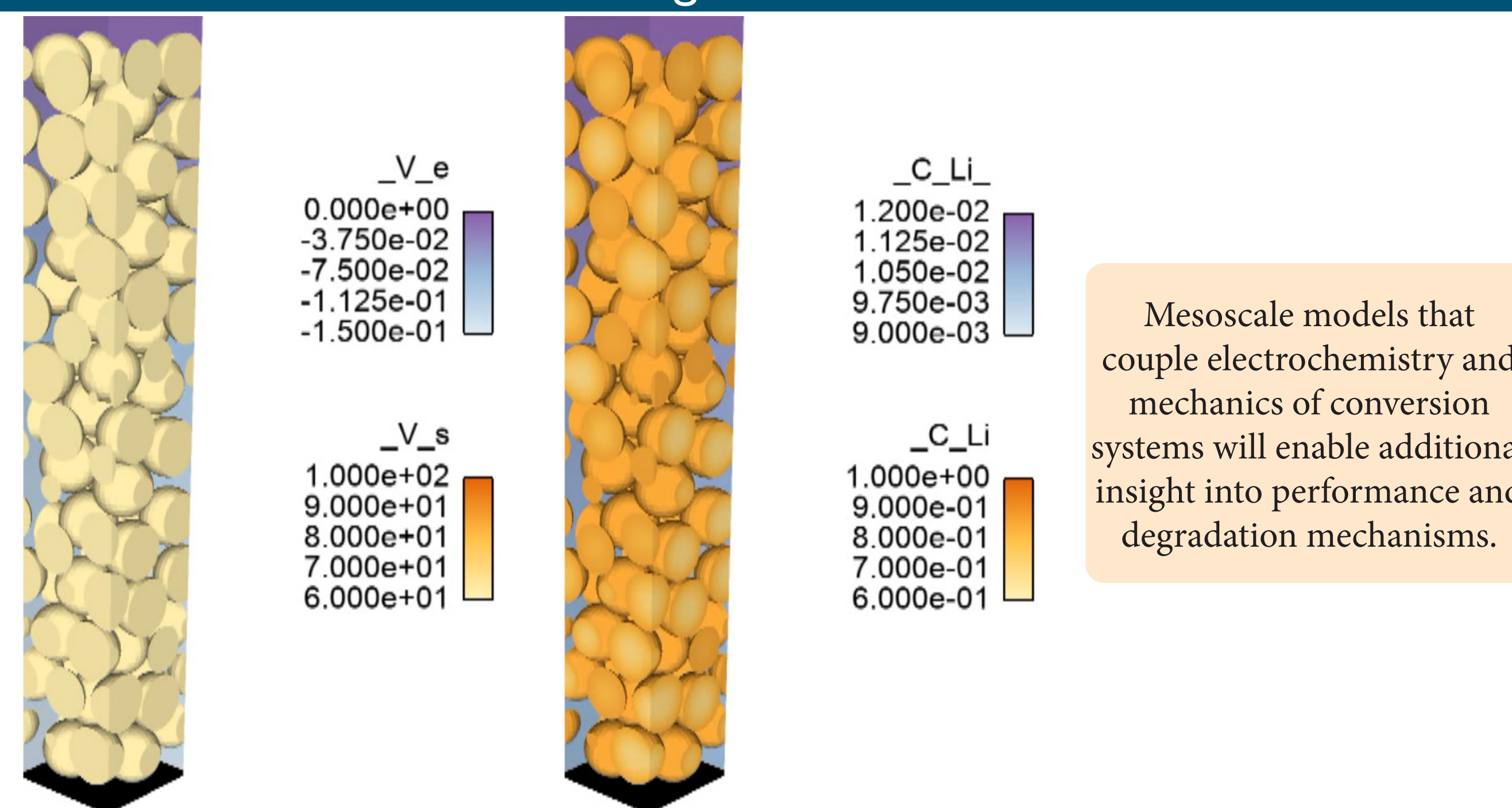


Pressure and porosity evolution during conversion highly nonlinear, requiring contact mechanics model

P3.5D model for pillar electrodes⁵



Mesoscale modeling of conversion electrodes⁴



Mesoscale models that couple electrochemistry and mechanics of conversion systems will enable additional insight into performance and degradation mechanisms.

References

1. D. S. Ashby, et al., "Understanding the Electrochemical Performance of FeS₂ Conversion Cathodes," in press at ACS Applied Materials & Interfaces
2. J. S. Horner, et al., "A Pseudo-Two-Dimensional (P2D) Model for FeS₂ Conversion Cathode Batteries" under review
3. J. S. Horner, et al., "Electrochemical Modeling of GITT Measurements for Improved Solid-State Diffusion Coefficient Evaluation," ACS Applied Energy Materials (2021), doi: 10.1021/acsami.1c02218
4. J. S. Horner, et al., To be a FeS₂ mesoscale modeling paper submitted this year
5. J. S. Horner, et al., To be a P3.5D FeS₂ modeling paper submitted this year