Topology optimization of porous electrodes for redox flow batteries using the finite element method

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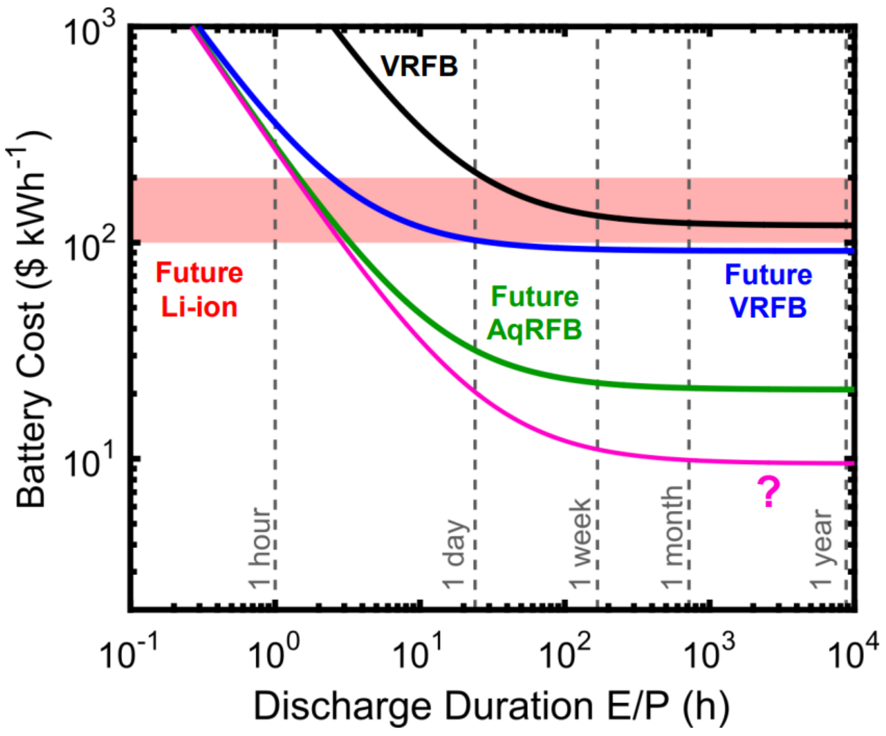
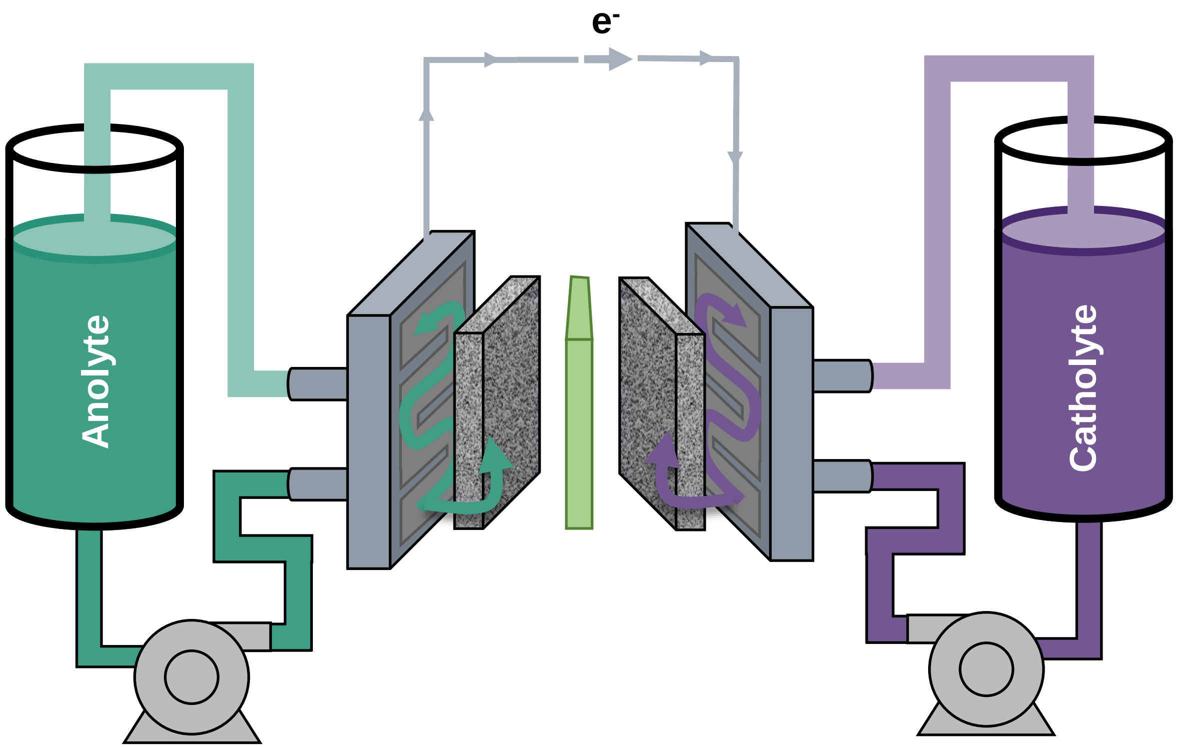
Redox Flow Batteries (RFBs)

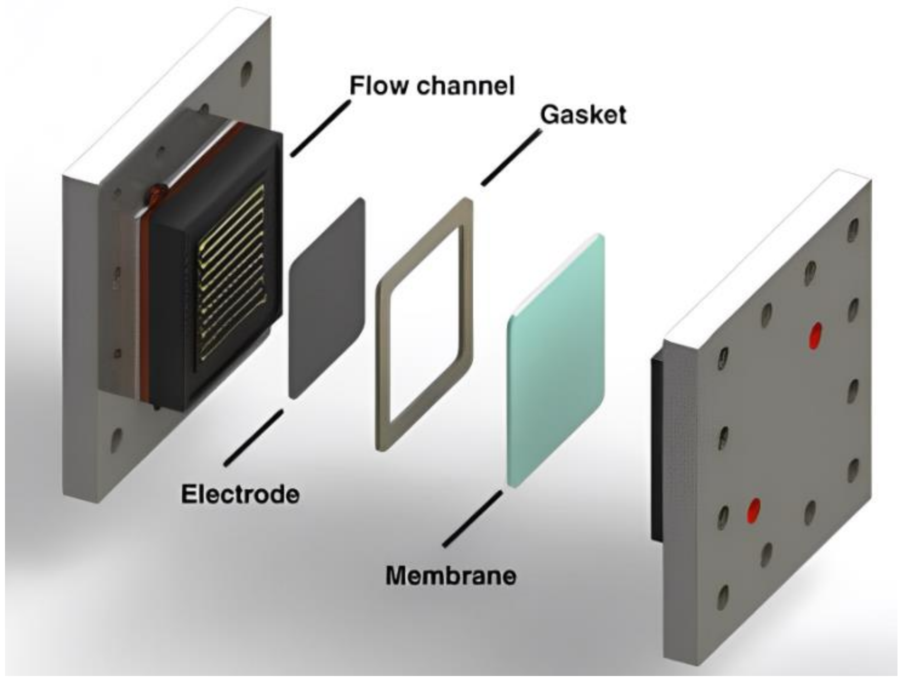
- Inexpensive durable energy storage
- Cost efficiency for grid-scale
- Decoupled energy and power



Darling & Gallagher et al., Energy Environ. Sci., 7 (2014) 3459. Dmello & Milshtein et al., J. Power Sources, 330 (2016) 261.

RFB Mechanism

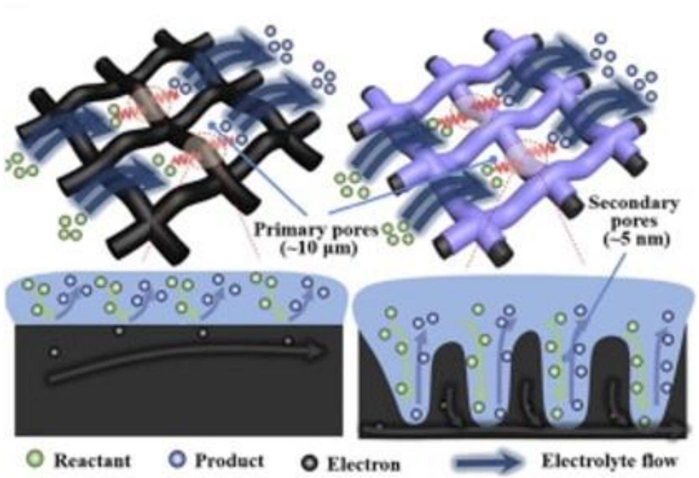




Su et al., in Rechargeable Batteries, Springer, (2015) 673.

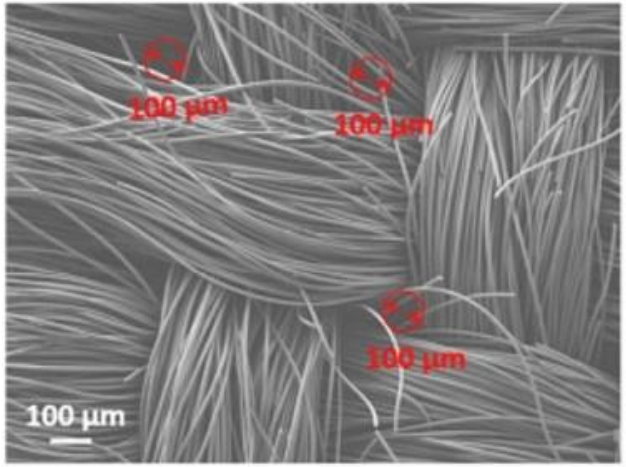
Electrodes: Performance-Defining Components

- Where the redox processes occur
- Tailoring the electrodes to improve performance:



Structure (micro-, macro-)

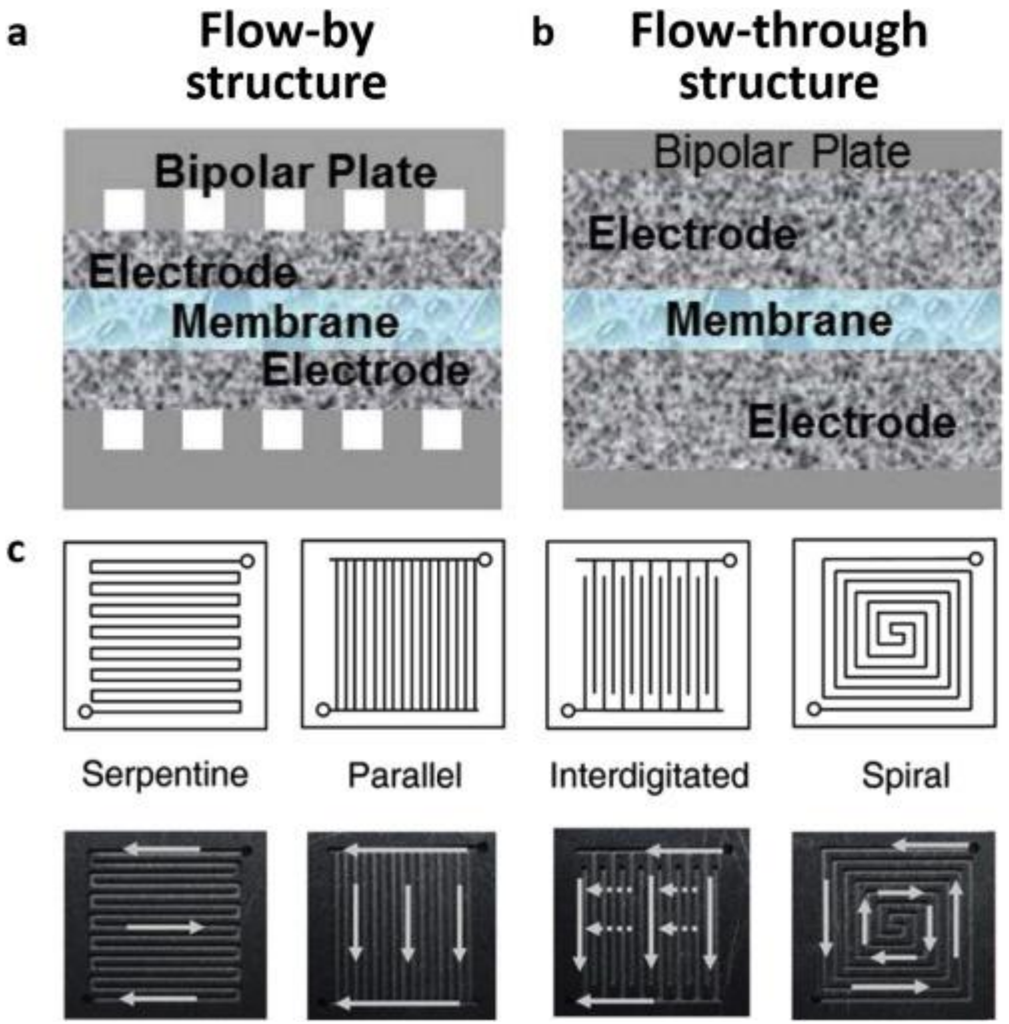
Surface properties



Zhou et al., J. Power Sources., 339 (2017)

Engineering Porous Electrodes

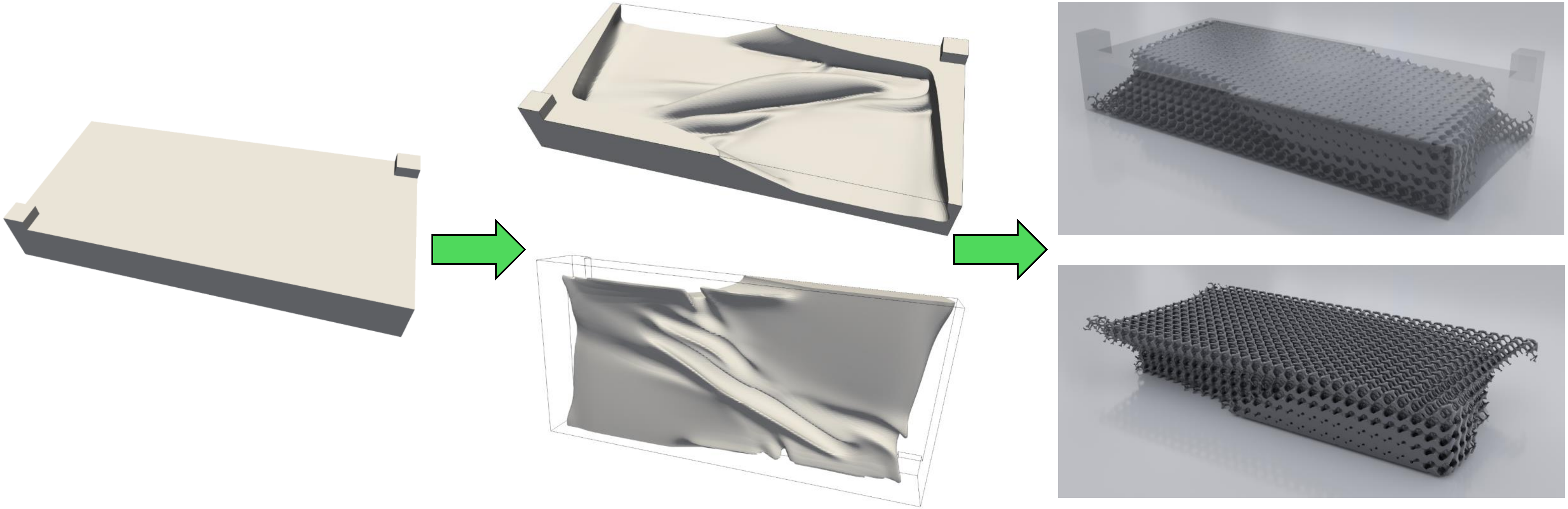
- What we want?
  - Surface area ↑
  - Mass transport ↑
  - Pressure drop ↓
  - Electrochemical activity ↑
  - Mechanical stability ↑



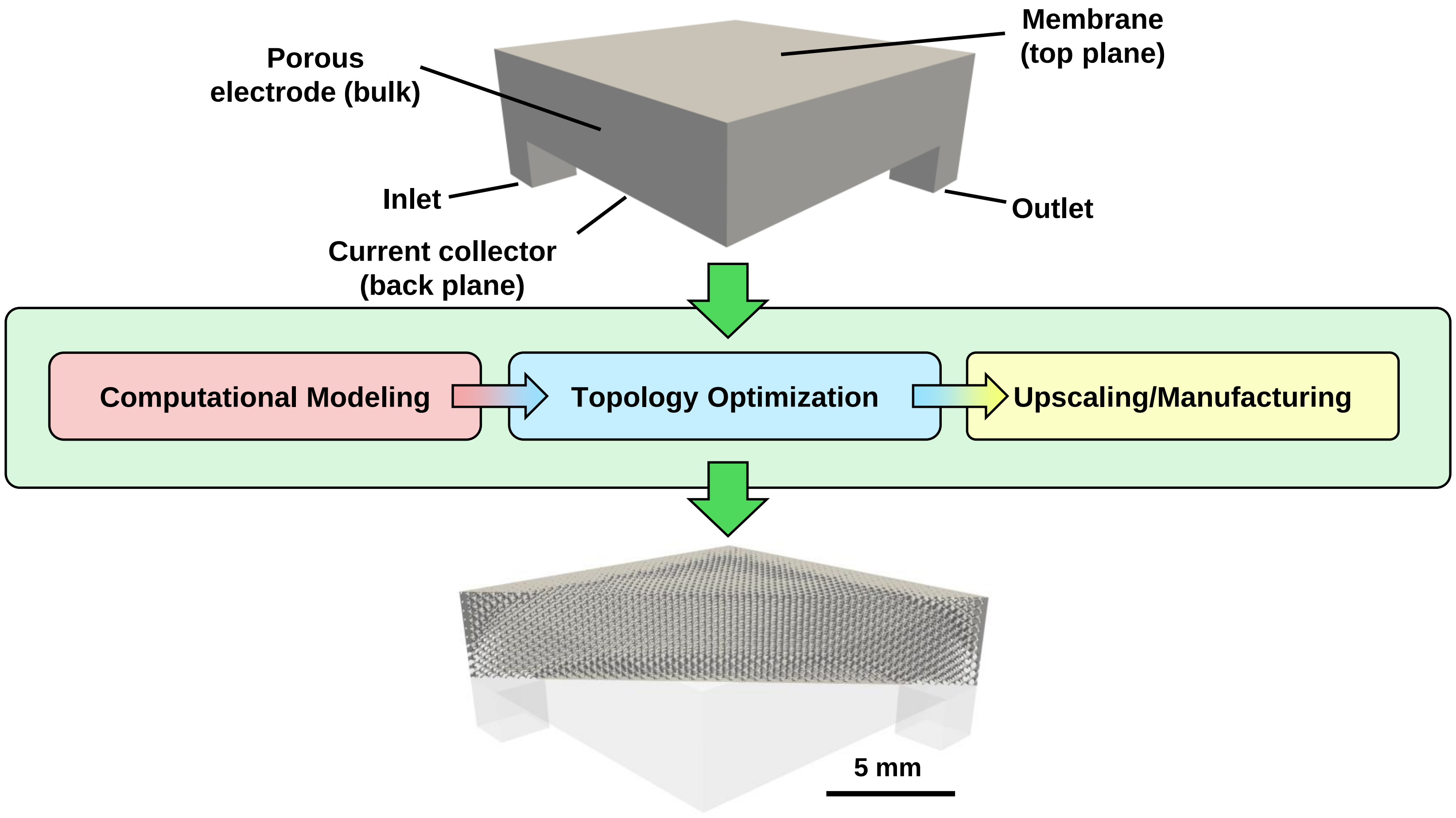
Zhou et al., J. Power Sources., 339 (2017)

Electrode Design via Engineering Optimization?

- Inverse design of electrodes for maximizing performance

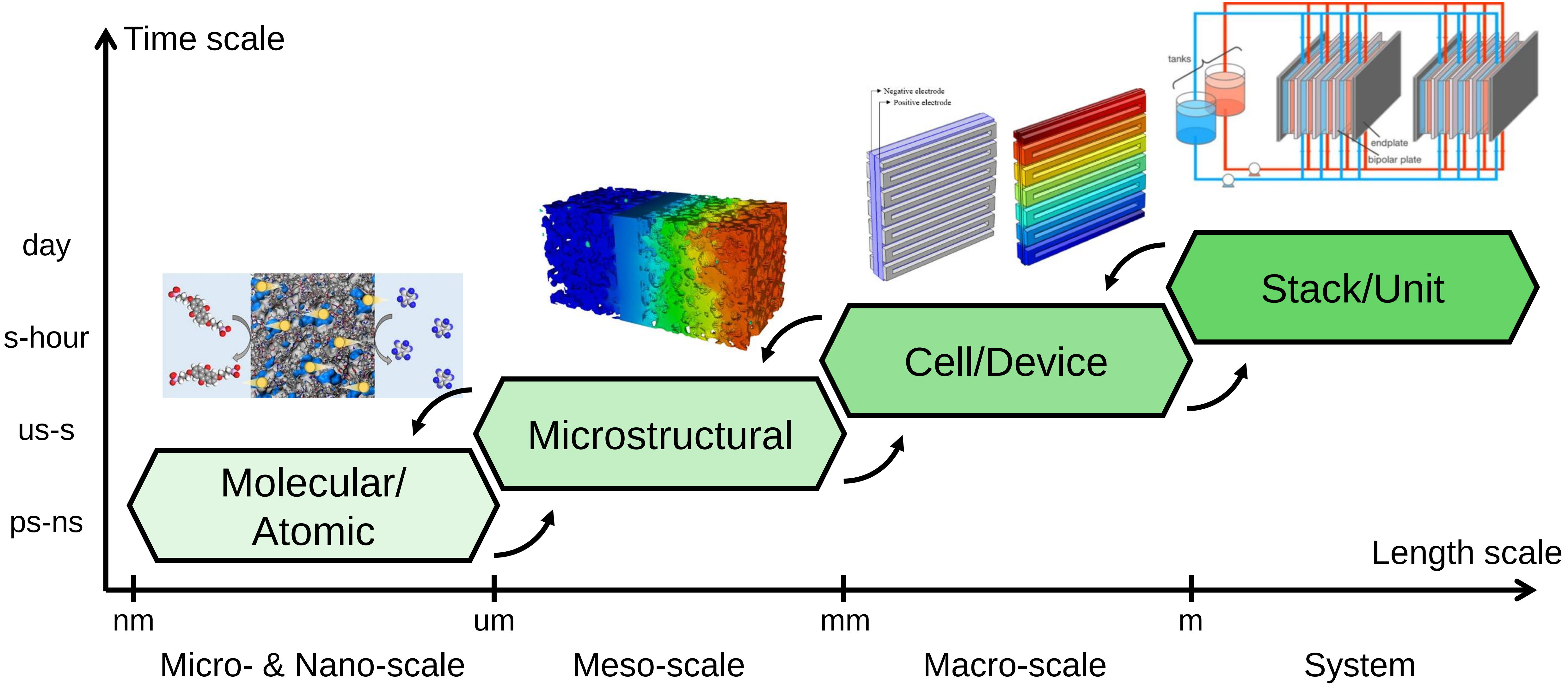


Modeling Workflow



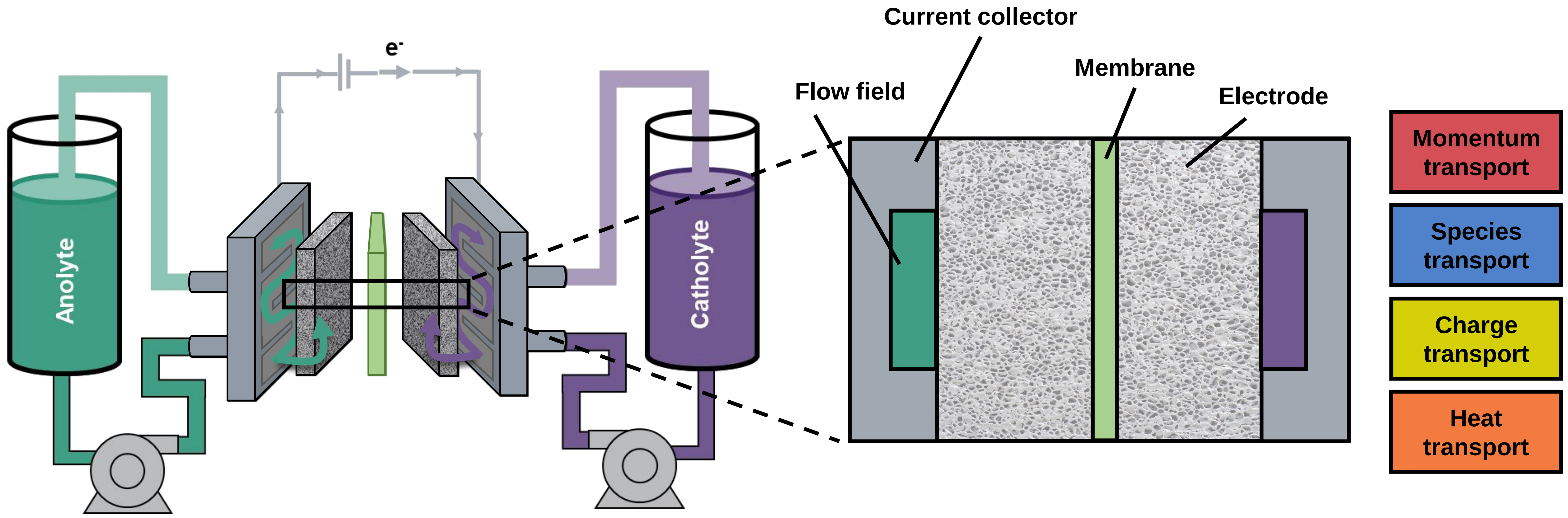
Computational Modeling of RFB Processes

Different Length & Time Scales in RFBs



Ye et al., Angew. Chem., 61 (2023) e202207580. De Lauri et al., ACS Appl. Energy Mater., 4 (2021) 13847.
Ali et al., J. Energy Storage, 28 (2020) 101208. Ma et al., J. Electrochem. Soc., 165 (2018) A2209.

RFBs as Multi-Physics Redox Systems



Mathematical Modeling of RFBs

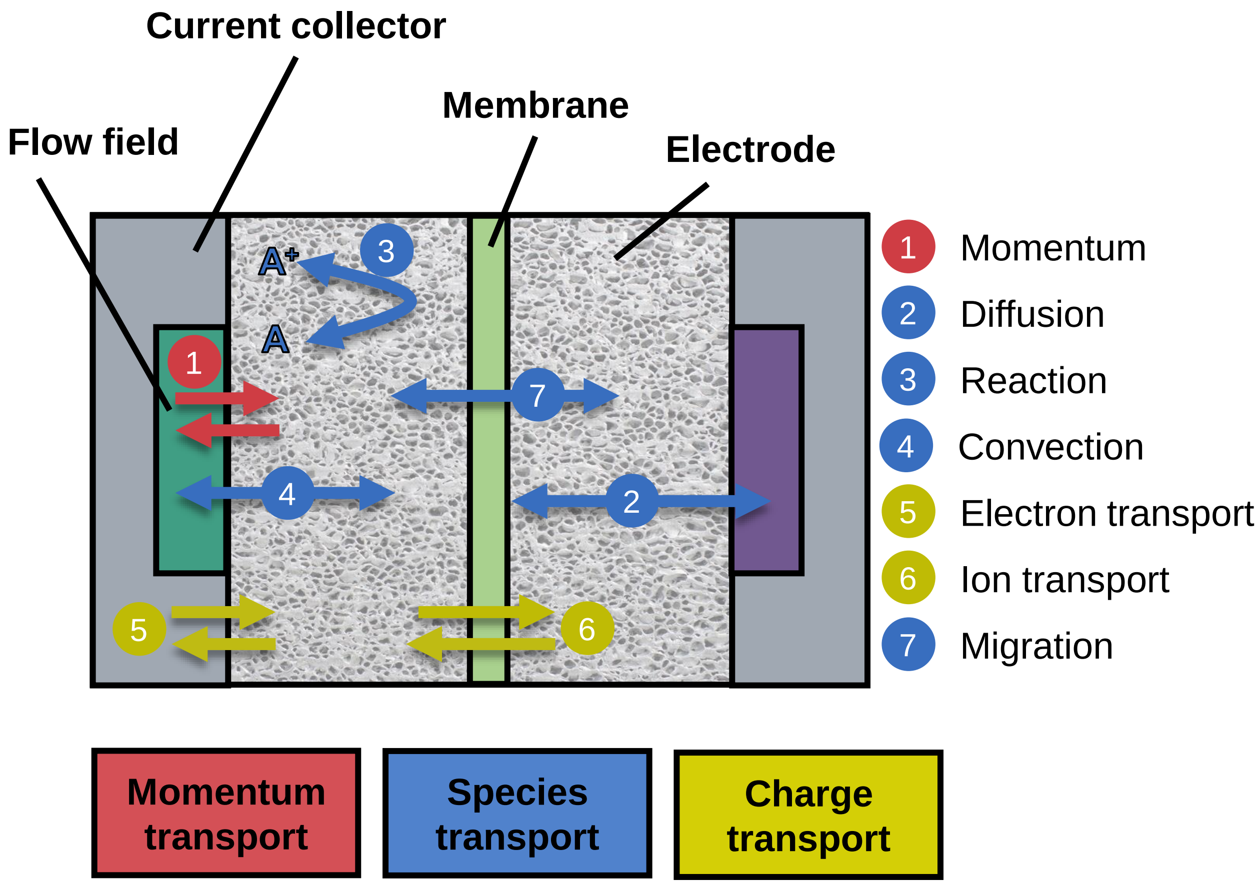
$$\begin{cases} \rho \dfrac{\partial \mathrm{u}}{\partial t} + \rho(\mathrm{u} \cdot \nabla \mathrm{u}) - \mu \nabla^2 \mathrm{u} + \nabla p = 0 \\ \nabla \cdot \mathrm{u} = 0 \end{cases}$$

$$\frac{\partial C_i}{\partial t} = \nabla.(D_i^e \nabla C_i) \; + R(C_i) \; - \nabla.(\mathrm{u}C_i) \; - \nabla.\left(\frac{zF}{RT} C_i \nabla \phi\right)$$

$$\nabla.(\sigma \nabla \phi_s) = -\nabla.(\kappa \nabla \phi_l) = a i_n(\phi_s, \phi_l)$$

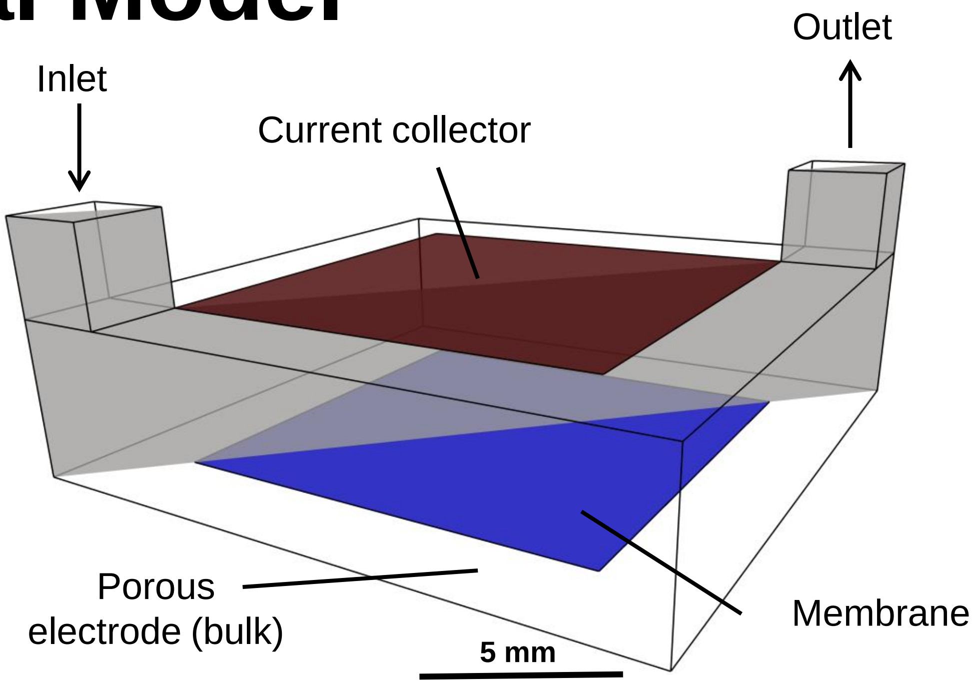
$$i_n(\phi_s, \phi_l) = \frac{i_0}{C_{\mathrm{ref}}}\left[C_R \exp\left(\frac{\alpha_A F}{RT}\Delta\phi\right) - C_O \exp\left(\frac{-\alpha_C F}{RT}\Delta\phi\right)\right]$$

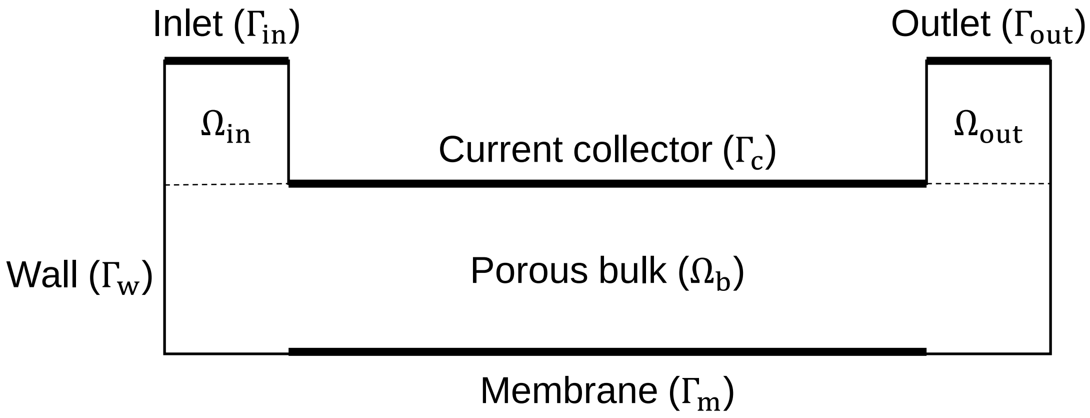
$$\Delta\phi = \phi_s - \phi_l - U_0$$



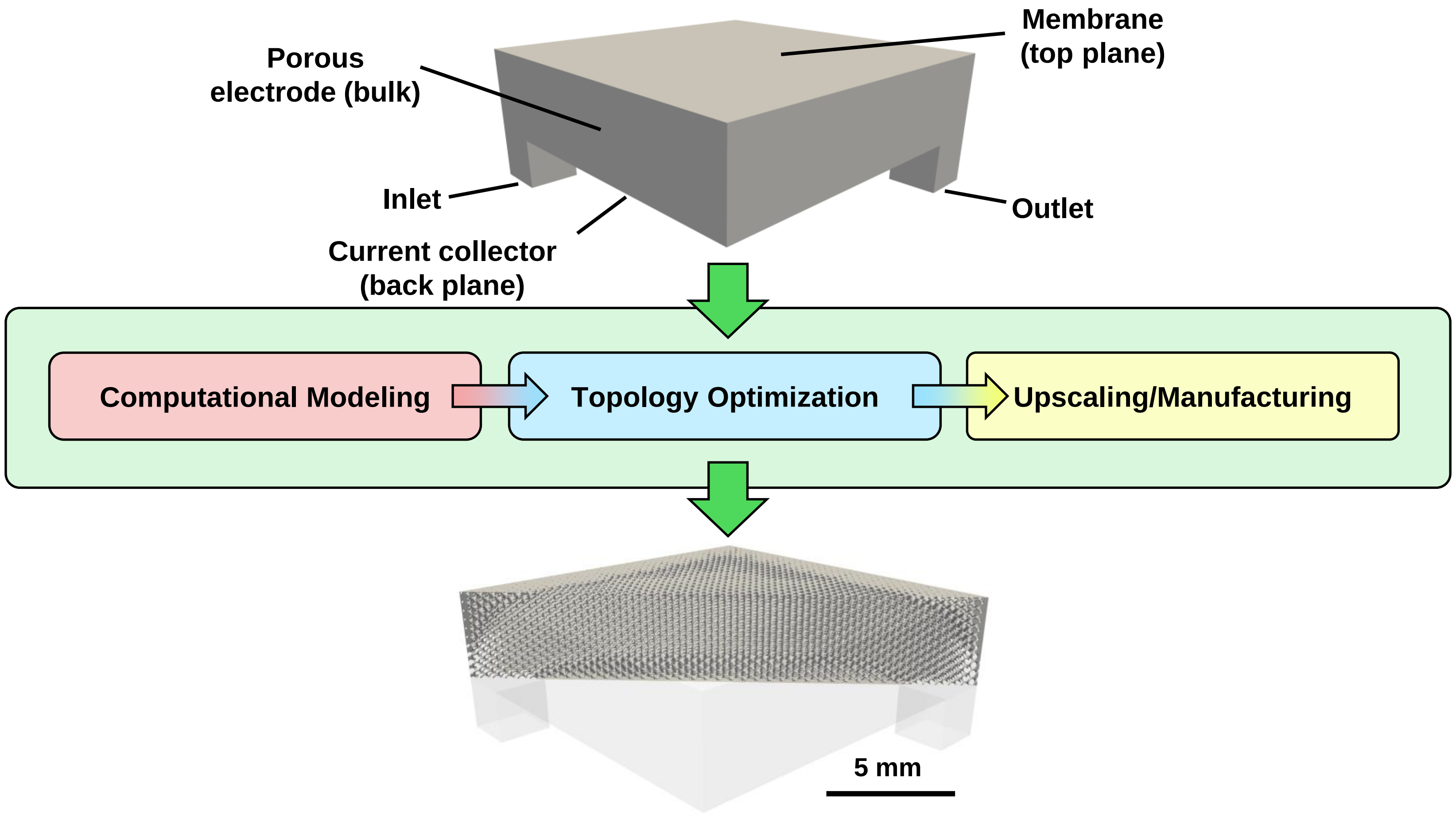
Constructing Computational Model

- Half cell
- 2D and 3D geometries
- Finite element formulation
- High-performance computing





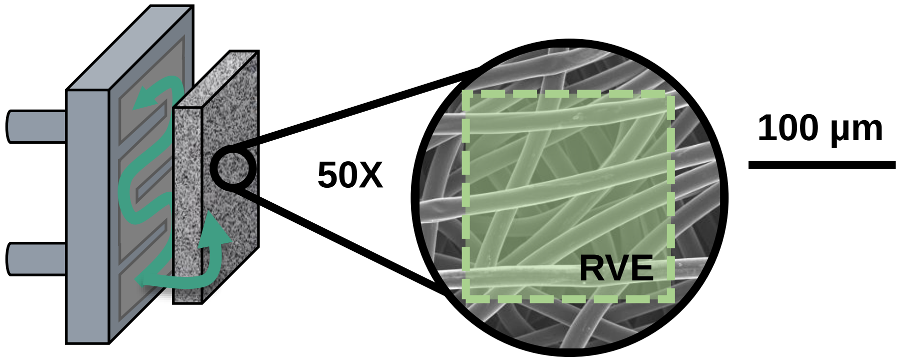
Modeling Workflow



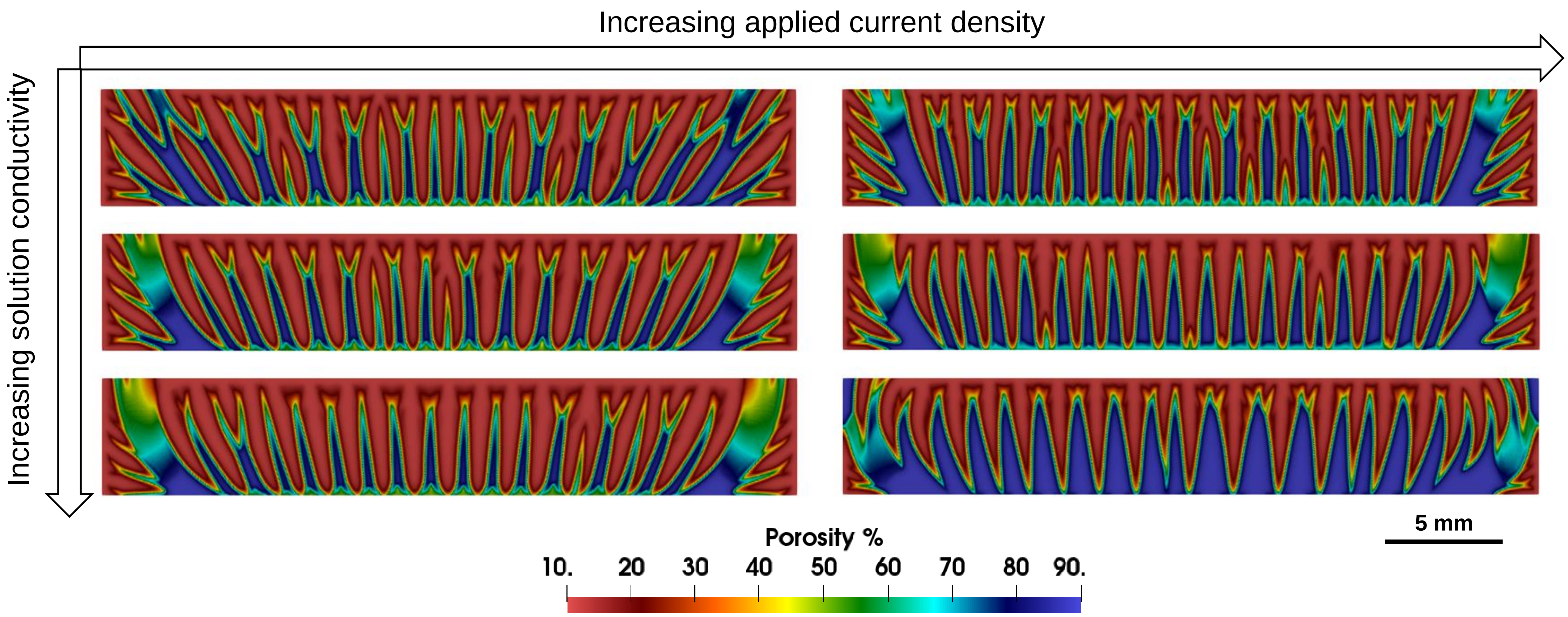
Topology Optimization of Porous Electrodes

Optimization Model

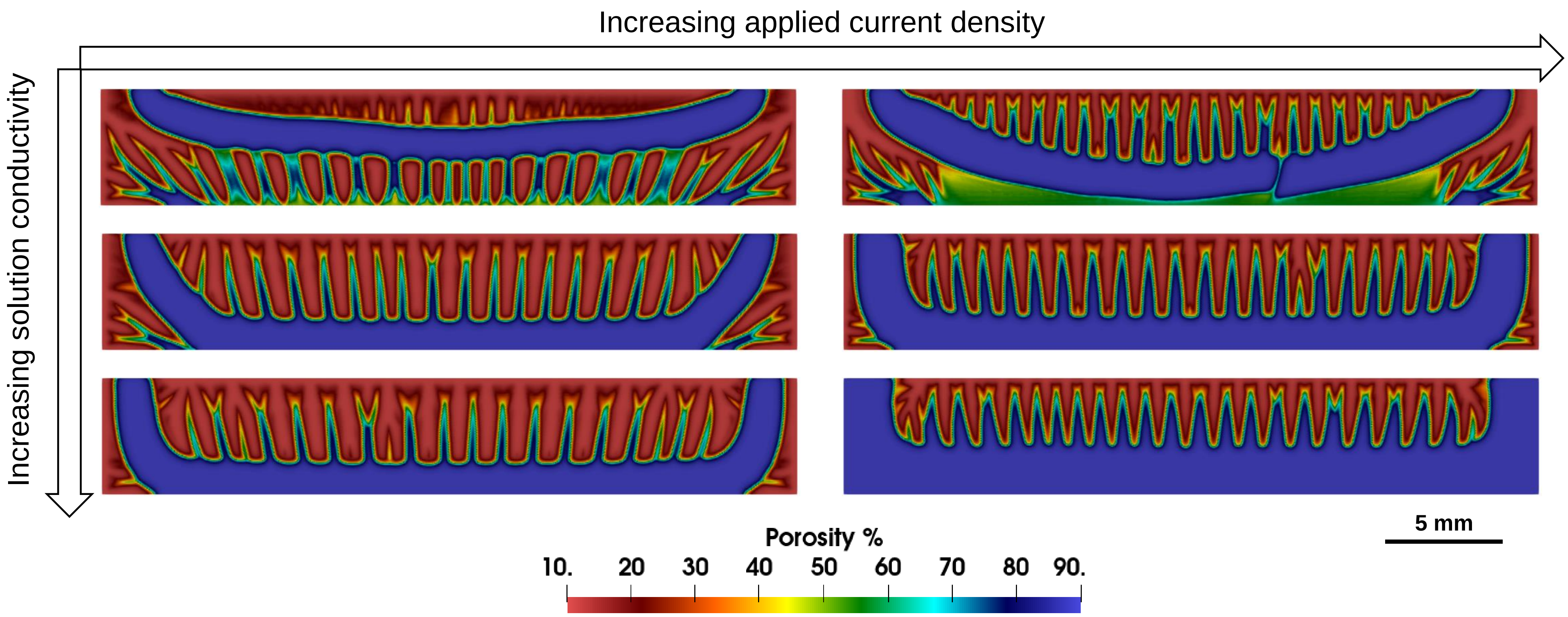
- Electrode as micro-porous material
- Skipping mass transfer effect
- Method of moving asymptotes
- Objective functions (normalized):
    - Power dissipation
    - Charge transferred on membrane



2D Results without Fluid Flow

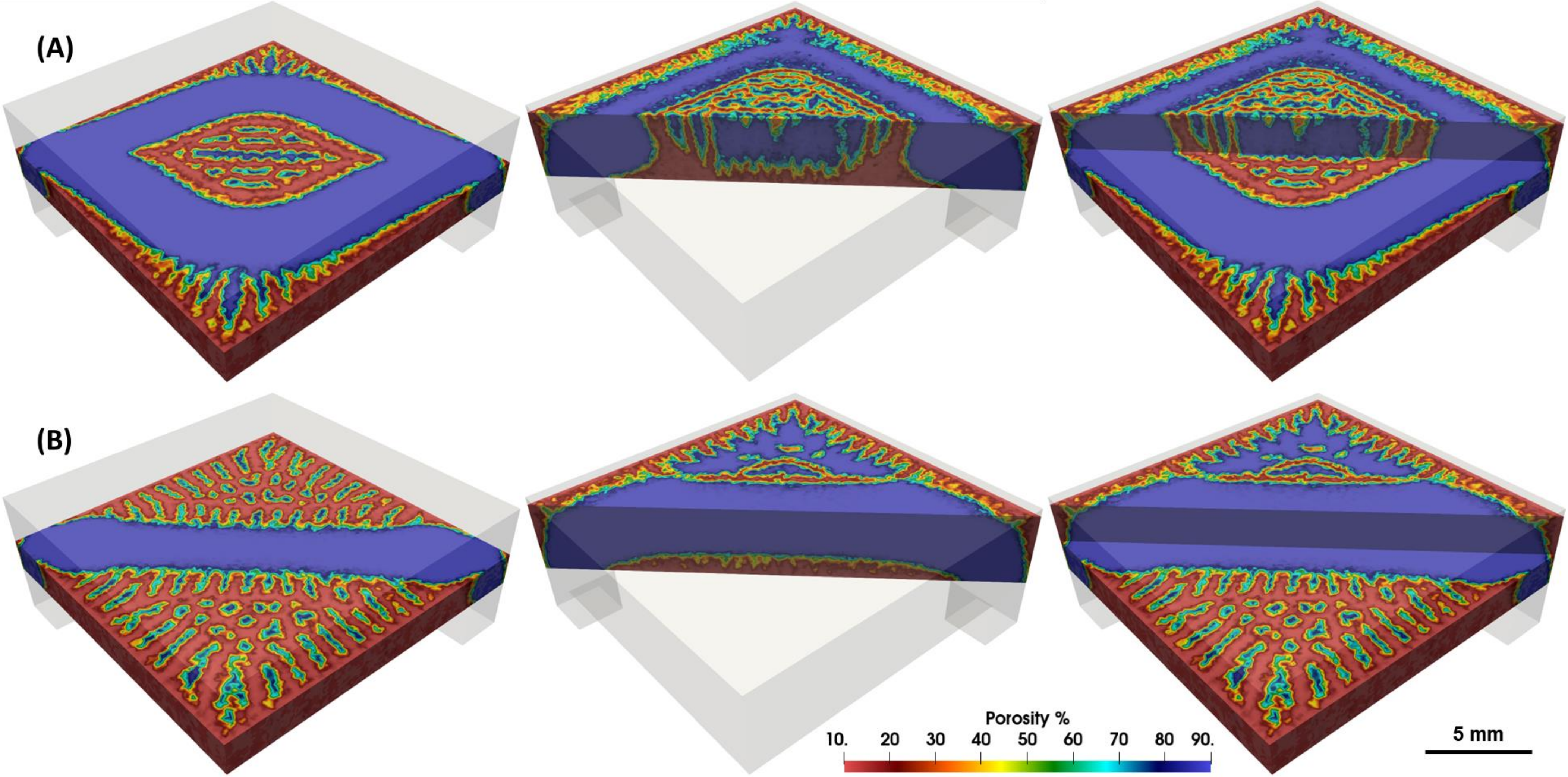


2D Results with Fluid Flow

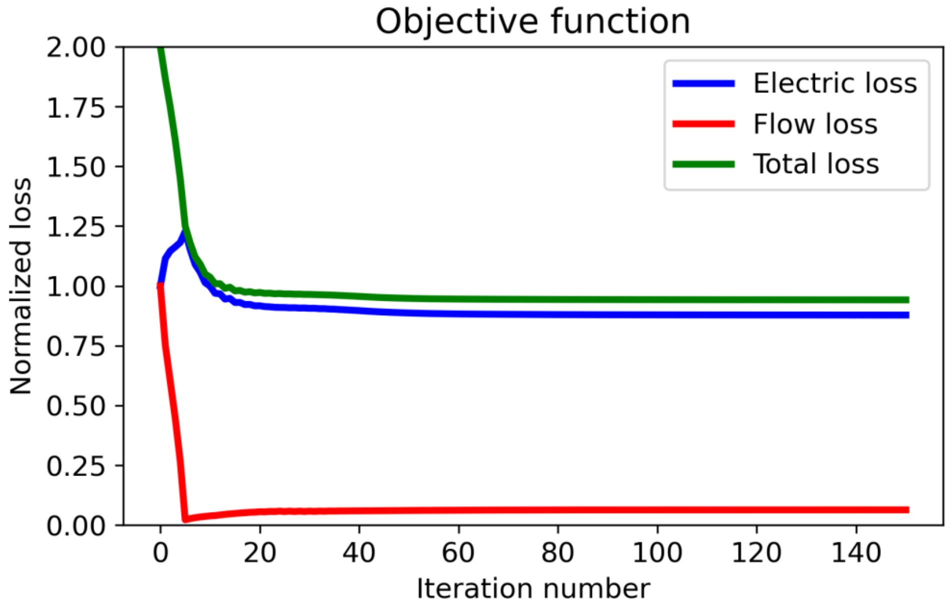


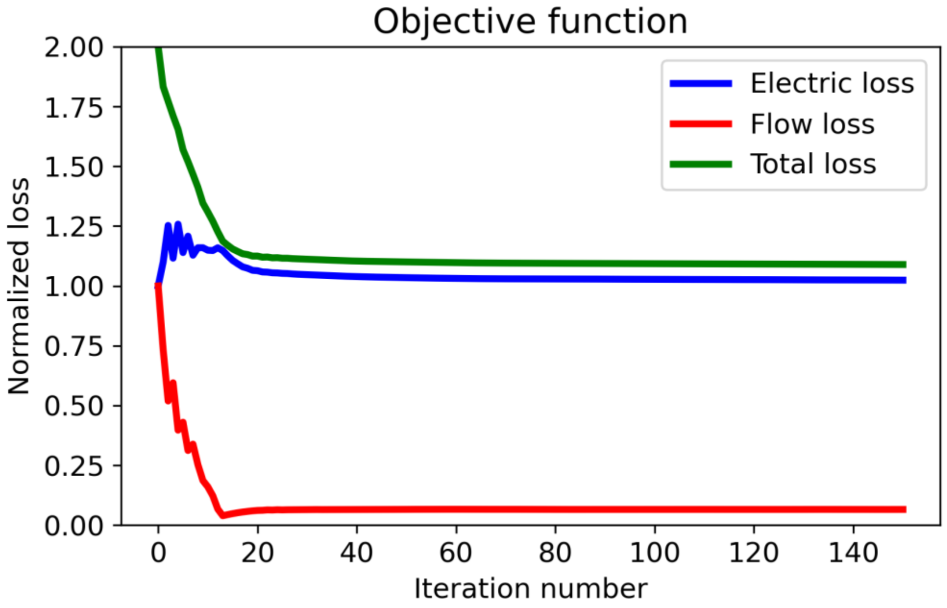
3D Results

Increased solution conductivity,
decreased applied current density

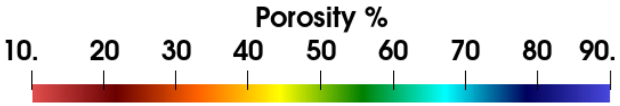


Convergence History



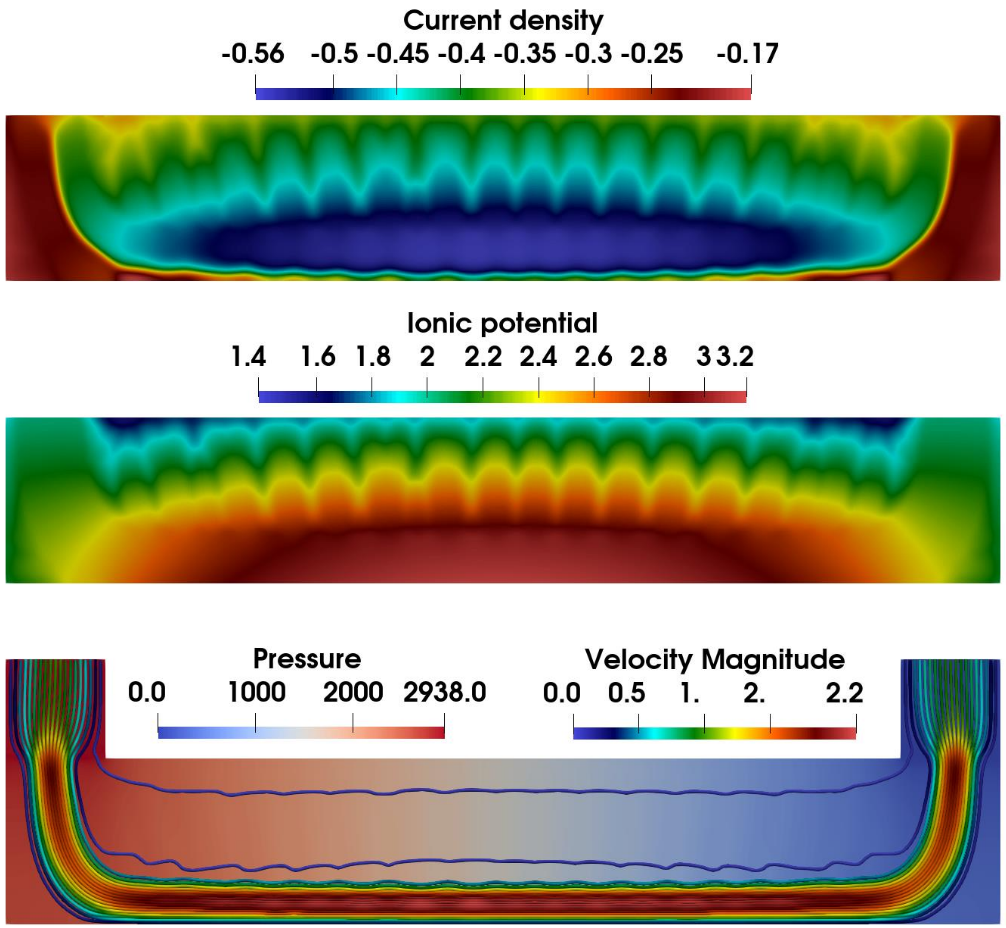


(High conductivity)

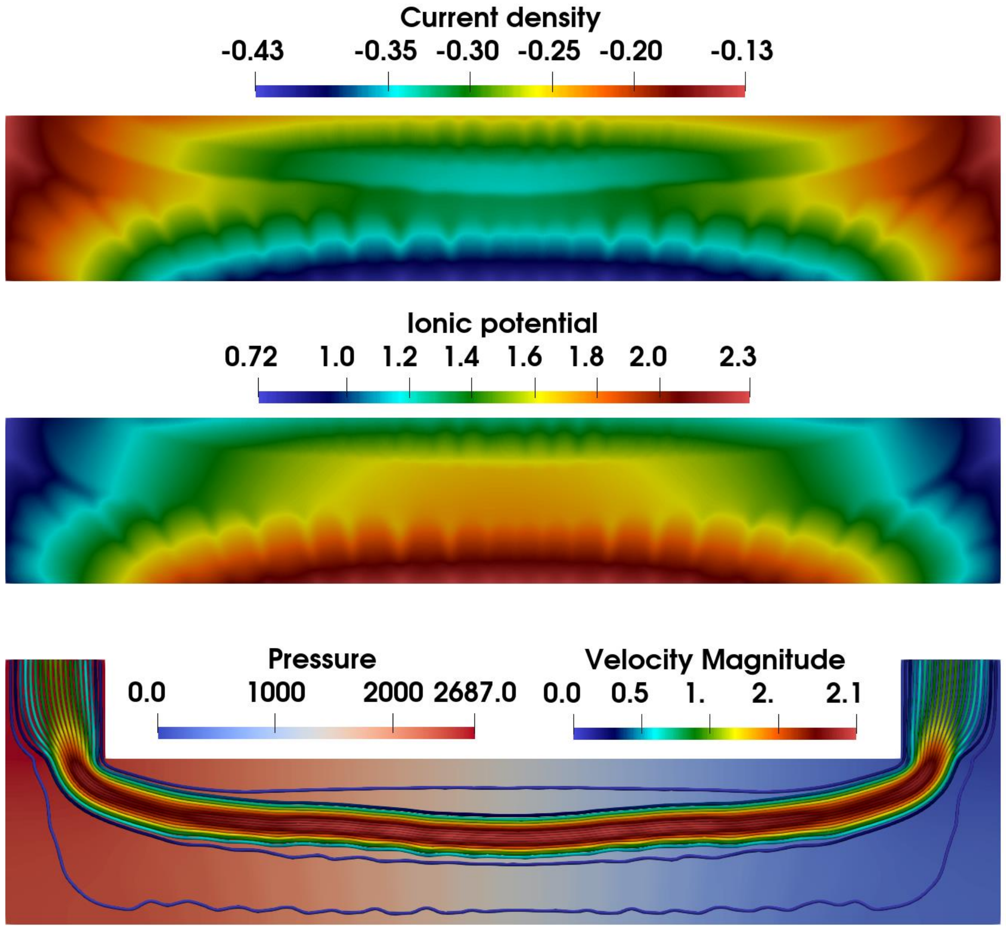


(Low conductivity)

Other Simulated Quantities

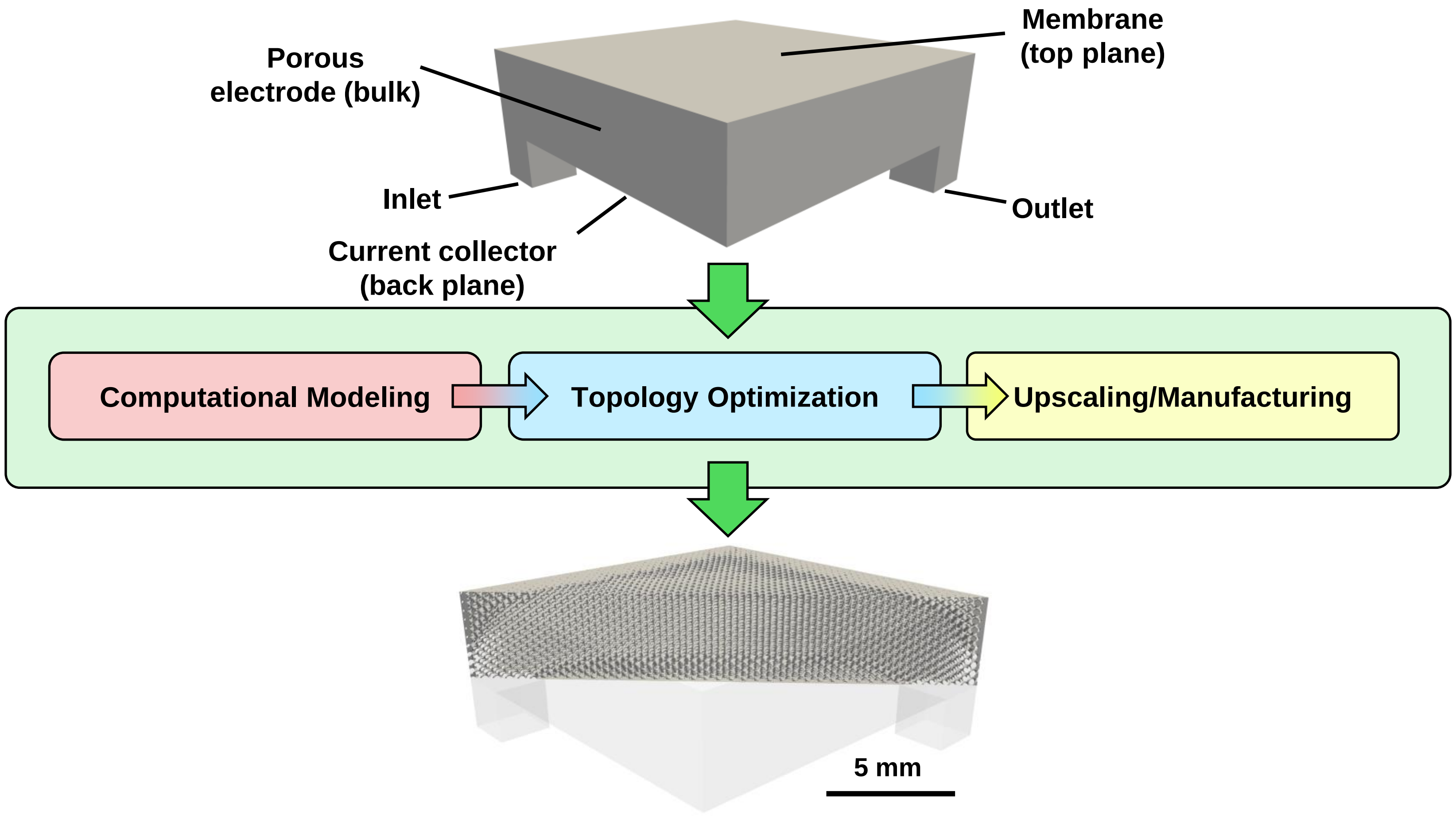


(High conductivity)



(Low conductivity)

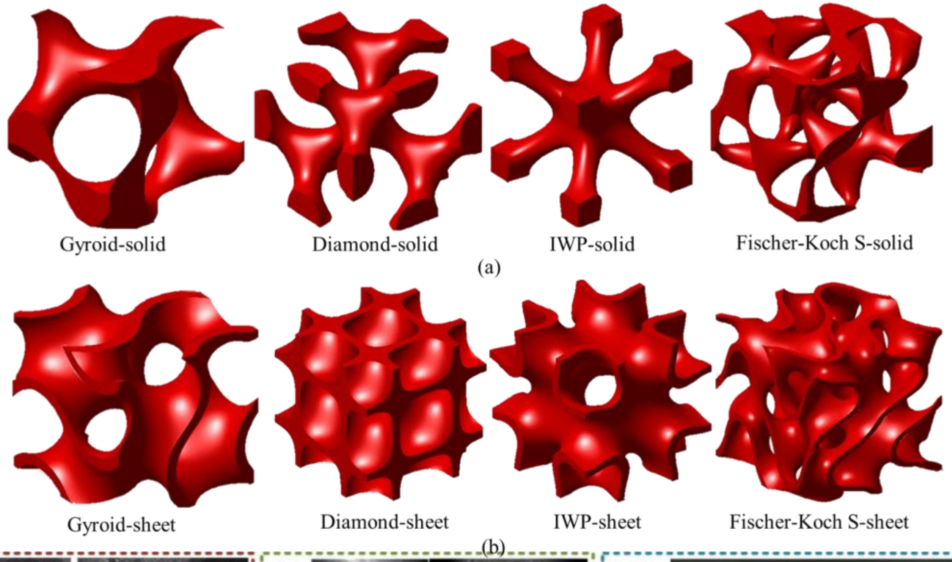
Modeling Workflow

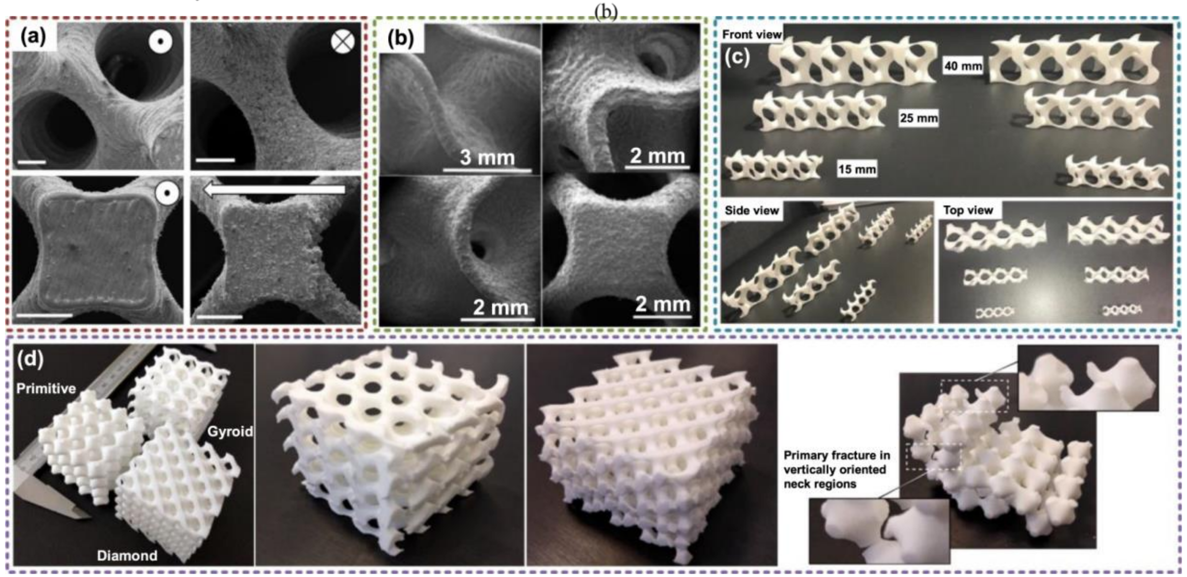


Upscaling using Periodic Surfaces

Triply Periodic Minimal Surfaces (TPMS)

- Smooth surfaces
- Highly interconnected porous architectures
- Mathematical controllable geometry features
- Excellent transport properties

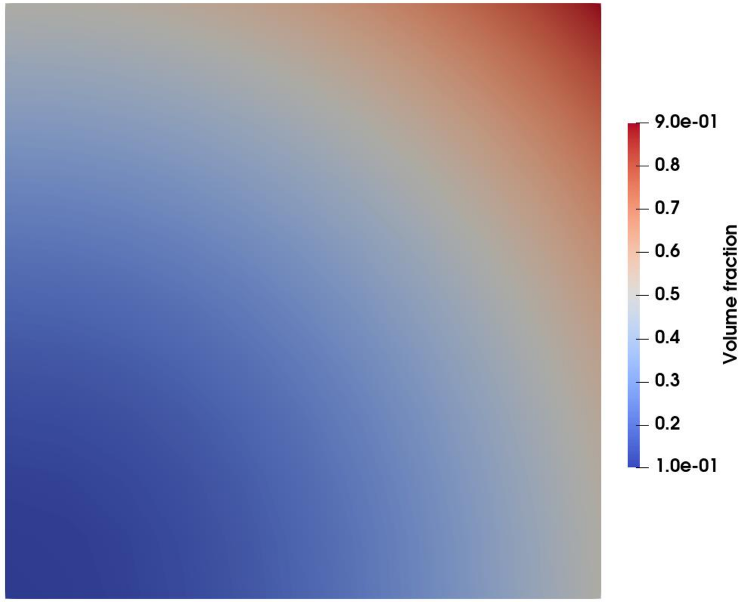




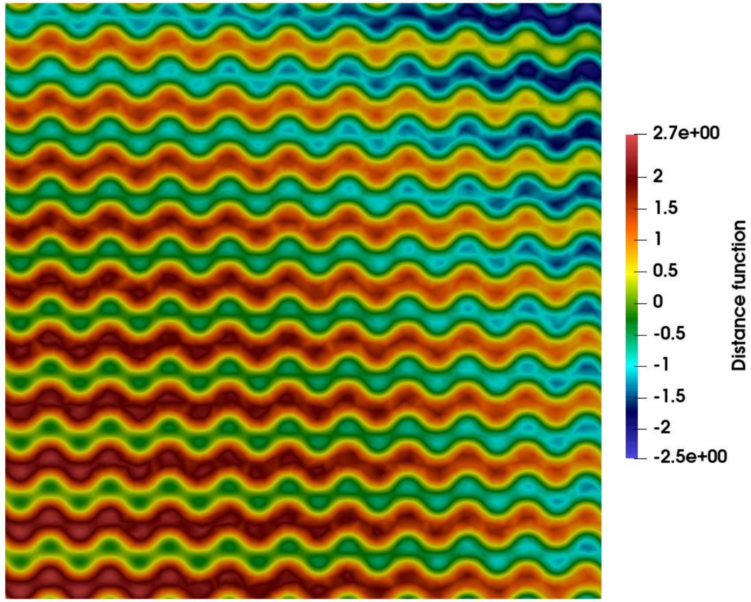
Feng et al., Int. J. Extreme Manuf., 4 (2022) 022001. Yeranee & Rao, Energies, 15 (2022) 8994.

Transforming Optimization Results

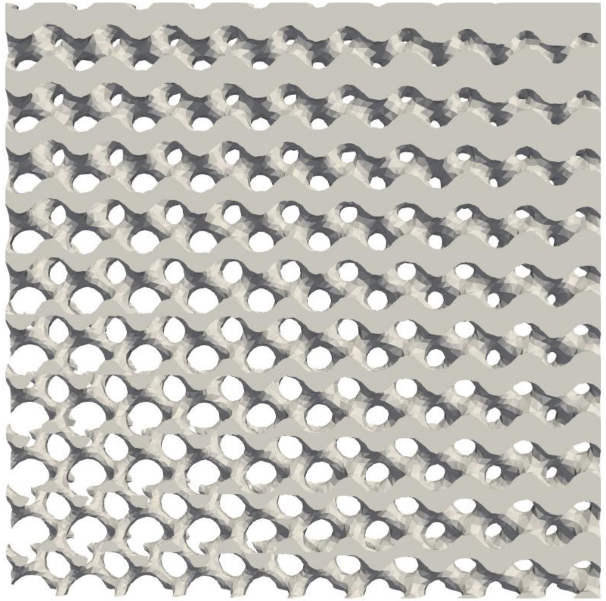
- Converting variable porosity to TPMS infills



(Volume fraction)

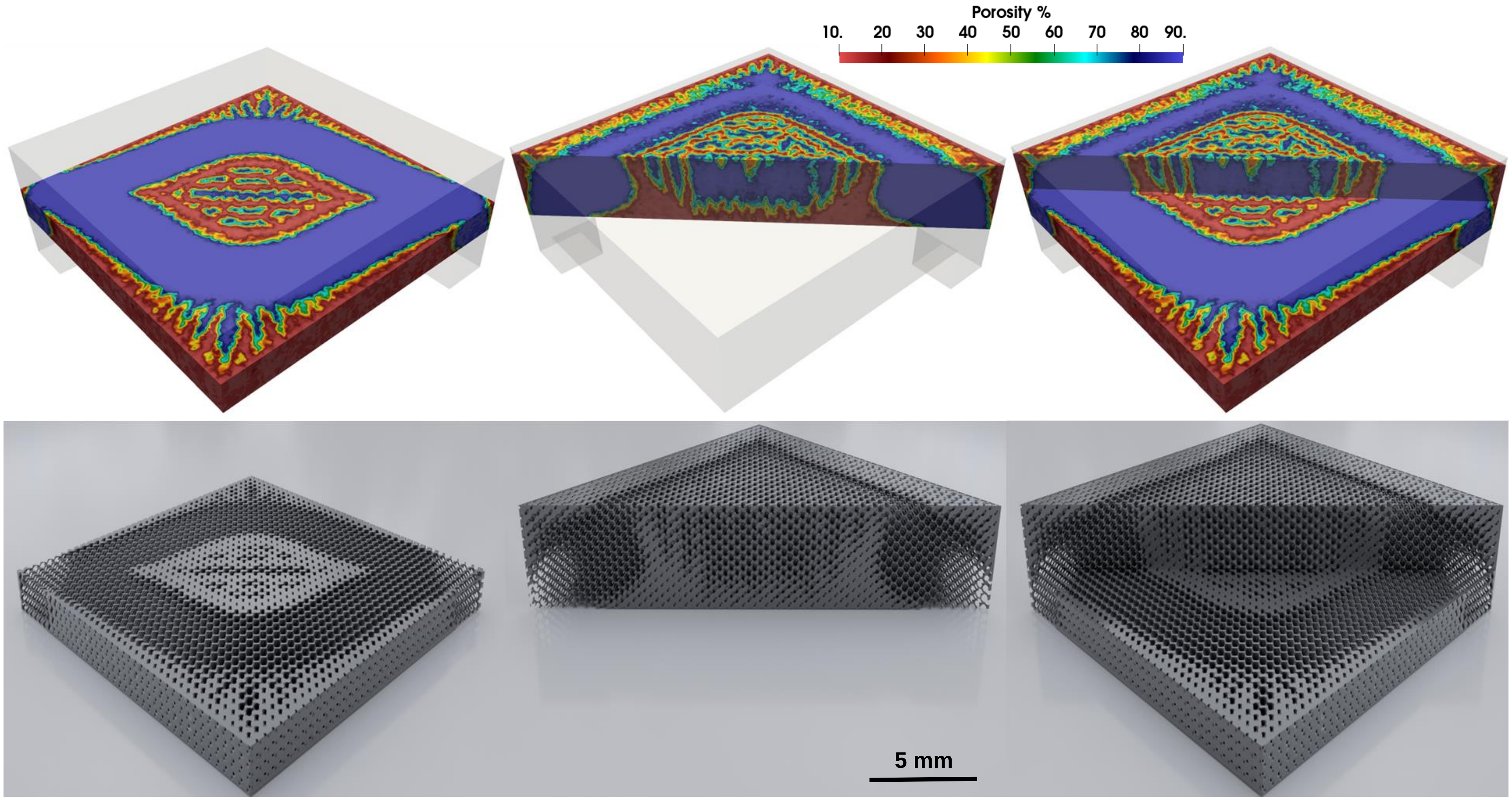


(Distance function)

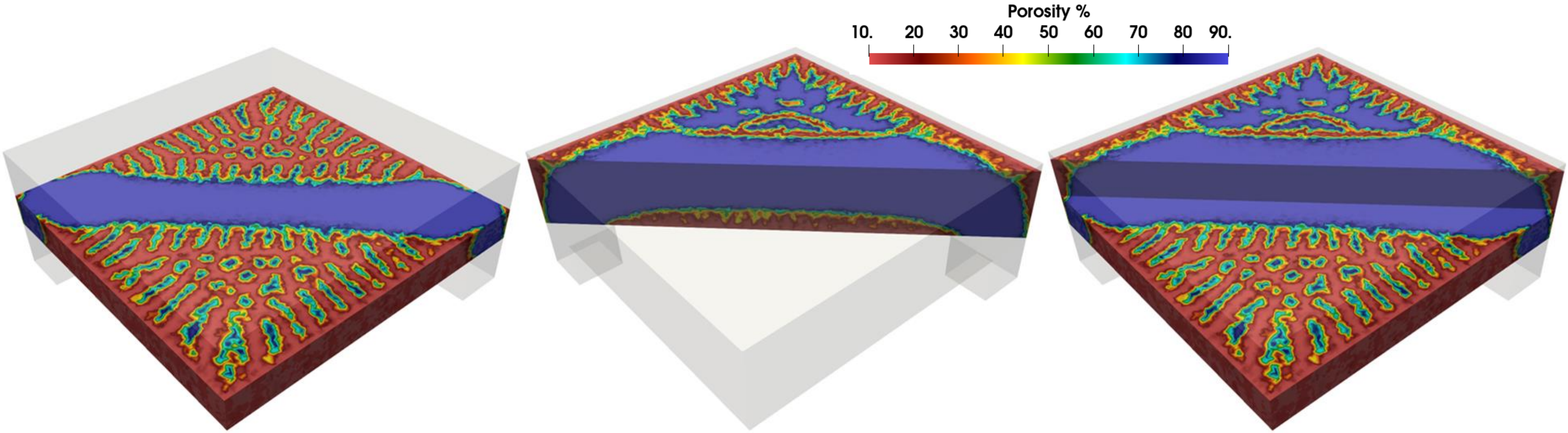


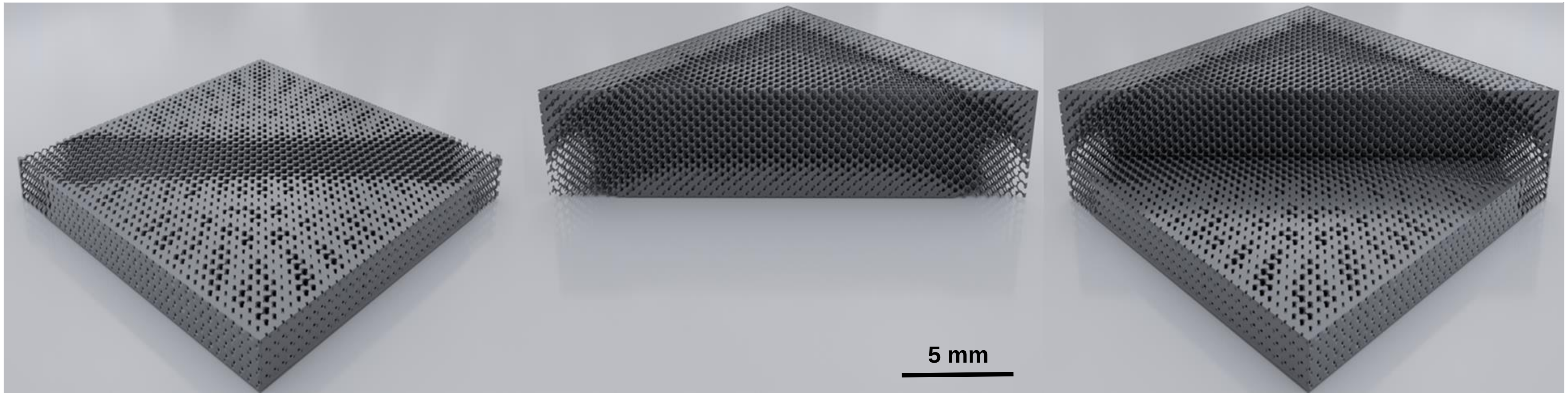
(TPMS infill)

Conversion Results #1 (conductivity ↓, current density ↑)



Conversion Results #2 (conductivity ↑, current density ↓)



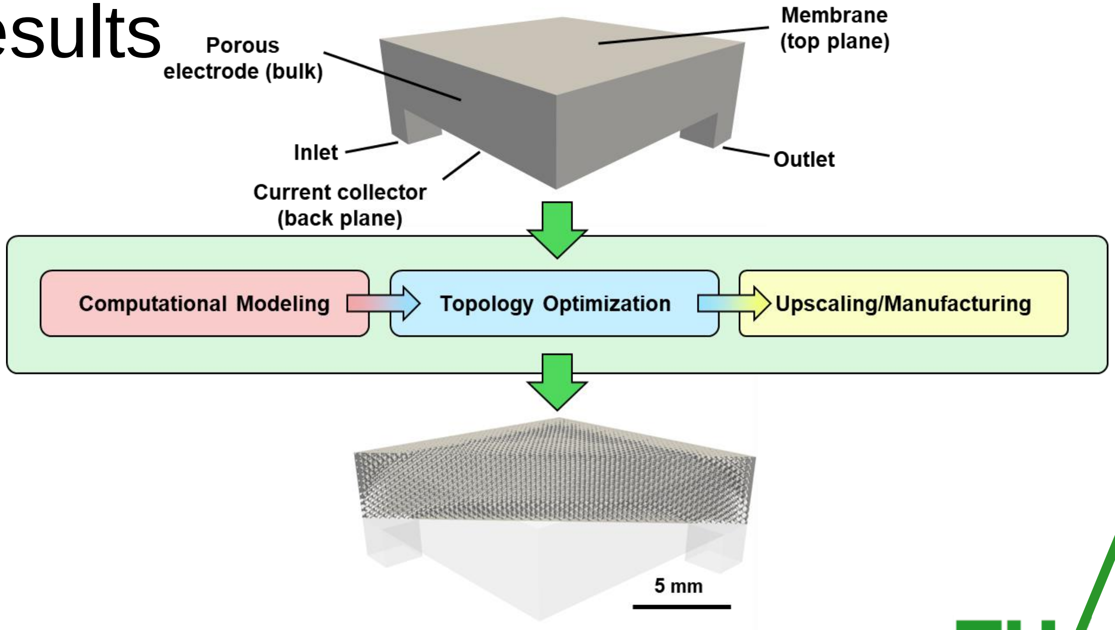


Employed Tools are all Open-Source!

- Mesh generation
  - GMSH
  - SALOME
- Multiphysics modeling
  - Firedrake
  - PETSc
- Topology optimization
  - pyMMAopt
  - PETSc
- TPMS generation
  - ASLI
  - CGAL
  - FreeFEM
  - MeshLab
- Visualization
  - Blender
  - ParaView

Conclusion

- Numerical models for correlating local configuration/structure to overall redox cell performance
- Scalable topology optimization for engineering porous electrodes
- Manufacturability by transforming results to TPMS infills



Thank You for Your Attention!

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