Inverse Design of Porous Electrodes in Redox Flow Batteries: A Computational Approach Integrating Topology Optimization and Multi-Physics Modeling

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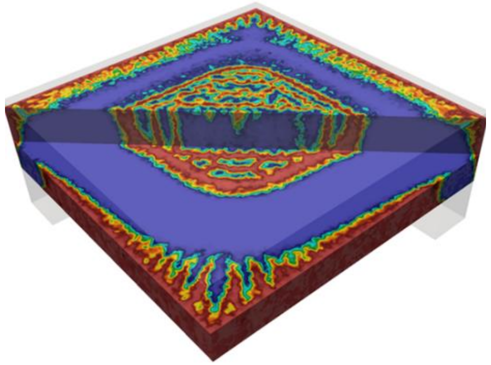
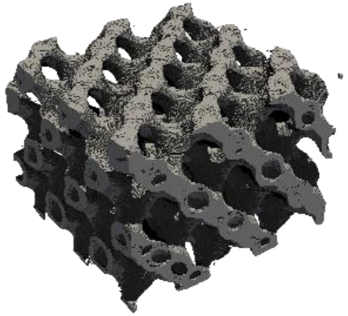
Maxime van der Heijden, Antoni Forner-Cuenca

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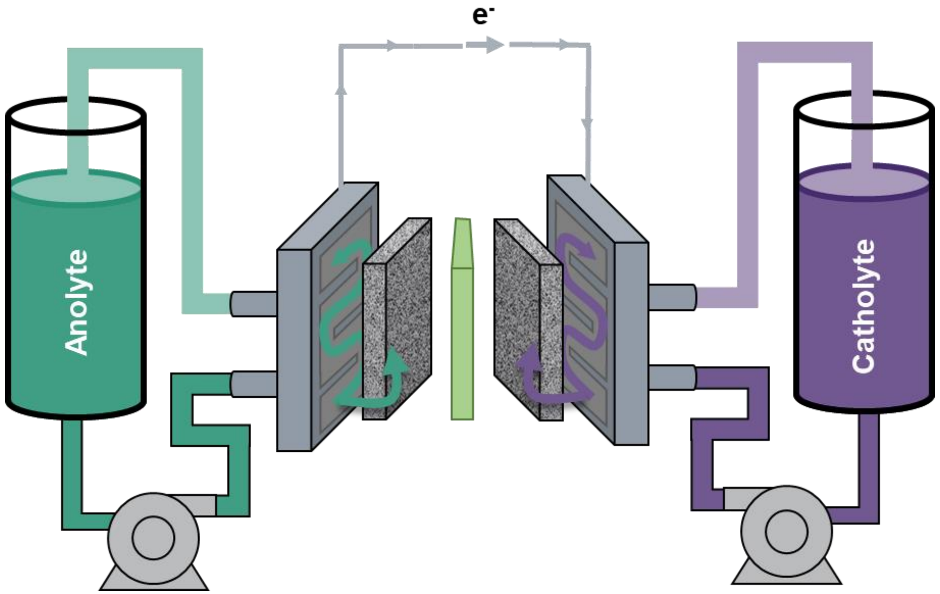
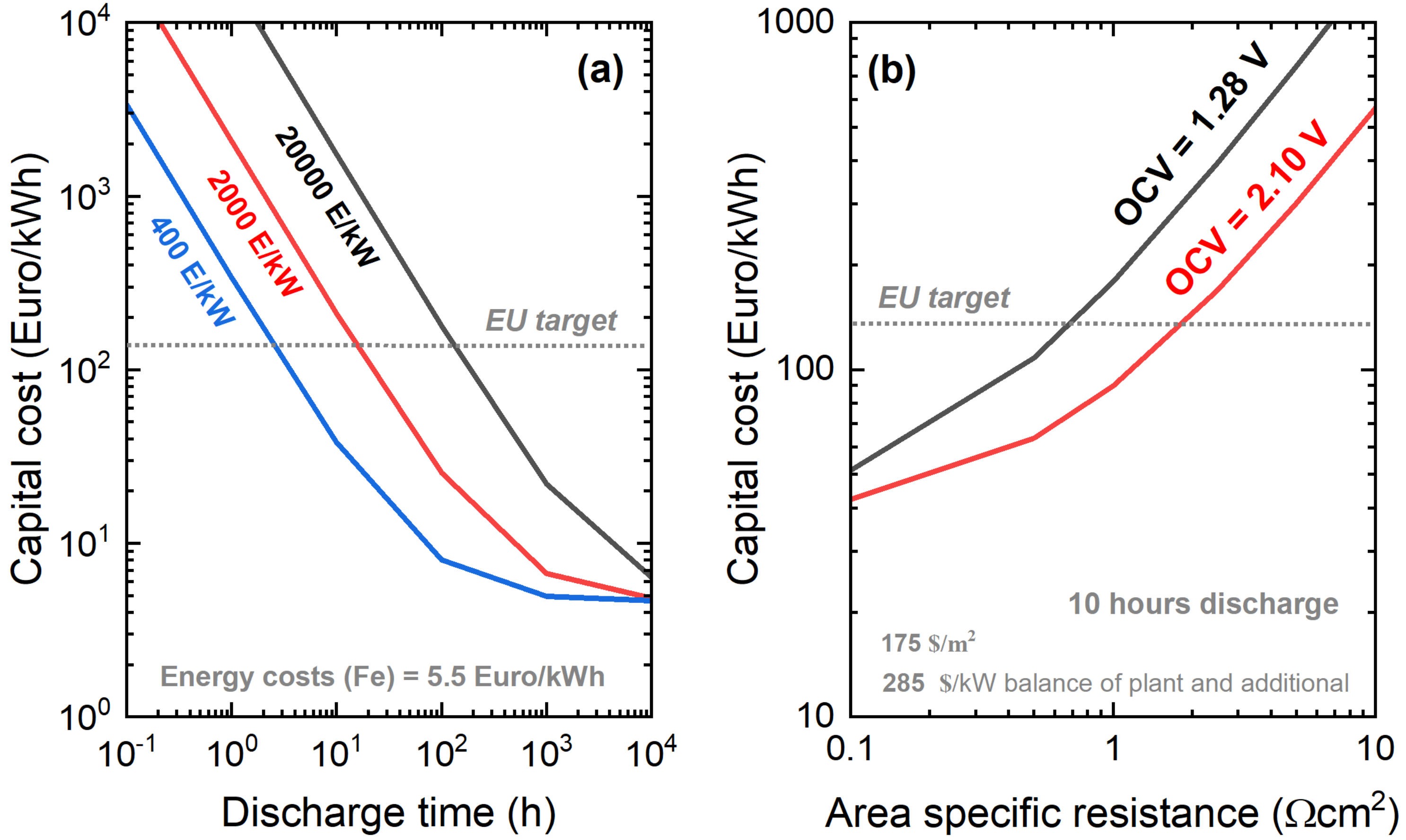
Honolulu, 6th October 2024

How does increasing power density and reducing materials costs affect the economics?



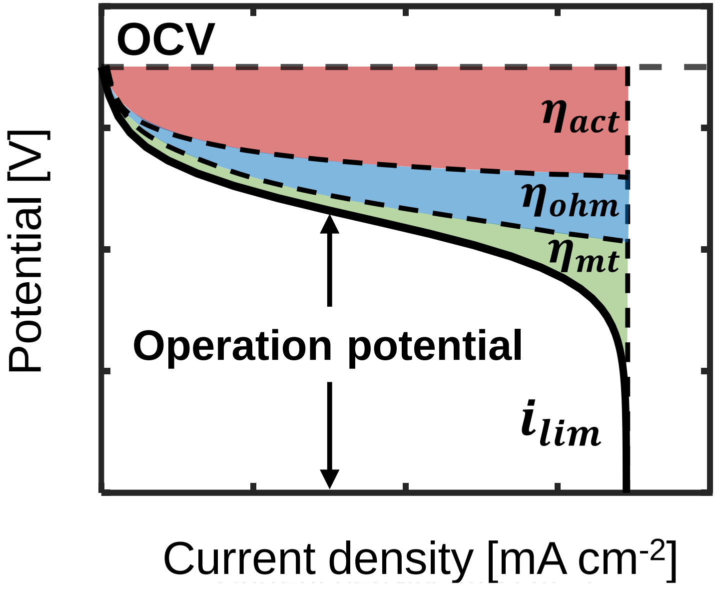


Minimizing area-specific resistance is a powerful strategy for reducing reactor cost contributions to the total system cost

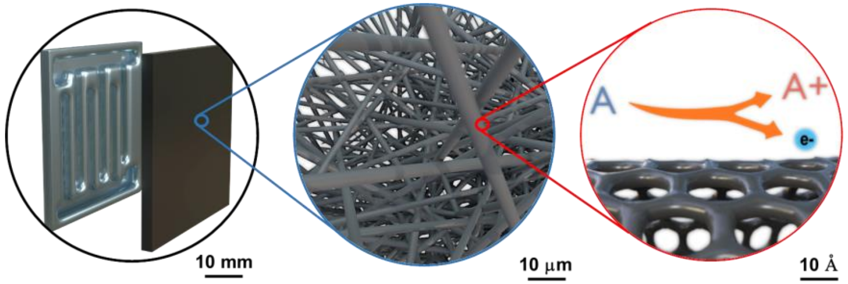
References to TE analysis: Darling et al., *Energy Environ. Sci. 7, 3459 (2014)*, Noack et al., *Energies 9(8) 627 (2016).*

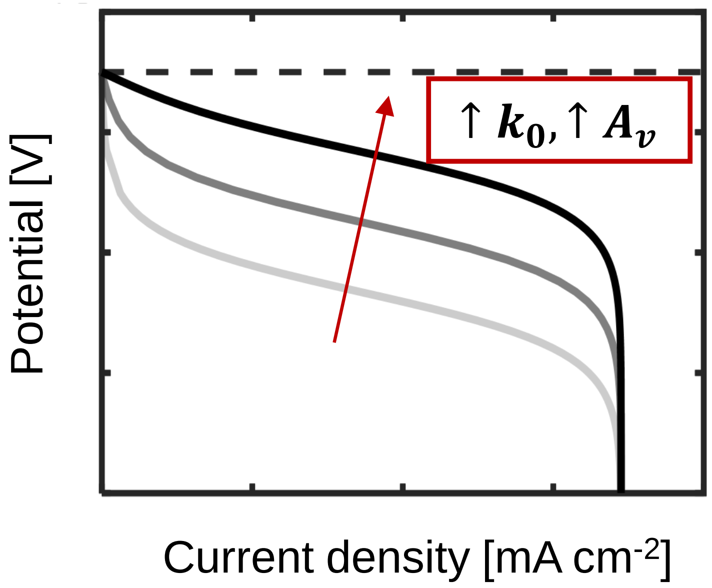
TU/e

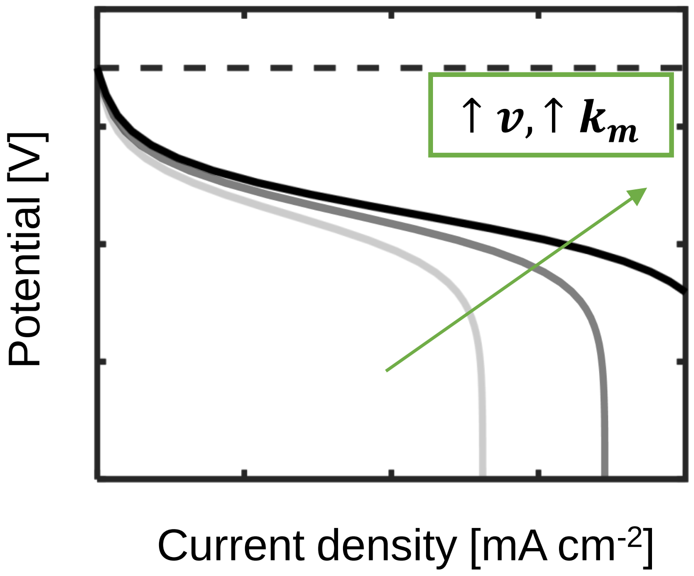
Electrode performance determines cell overpotentials



Assuming: *OCV = 1.50 V; HFR = 130 mΩ cm²* (e.g. VRFB)
What is the maximum (theoretical) power density?
(assuming an infinitely fast charge and mass transfer)
Power density = ***4.40 W cm⁻²***







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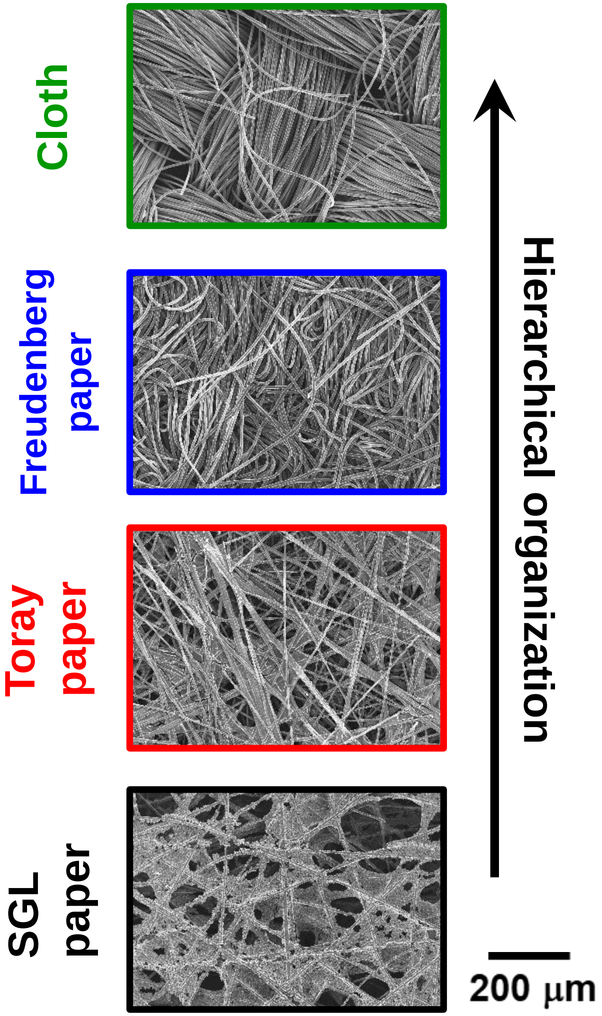
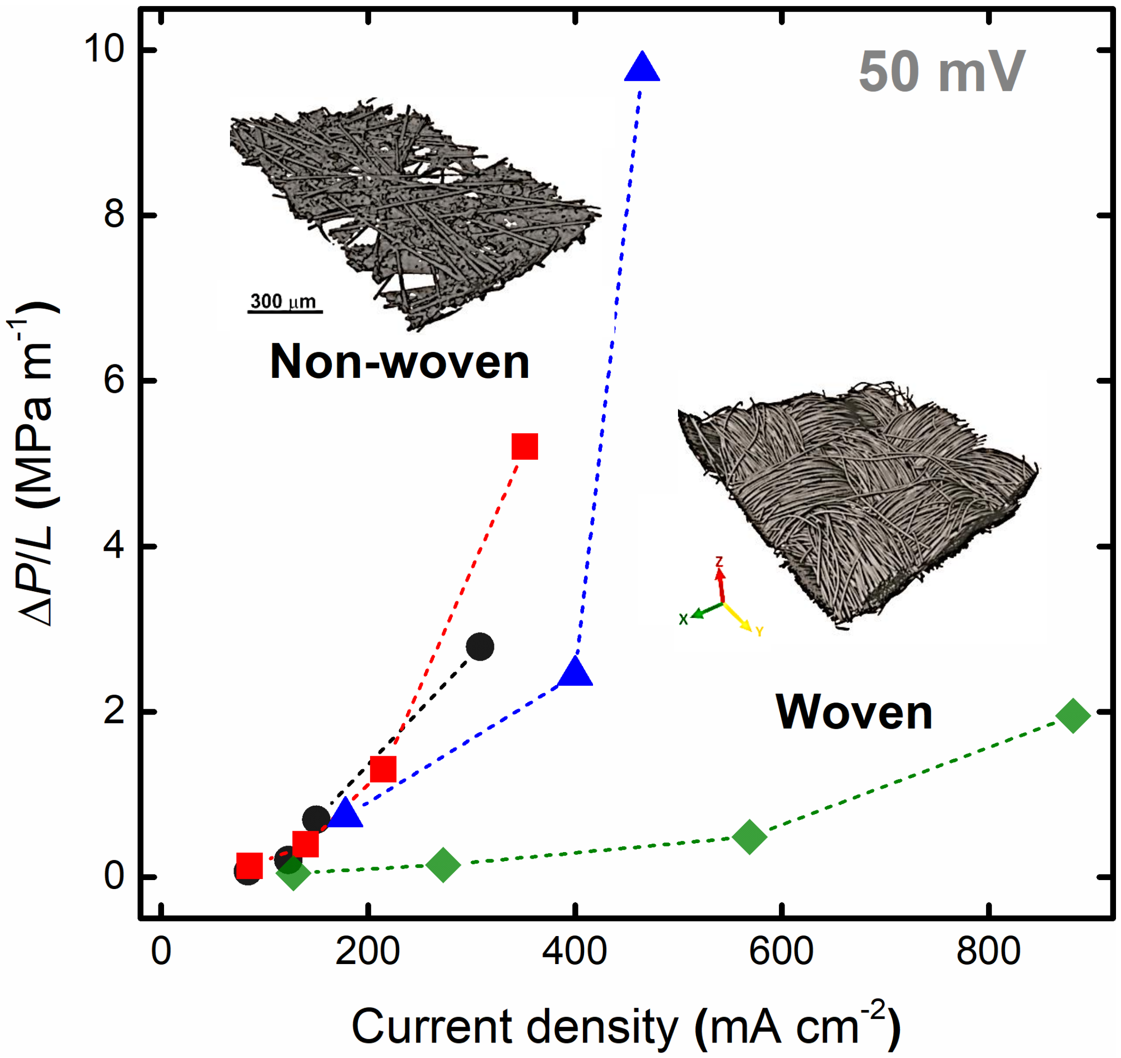
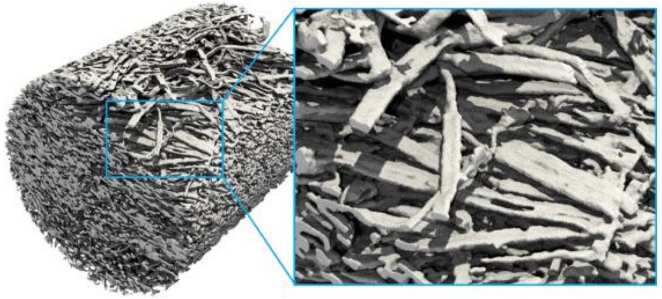
Electrode microstructure governs performance





Forner-Cuenca et al., *Current Opinion in Electrochemistry*, 18, 113 (**2019**). *Journal of The Electrochemical Society*, 166(10) A2230-A2241 (**2019**)

Engineering electrode microstructures…



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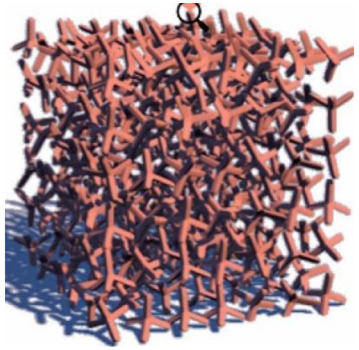

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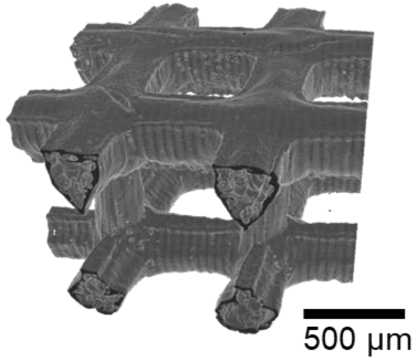

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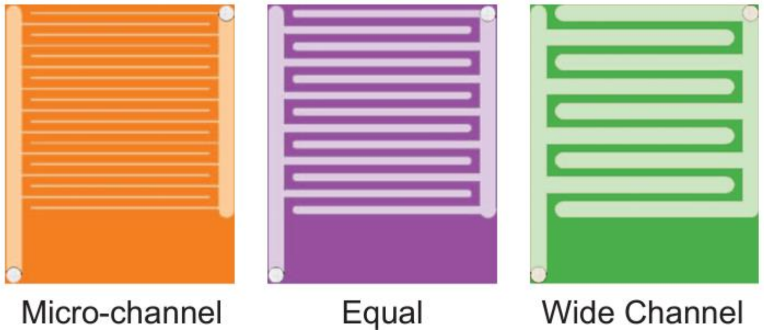
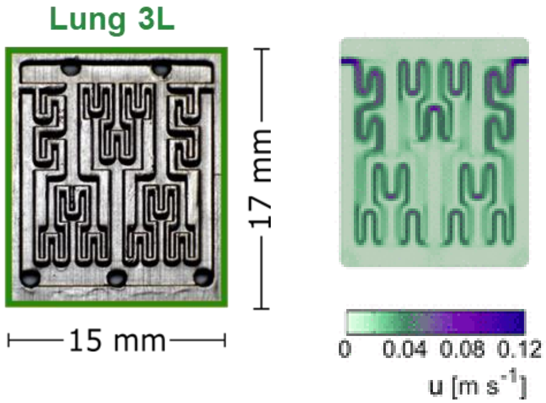
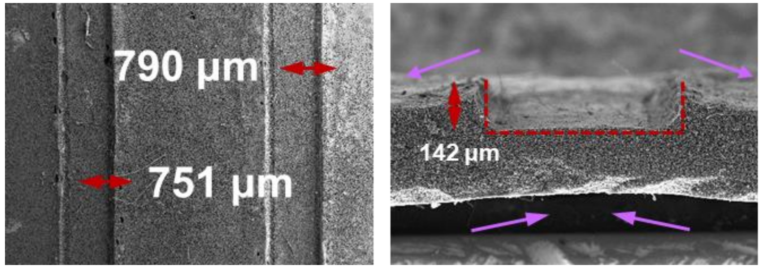
… and flow field geometries



Gerhardt, J. Electrochem. Soc, 165 (2018)



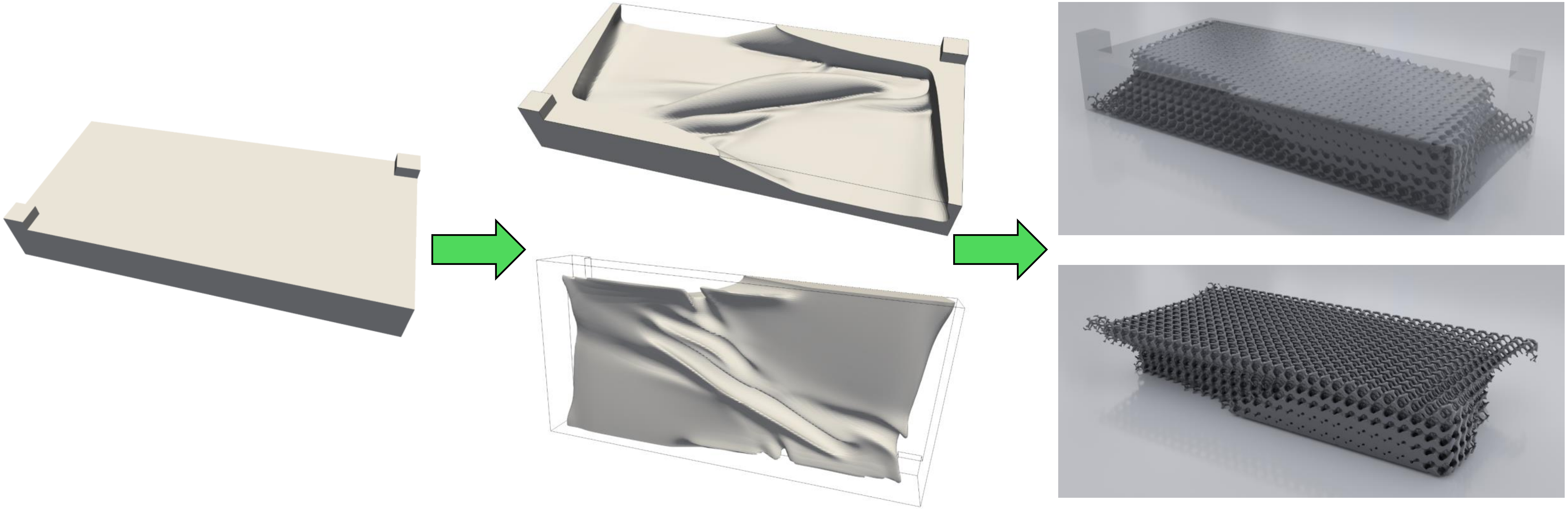
Munoz-Perales, ACS Sust. Chem. Eng., 11 (2023)



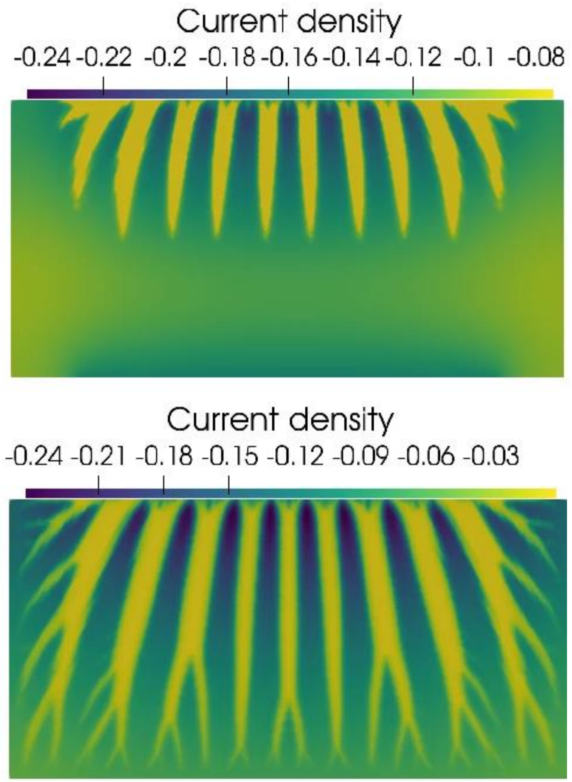
Liu et al., In preparation.

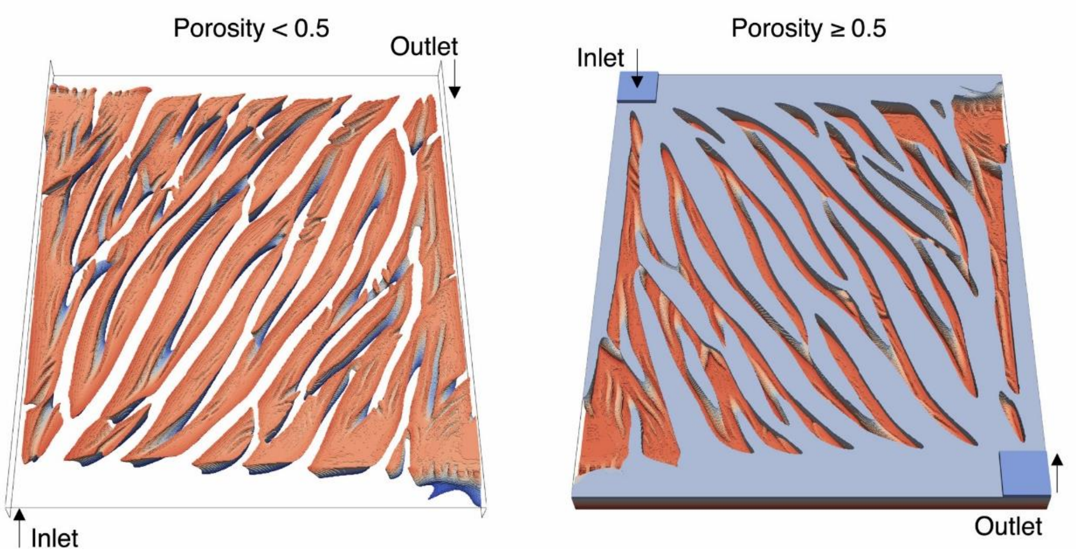
Electrode design via engineering optimization?

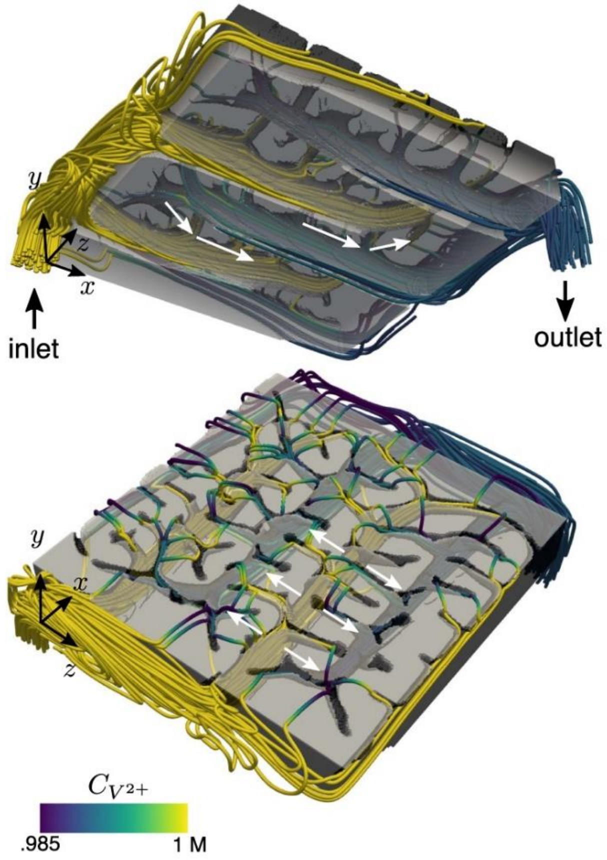
- Inverse design of electrodes for maximizing performance



Topology optimization in electrochemistry

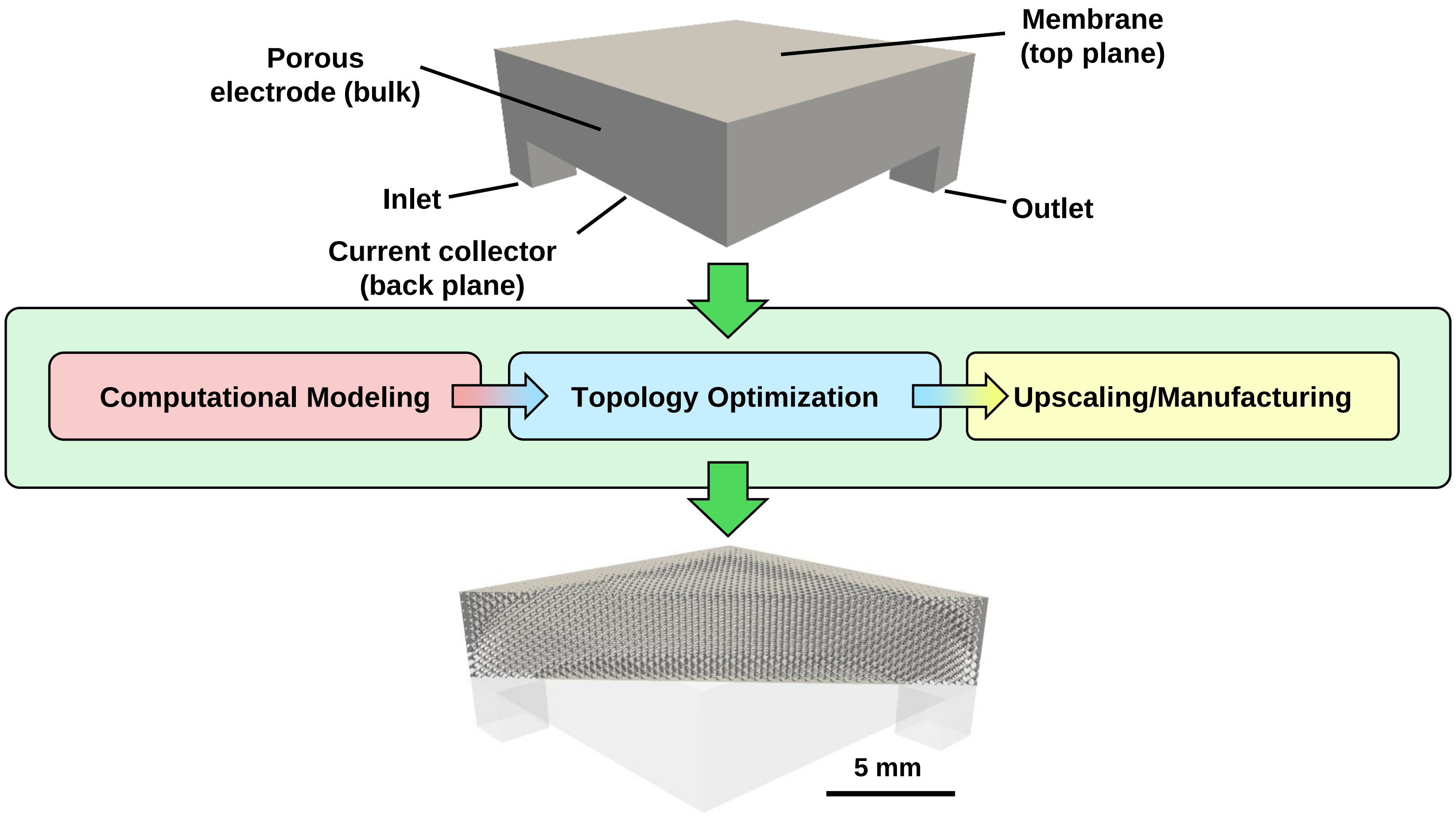






Roy et al., Struct. Multidiscipl. Optim., 65 (2022); Beck et al., J. Power Sources., 512 (2021); Tiras et al., J. Electrochem. Soc., 169 (2022)

Modeling workflow

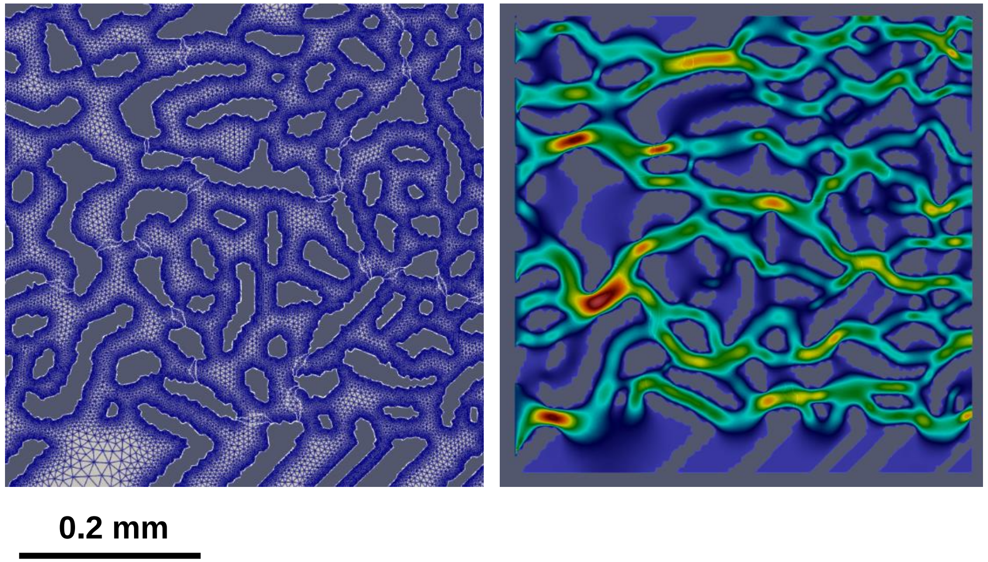


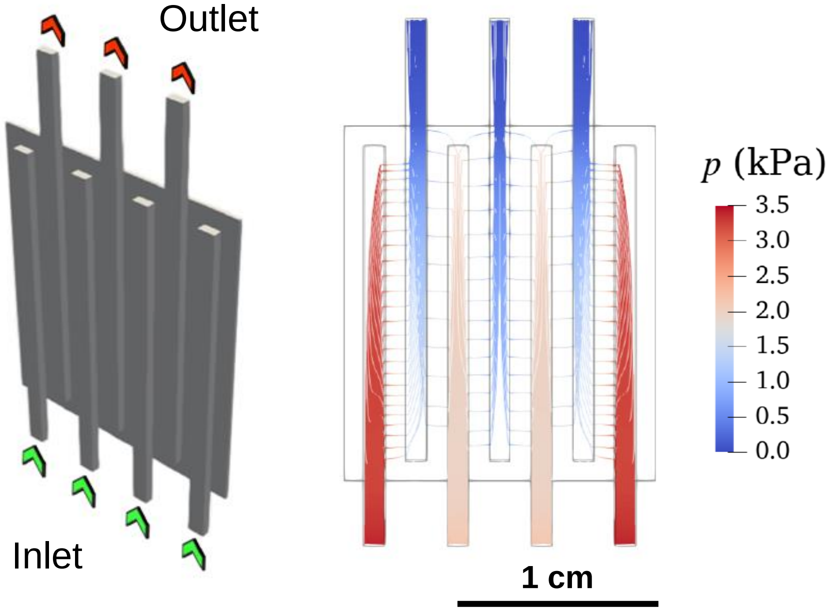
Multi-{scale, physics} modeling

Multi-scale computational modeling of RFB processes

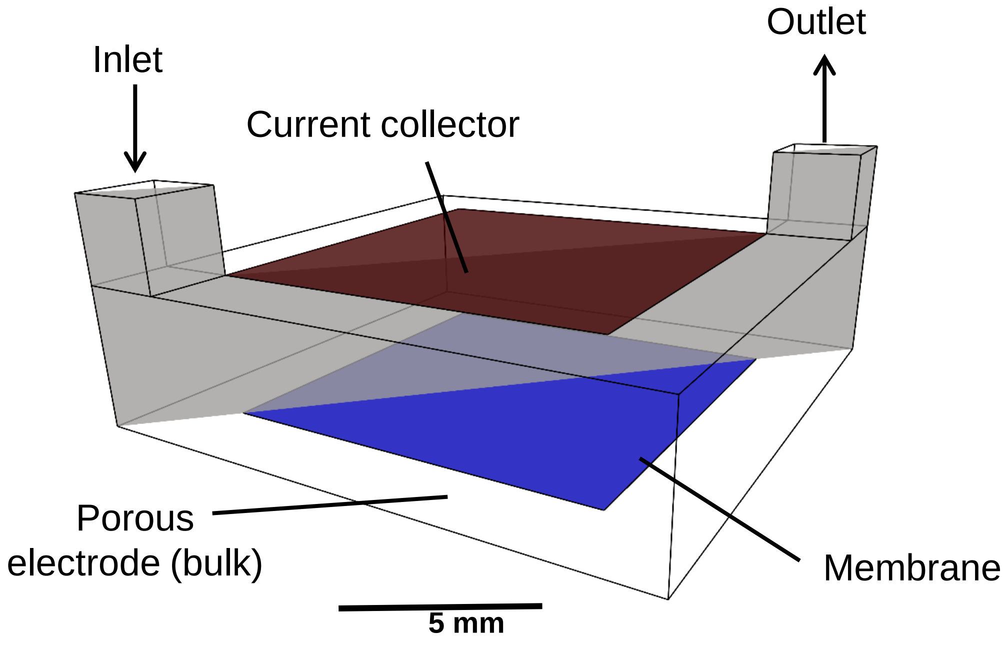
Validation, assessment and performance analysis of developed models

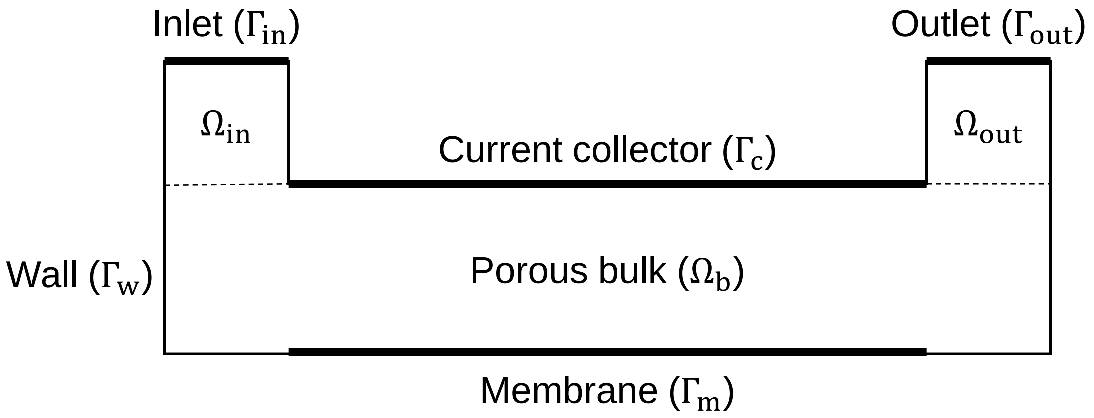
Pore-scale transport models → Cell-level models of transport phenomena





Computational model





- Half cell (symmetric)
- Finite element formulation
- Neglecting mass transfer
- Porous electrode theory

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

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

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

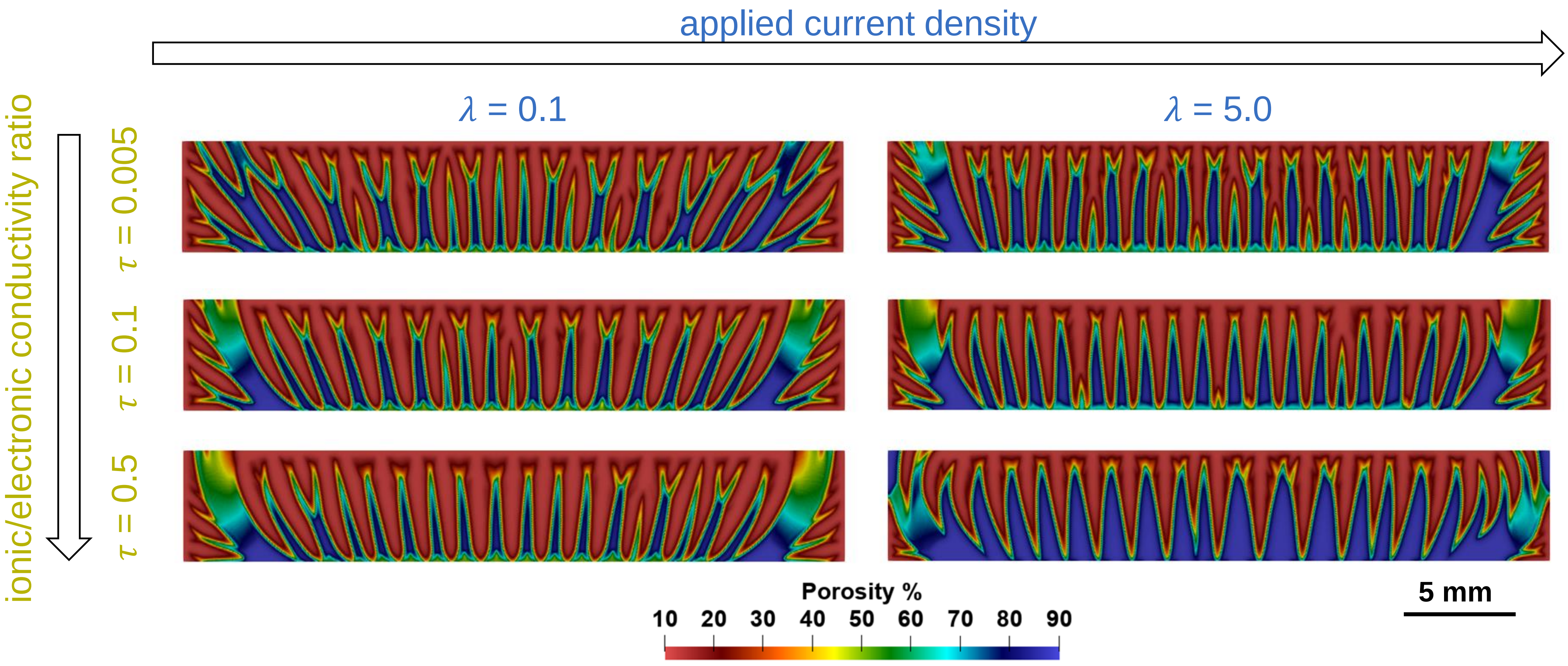
$$\Delta\phi = \phi_s - \phi_l - U_0$$

$$\delta = \frac{\text{ohmic resistance}}{\text{kinetic resistance}} \qquad \tau = \frac{\text{ionic conductivity}}{\text{electronic conductivity}}$$

$$\lambda = \text{applied current density}$$

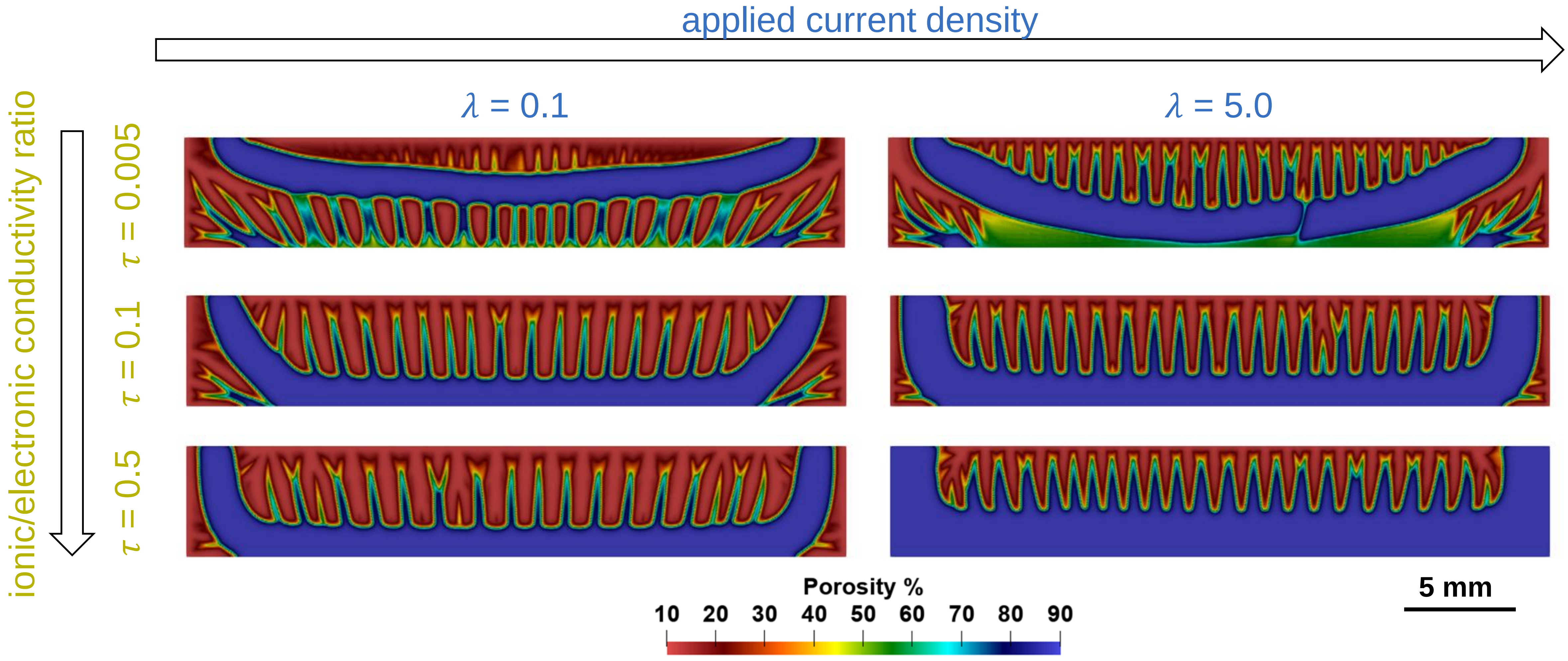
2D Results without fluid flow

$\delta = 1.0$



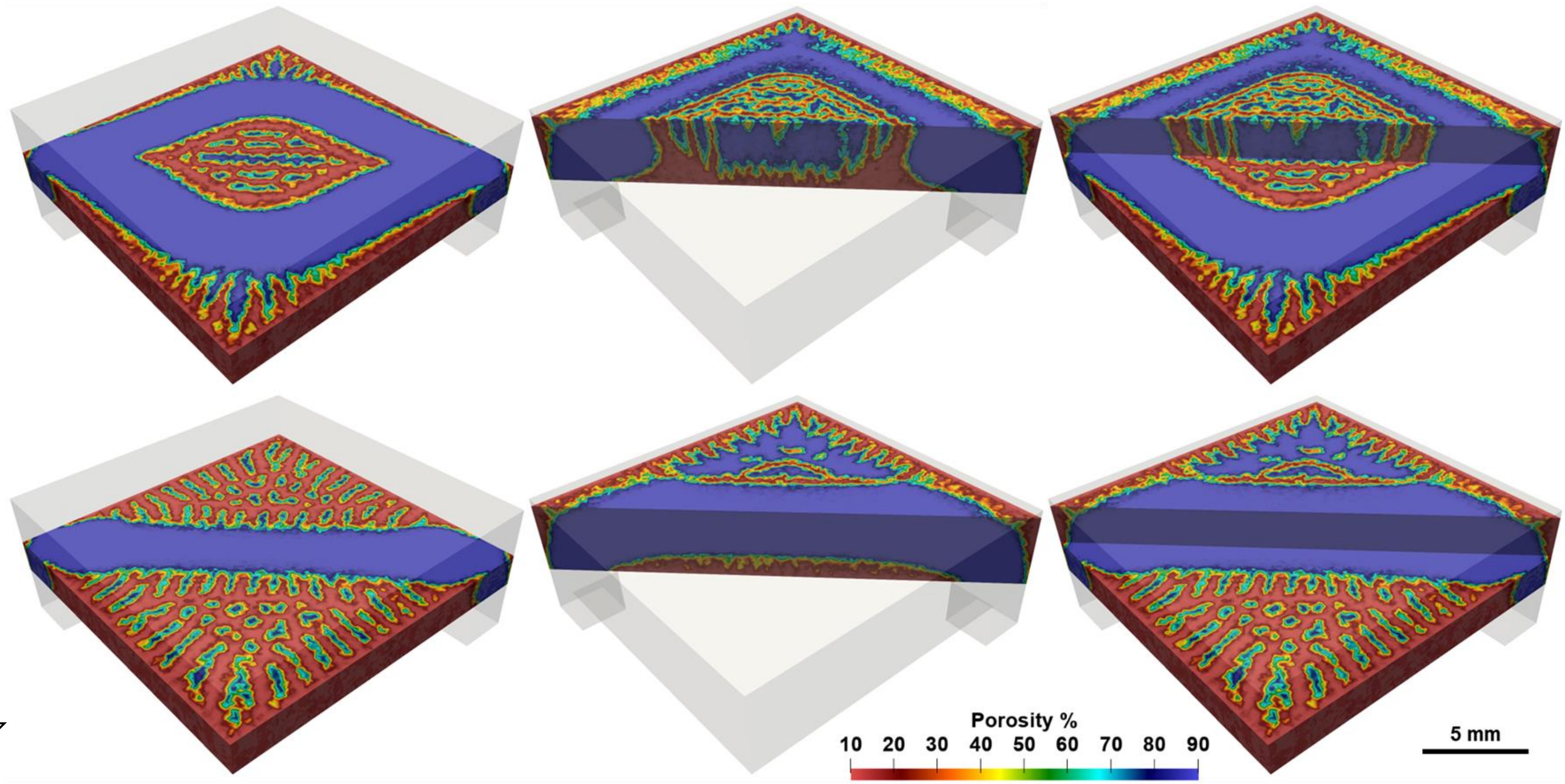
2D Results with fluid flow

$\delta = 1.0$



3D results

Increased ionic/electronic conductivity ratio, decreased applied current density

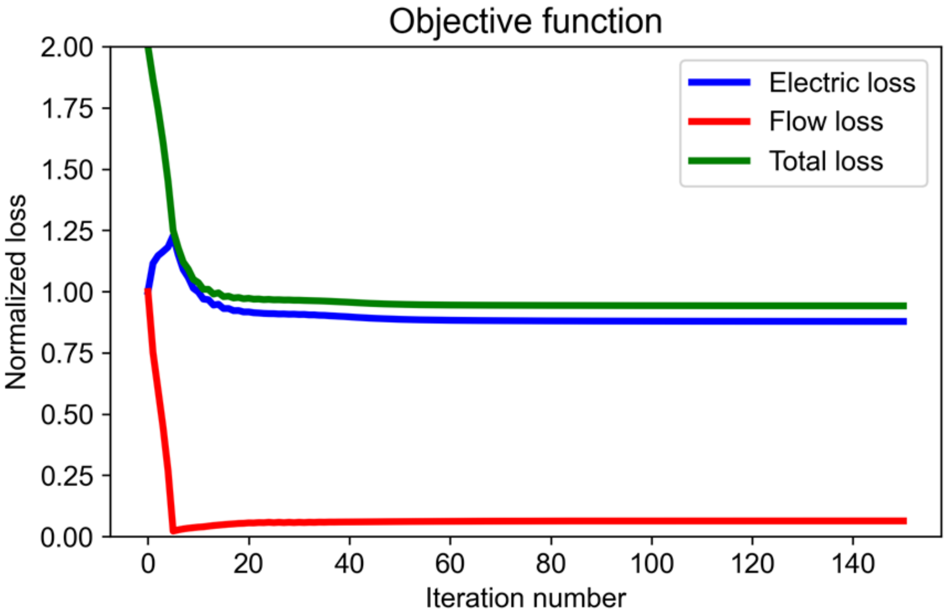


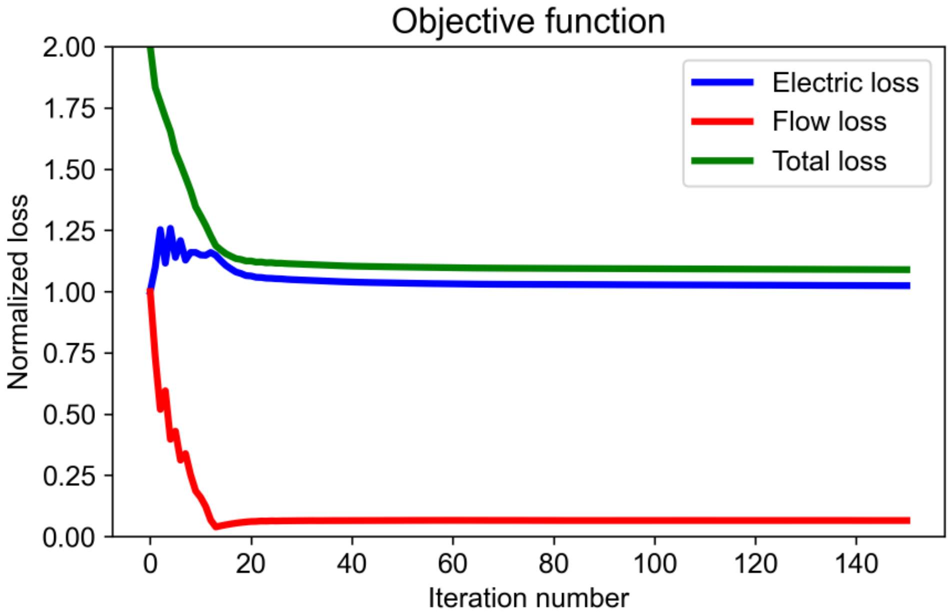
$\tau = 0.005$
$\lambda = 5.0$
$\delta = 1.0$

$\tau = 0.5$
$\lambda = 1.0$
$\delta = 1.0$

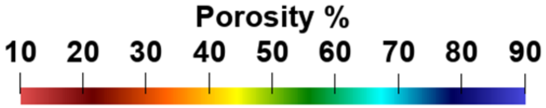
Convergence history

- Density-based topology optimization
- Objective functions: (1) Power dissipation, (2) Overpotential losses



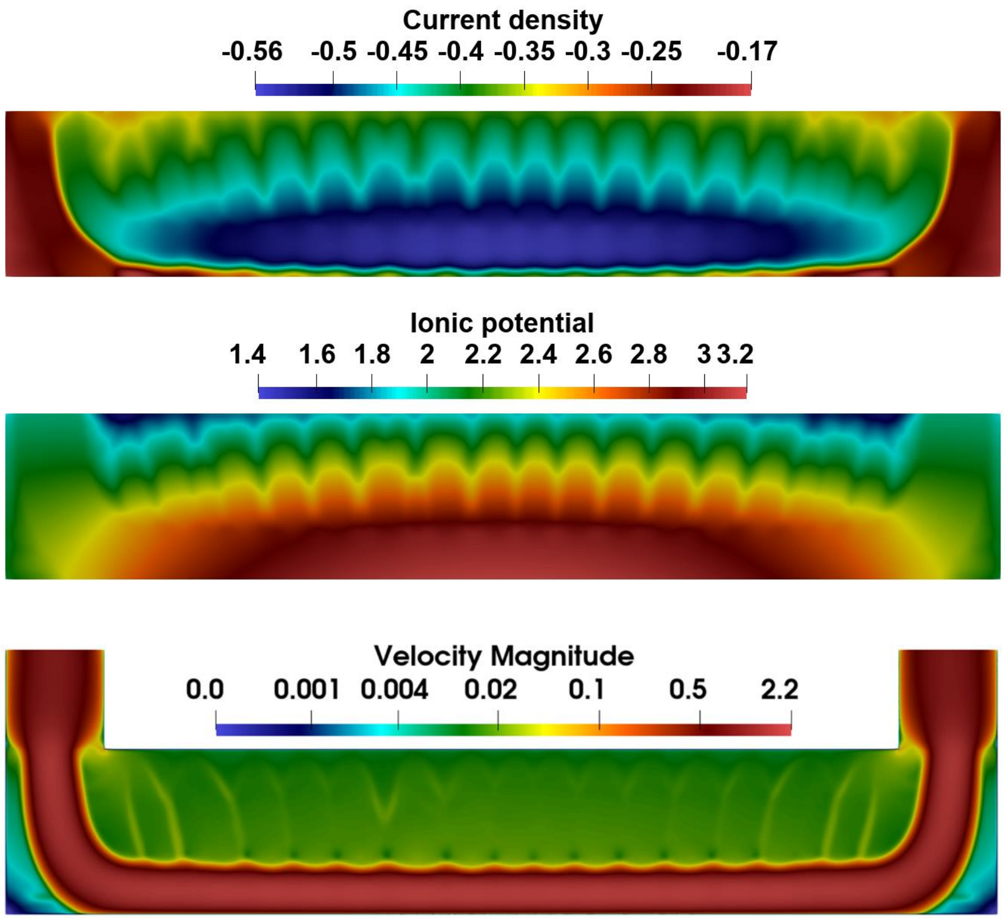


$\tau = 0.5$
$\lambda = 0.1$
$\delta = 1.0$

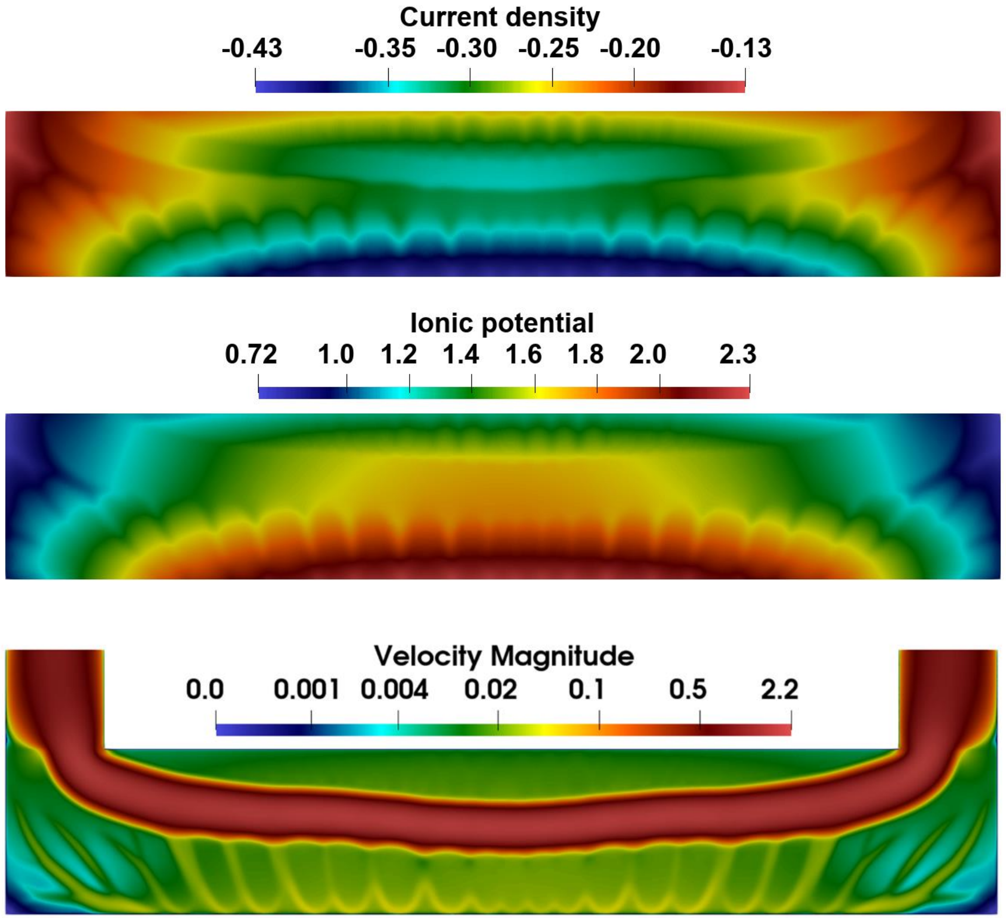


$\tau = 0.005$
$\lambda = 0.1$
$\delta = 1.0$

Other simulated quantities

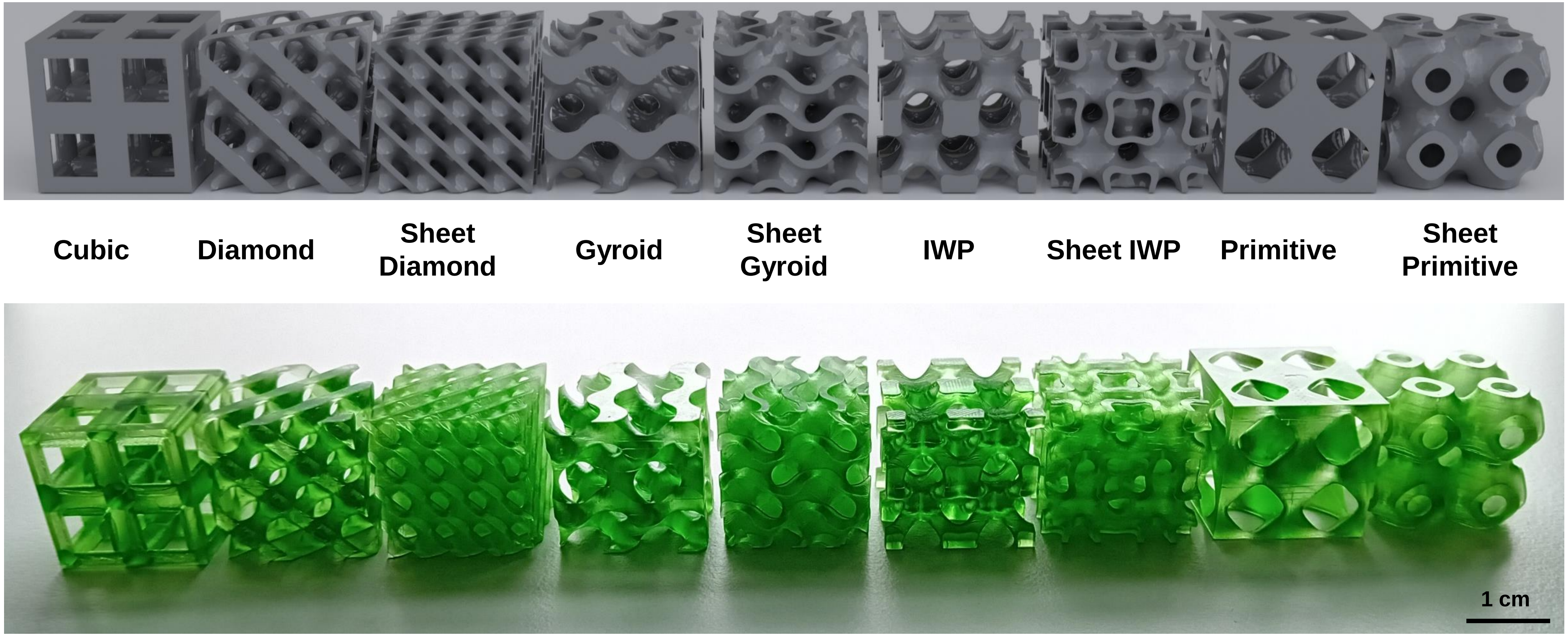


$\tau = 0.5$
$\lambda = 0.1$
$\delta = 1.0$



$\tau = 0.005$
$\lambda = 0.1$
$\delta = 1.0$

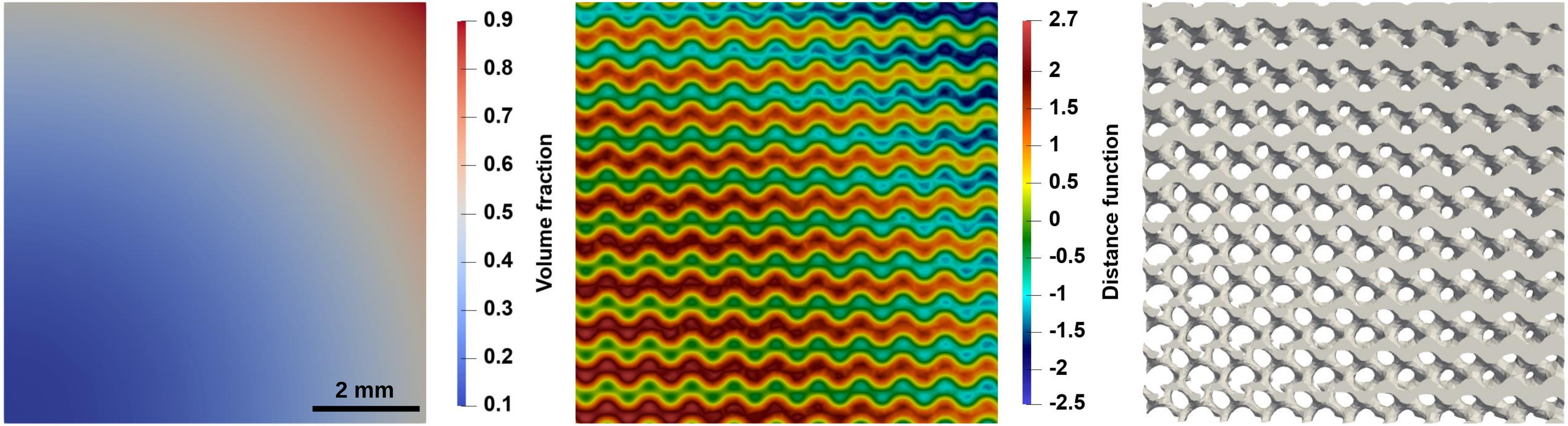
Triply periodic minimal surfaces (TPMS)



- Porous infills
- Highly interconnected
- Math-friendly!
- Ability to control transport properties

Transforming optimization results

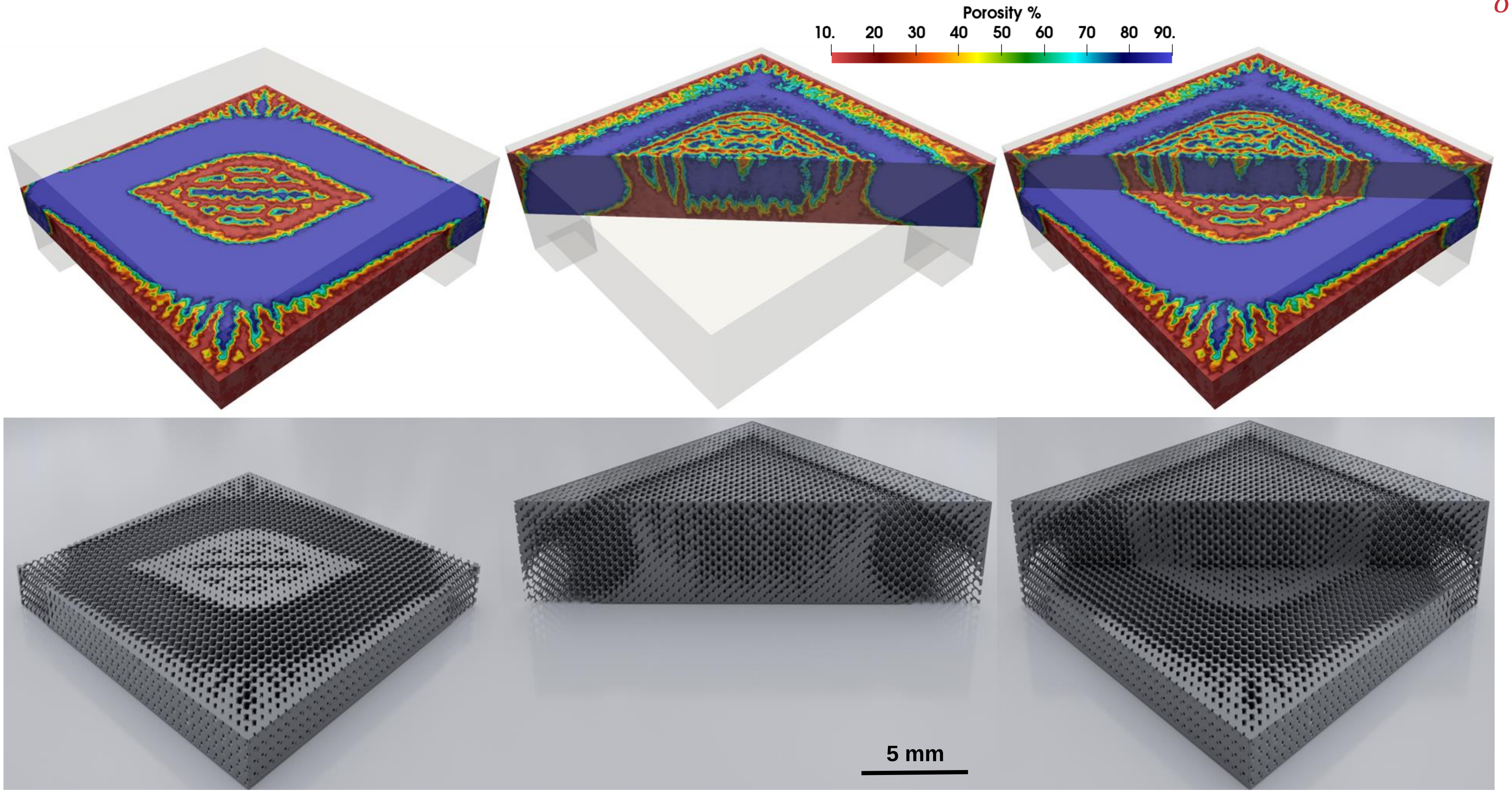
- Converting variable porosity to TPMS infills



(Volume fraction) (Distance function) (TPMS infill)

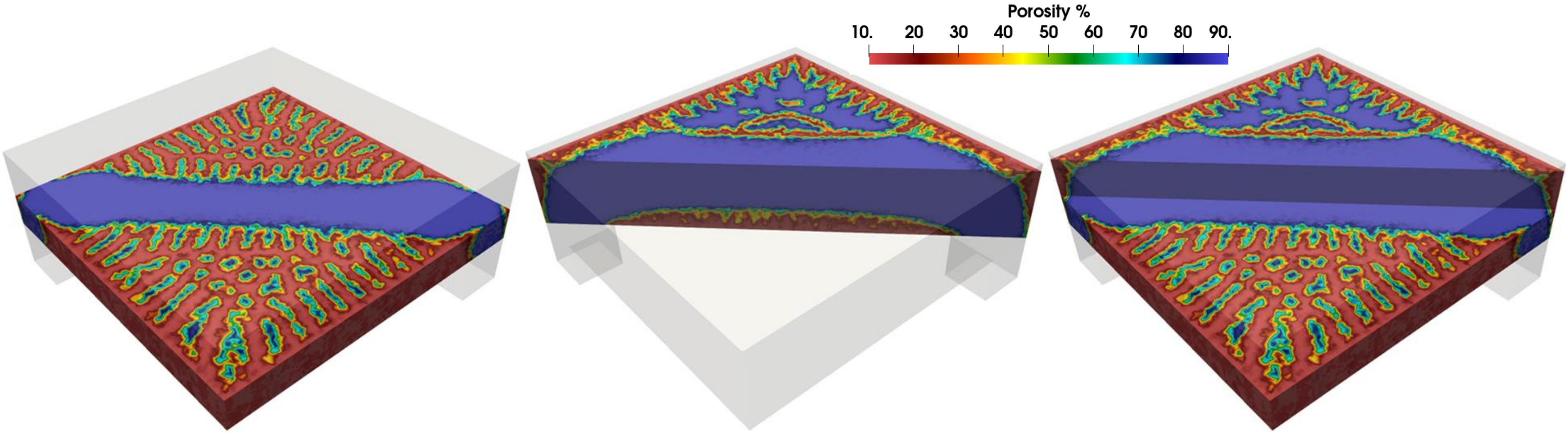
Conversion results #1 (conductivity ratio ↓, current density ↑)

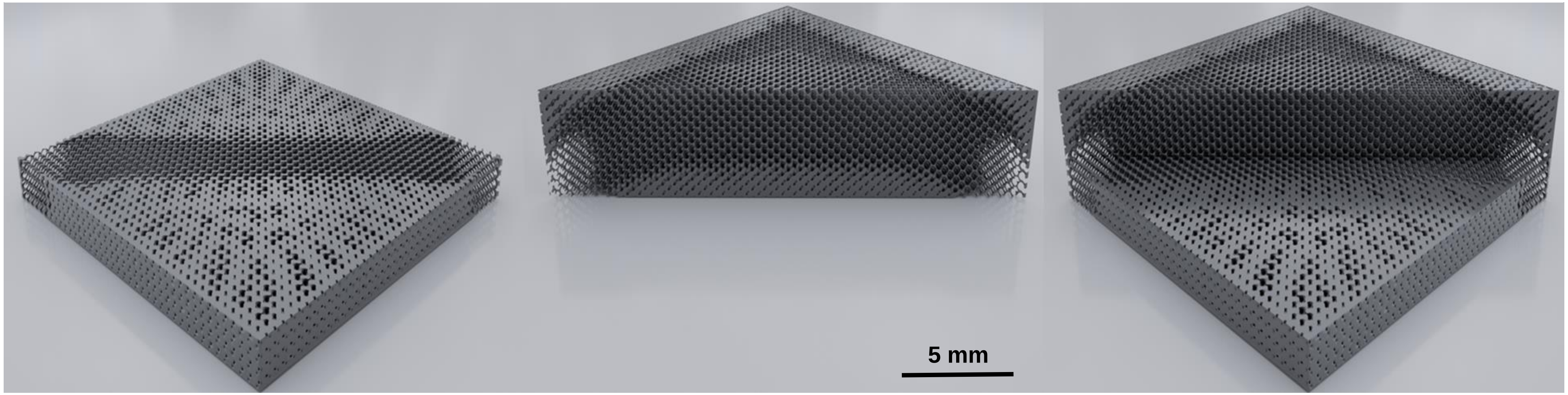
$\tau = 0.005$
$\lambda = 5.0$
$\delta = 1.0$



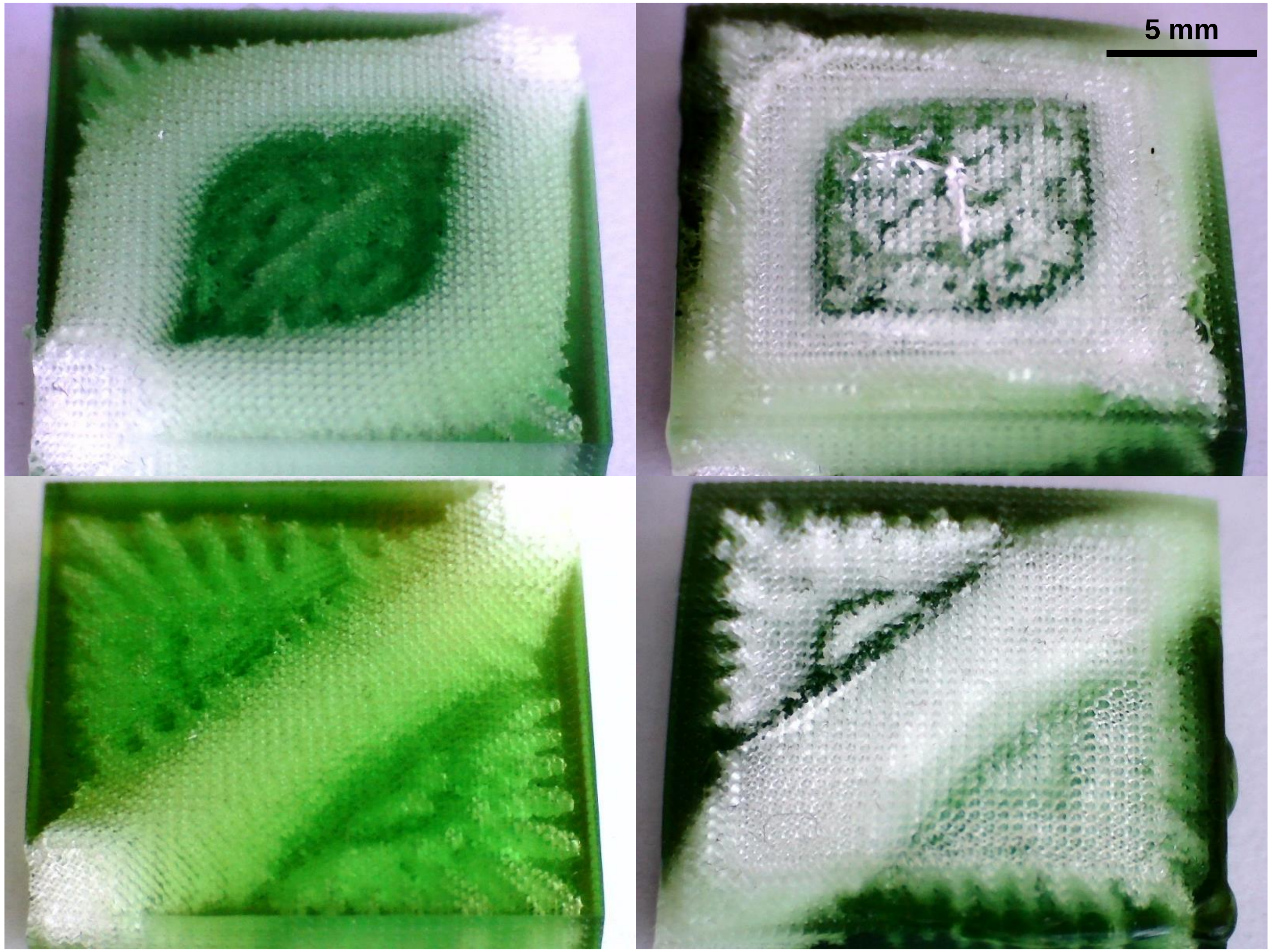
Conversion results #2 (conductivity ratio ↑, current density ↓)

τ = 0.5
λ = 1.0
δ = 1.0





3D printed optimized samples



Increased ionic/electronic conductivity ratio, decreased applied current density

$\tau = 0.005$
$\lambda = 5.0$
$\delta = 1.0$

$\tau = 0.5$
$\lambda = 1.0$
$\delta = 1.0$

Employed tools are all open-source!

Mesh generation
- GMSH
- SALOME

Multiphysics modeling
- Firedrake
- PETSc
- OpenFOAM

Topology optimization
- pyMMAopt
- PETSc

TPMS generation
- ASLI
- CGAL
- FreeFEM
- Mmg

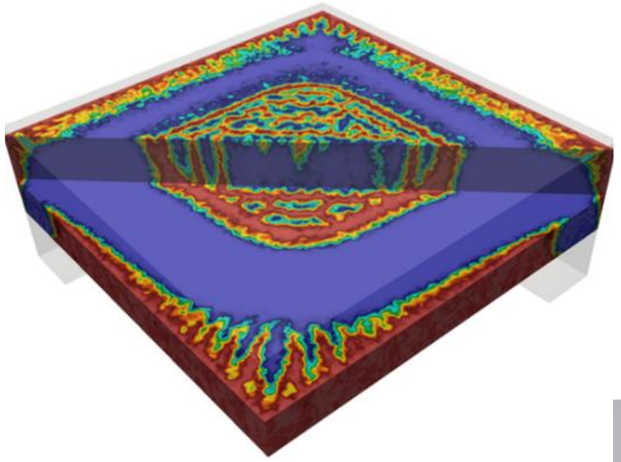
Postprocessing
- Blender
- ParaView
- MeshLab

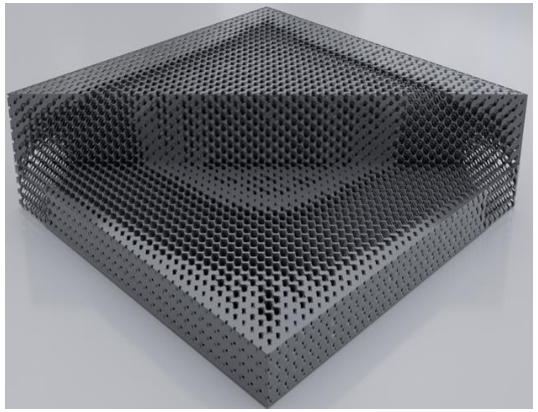
Conclusion

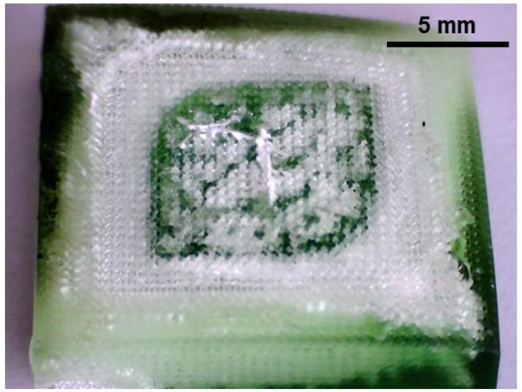
- Numerical models for correlating local configuration/structure to overall redox cell performance

- Scalable topology optimization for engineering porous electrodes

- Manufacturability by transforming results to TPMS infills







Thank You for Your Attention!

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