A Hybrid 3D/2D Model for Performance Predictions of Organic Flow Battery Cells

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Introduction

Flow batteries are an emerging technology to address the growing need for large-scale energy storage of intermittent energy sources, such as photovoltaics and wind. While the global electrochemical storage market is currently dominated by lithium-ion batteries, flow battery systems offer an attractive alternative thanks to their long lifetime, fire safety, and flexibility in scaling capacity and power.

Organic compounds offer a vast chemical space for potentially viable redox species. The possibility of local synthesis reduces the risks associated with global supply chains and represents a path towards more sustainable energy storage systems.

To accelerate the prototyping of new flow cells, we have developed a steady-state, non-isothermal model for performance predictions of single flow battery cells in COMSOL Multiphysics® [5]. The model is based on a macrohomogeneous description of the transport processes in the flow cell, which consists of current collectors, flow field channels, porous electrodes, and a semi-permeable membrane.

Hybrid 3D/2D Approach

- The cell model is divided into two submodels: a *3D electrolyte flow submodel* and a *coupled 2D electrochemical submodel*.
- The two submodels are coupled by using the averaged 3D velocity field of the flow model in the 2D electrochemical model.
- This hybrid approach allows for a significant reduction of the required computational resources, while capturing the impact of the 3D flow field.

3D Electrolyte Flow Submodel

- Solves for 3D velocity and pressure fields of the electrolyte flow described by:
  - Incompressible Navier-Stokes equation in the flow channels:

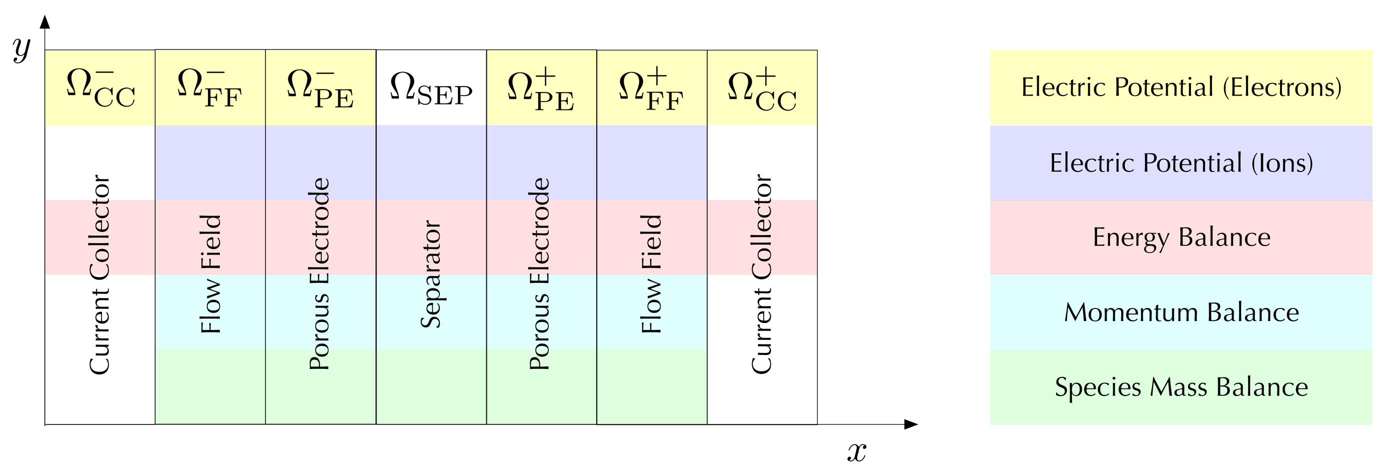
$$\rho(\mathbf{v}\cdot\nabla)\mathbf{v} = -\nabla p + \mu\nabla^2\mathbf{v}, \quad \nabla\cdot\mathbf{v} = 0$$

  - Brinkman equation in the porous electrode domains:

$$\mathbf{v} = -\frac{K_h}{\mu}\nabla p + \frac{K_h}{\varepsilon}\nabla^2\mathbf{v}$$

2D Electrochemical Submodel

- Solves coupled system of mass, charge, and heat transport.
- 2D cell domain extends along the
  - through-plane direction (x) of the cell assembly
  - and the forced-flow direction (y)
- 2D velocity field is obtained by averaging the 3D velocity field of the flow model along the z-direction (width of cell).



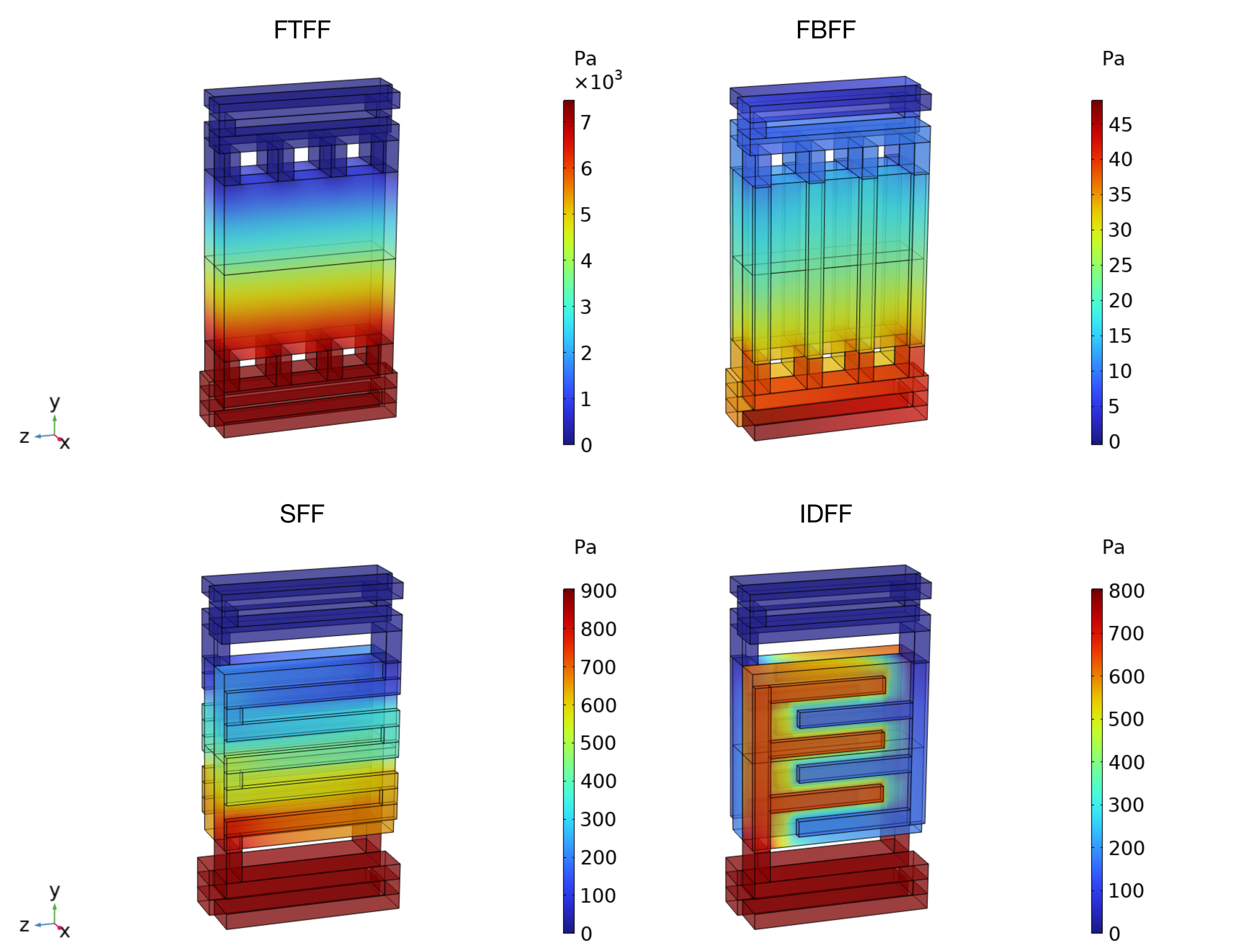
- Diffusion coefficient is evaluated based on an effective porous electrode model [2].
- Contact resistance between current collector and porous electrode is accounted for by an effective model $R_c(p_c) = a(p_c/p_c^{\mathrm{ref}})^{-b}$ $[\mathrm{m}\Omega\cdot\mathrm{cm}^2]$ fit to experimental data.
- Summary of balance laws in the electrochemical submodel:

Variable	Flux	Balance Law
ϕ_s	$\boldsymbol{J}_s = -\sigma_s^{\mathrm{eff}}\nabla\phi_s$	$\nabla\cdot\boldsymbol{J}_e = S_i$
ϕ_l	$\boldsymbol{J}_l = -(\sigma_l^{\mathrm{eff}}\nabla\phi_l + \sum_\alpha z_\alpha\varepsilon D_\alpha^*\nabla c_\alpha)$	$\nabla\cdot\boldsymbol{J}_l = -S_i$
c_α	$\boldsymbol{N}_\alpha = -\varepsilon(D_\alpha^*\nabla c_\alpha + z_\alpha D_\alpha^{\mathrm{eff}} c_\alpha\nabla\phi_l)$	$\nabla\cdot(c_\alpha\boldsymbol{v} + \boldsymbol{N}_\alpha) = S_\alpha$
T	$\boldsymbol{q} = -\lambda\nabla T$	$\nabla\cdot(c_p\boldsymbol{v}T + \boldsymbol{q}) = S_T$

- Here we consider the all-organic MV/TEMPTMA system with half-cell reactions:

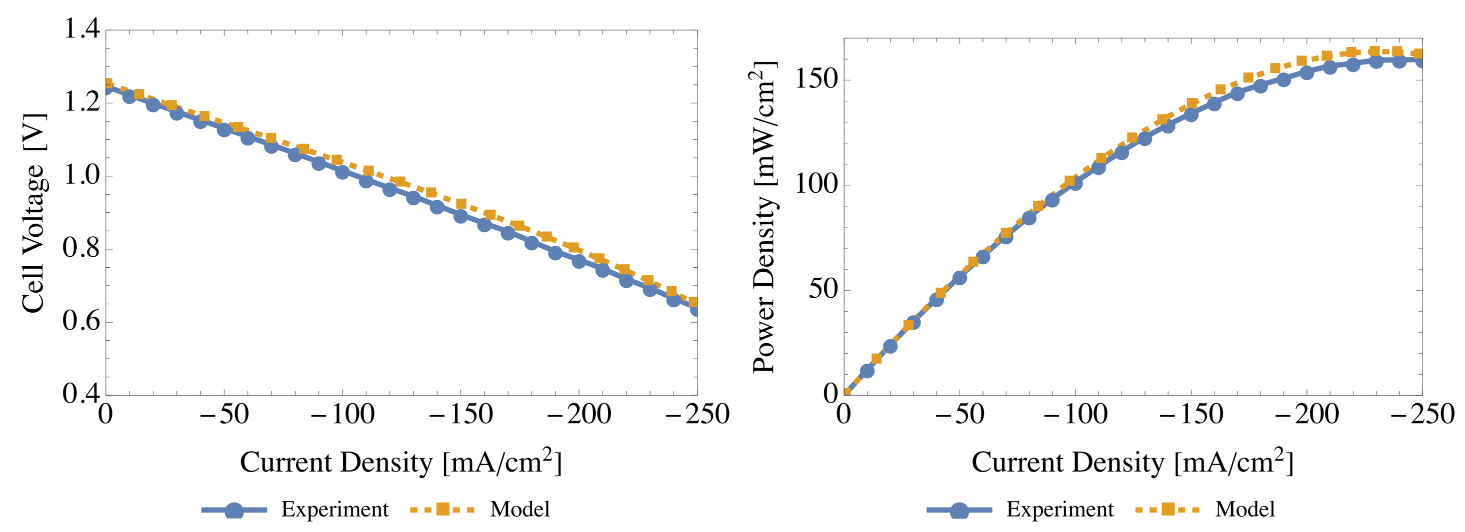
$$\mathrm{MV}^{2+} + \mathrm{e}^- \underset{\text{Discharge}}{\overset{\text{Charge}}{\rightleftharpoons}} \mathrm{MV}^{+}, \quad \mathrm{T}^{2+} + \mathrm{e}^- \underset{\text{Charge}}{\overset{\text{Discharge}}{\rightleftharpoons}} \mathrm{T}^{+}$$

Pressure Distribution with Different Flow Fields



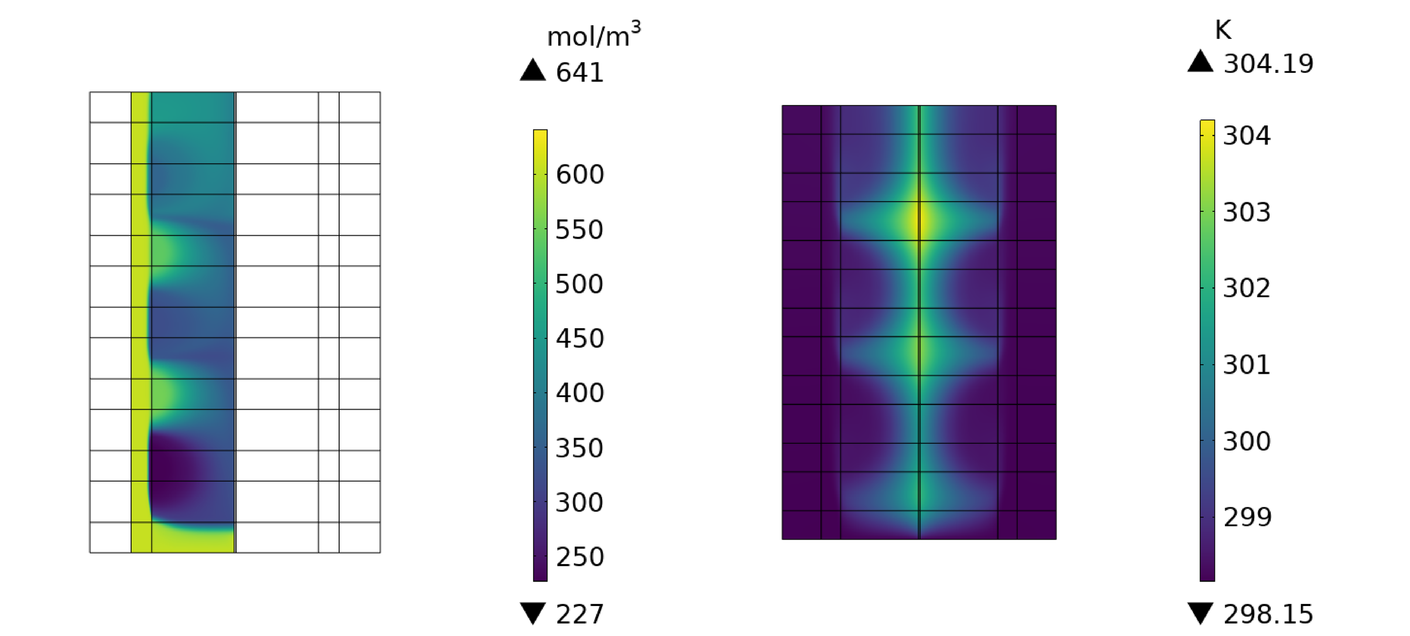
Pressure drop at a fixed flow rate $Q = 16\,\mathrm{mL/min}$ for the flow-through flow field (FTFF), flow-by flow field (FBFF), serpentine flow field (SFF), and interdigitated flow field (IDFF).

Validation with the MV/TEMPTMA System



Predicted polarization curves and experimental data of a lab-sized $5\,\mathrm{cm}^2$ cell [1].

Resolved Fields for the IDFF Geometry



Species concentration of MV^{2+} in the flow field channels and porous electrode in the negative half-cell and temperature distribution at $i_{\mathrm{cell}} \approx -40\,\mathrm{mA\,cm^{-2}}$.

References

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