OpenFCST: Fuel Cell Simulation Toolbox

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ESDL ENERGY SYSTEMS DESIGN LABORATORY

UNIVERSITY OF ALBERTA

Overview

- Introduction and motivation
- OpenFCST framework
- Case studies:
  - Membrane electrode assembly model with multi-step reaction kinetics
  - Multi-component gas transport model
- Conclusions

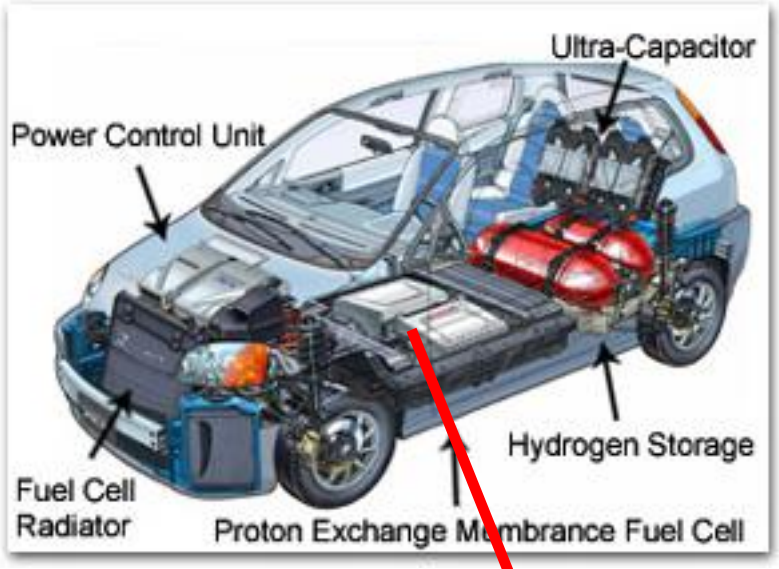
Introduction

- PEM fuel cells are efficient energy conversion devices
- Their fuel is usually pure hydrogen and their only emission is water vapour
- They are an alternative to internal combustion engines and batteries for transportation and portable applications
- PEM fuel cell cars and buses are currently operating in both Europe and North America

Source: http://www.iveho.com/2011/03/18/mercedes-benz-vehicles-circle-the-globe/

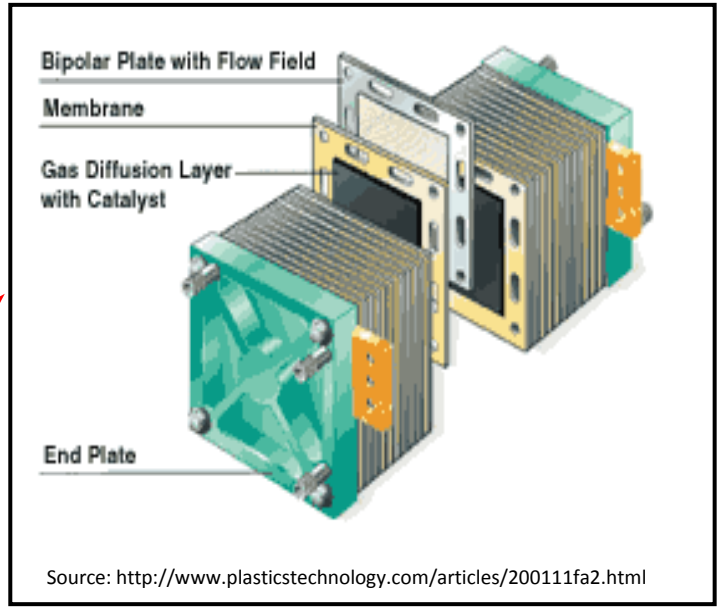
Source http://www.isecorp.com/gallery/albums/BC-Transit-Fuel-Cell-Bus/BCTransit_fuel_cell_bus.jpg

Introduction



Source: http://www.h2education.com/images/misc/ge_content/hondaFCX.jpg

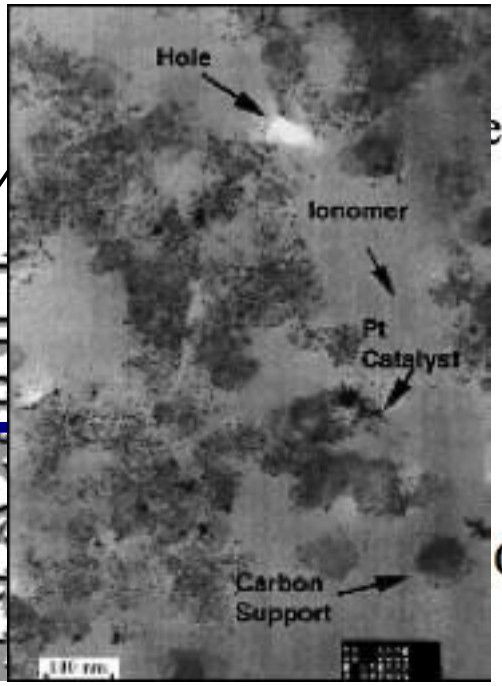
Source: http://www.palcan.com/s/Products.asp



Source: http://www.plasticstechnology.com/articles/200111fa2.html

Introduction

Source: http://www.me.ust.hk/~mezhao/research/diffusion.html



Source:Siegel et al, "Single domain PEMFC model based on agglomerate catalyst geometry",JPS, 2003

Introduction

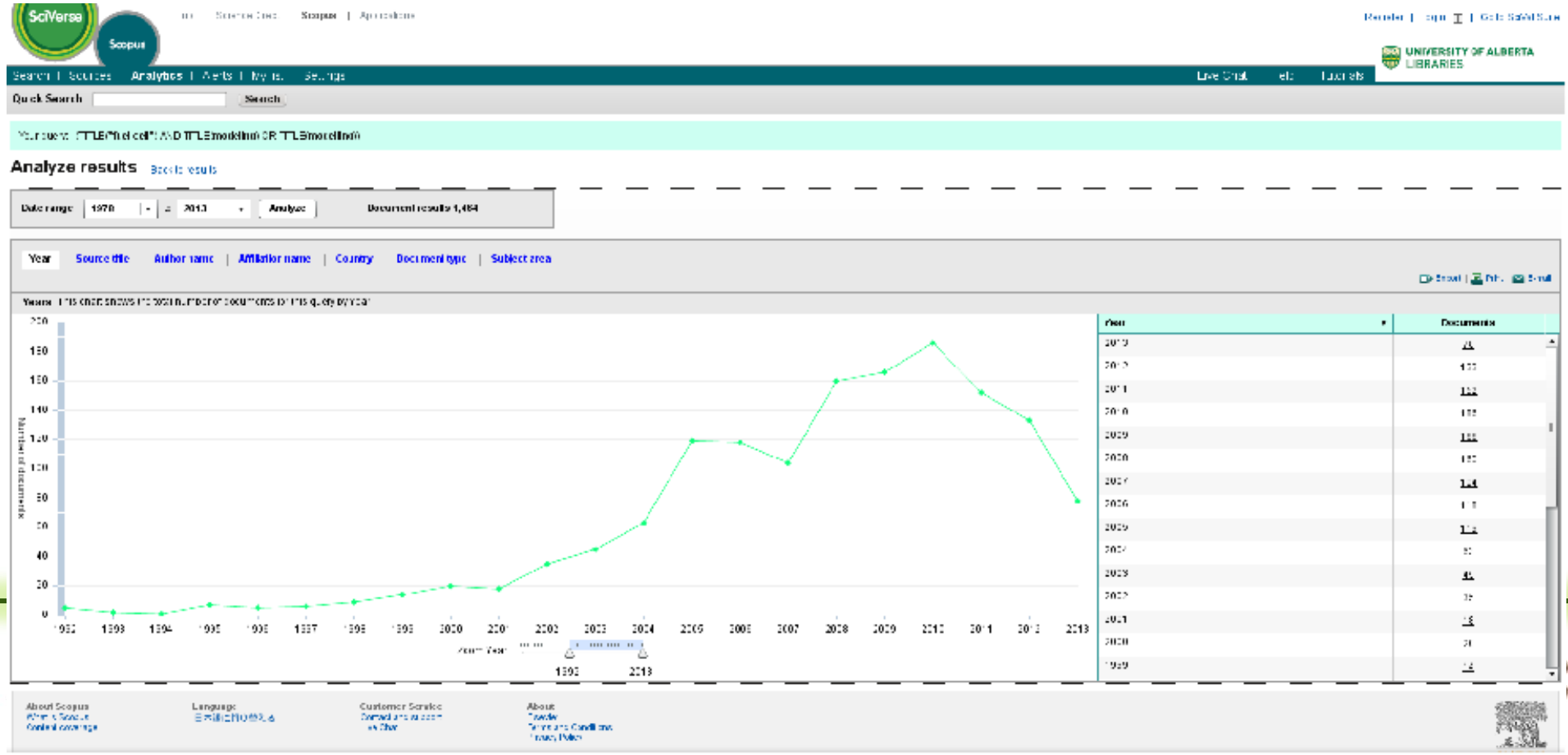
- Fuel cell operation involves a myriad of coupled physical process
  - Multi-component reactant gas transport
  - Charge transport: Proton and electron transport
  - Liquid water transport: Two-phase flow
  - Heat transport
  - Electrochemical reaction
- Fuel cell mathematical models must account for all these physical process simultaneously
  - Complex coupled problem

Motivation

- Fuel cell mathematical models are constantly evolving due to its coupled, multi-physics nature
  - Multi-component gas transport in micro- and meso-porous materials is not well understood
  - Two-phase flow models are still in development such as a two-fluid models with a mixed wettability pore-size distribution closure
  - Multi-step electrochemical reactions only now being introduced
  - Transient models need to be further developed to analyze cold-start, purge system and degradation studies

Motivation

- A Scopus search for “fuel cell” AND “modelling” returned over 100 articles per year over the past decade



Motivation

- Most article are based on:
  - Mathematical models already available in commercial software
    - Difficult to develop new physical models → Limited physical insight
  - In-house codes
    - Codes are not made available to other research groups → Re-invent the wheel
    - Focused on one set of novel physical phenomena → Difficult to assess the true impact of the new model due to lack of coupling

Motivation

- There is a need to develop a collaborative fuel cell mathematical modelling software that can be shared within the fuel cell community
- The package should be:
  - Open-source and available to the community
  - Useful to both users and mathematical model developers
  - Easily expandable

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What is OpenFCST?

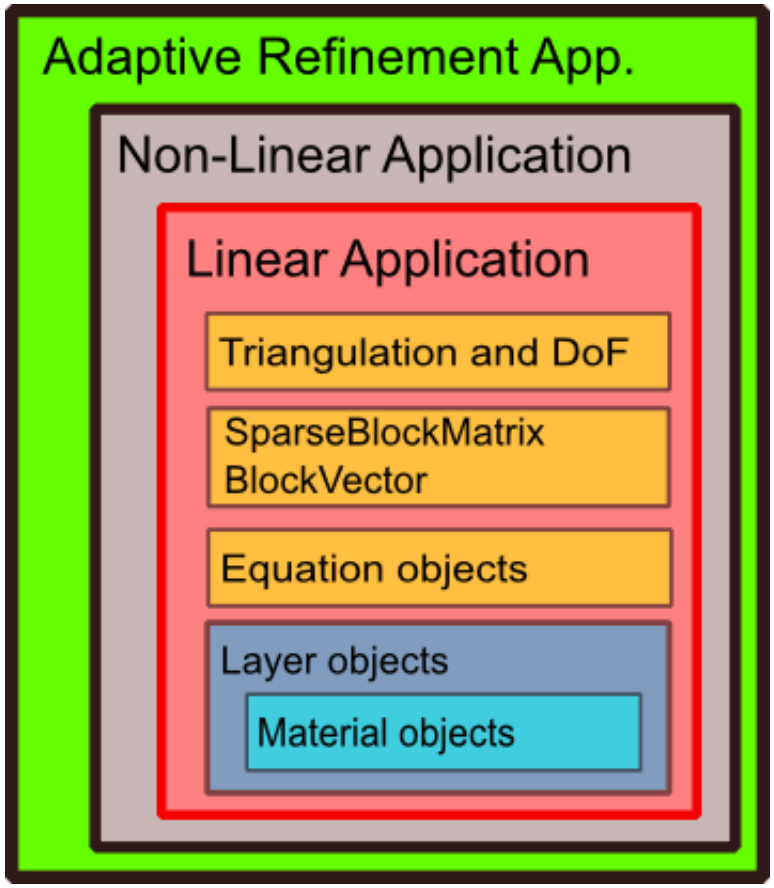
- The Fuel Cell Simulation Toolbox (FCST) is an open-source mathematical modelling software for polymer electrolyte fuel cells
- It contains
  - Pre-processors: A fuel cell specific grid generator and a class to read meshes from a mesh generator (we use SALOME, i.e. UNV)
  - Solvers: A library of materials, layers and physical equations as well as linear and non-linear solvers**. FEM routines provided by deal.II**.
  - Post-processors: Functional evaluation algorithms and a VTK file generator
  - Design and optimization capabilities: Parametric studies, parameter estimation and optimization algorithms. **Optimization functionality provide by Dakota.**

Philosophy

- Develop an easily expandable toolbox by developing a flexible interface for each component of a fuel cell
- Accessible to both industry and academia
  - Released under MIT License
  - Supported and used by the Automotive Fuel Cell Cooperation Corp.
- Accessible to users and code developers:
  - Users:
    - Many options available through text/XML input files
  - Developers:
    - Easy to develop and integrate new material, layer, equation classes by means of inheritance, well documented base classes and already available examples
    - Easy to develop and integrate your own applications using already available material, layer and equation classes

Main components of the code

- Pre-processor
- Application framework
- Equation framework
- Reaction database
- Layer database
- Materials database
- Post-processor

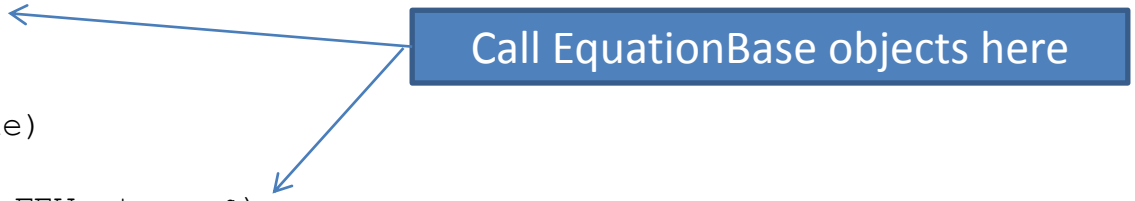


Application framework

- Based on Dr. Guido Kanschat ‘s AppFrame and MeshWorker framework
- Two main components
  - FEM applications
    - Generate and store Triangularization<dim>
    - Read parameter file and initialize equation, layer and material objects
    - Loop over cells and assemble the FEM global matrix and right hand side
    - Solve the linear system
  - Solver applications
    - Implements solution strategies for adaptive refinement, non-linear solvers and transient algorithms

FEM application interface

```
virtual void declare_parameters (ParameterHandler &param);

virtual void initialize (ParameterHandler &param);

virtual void remesh ();

virtual double residual (
    FEVector &dst,
    const FEVectors &src,
    bool apply_boundaries=true)

virtual void assemble (const FEVectors &)

virtual void solve (
    FEVector &start,
    const FEVectors &rhs)

virtual double estimate (const FEVectors &src)

virtual double evaluate (const FEVectors &src)

virtual void data_out (
    const std::string &filename,
    const FEVectors &src,
    const std::vector< std::string >)
```



Equation framework

- Equation classes implement the weak form of the PDE that needs to be solved
- Equation classes are used to
  - Provide applications with couplings between solution variables
  - Assemble the local cell matrix of the PDE
  - Assemble the local cell residual vector of the PDE
  - Assemble local boundary matrix and residual terms of the PDE based on Newmann/Robin B.C.
- Equations receive as input:
  - FEValues and Solution via CellInfo object in MeshWorker
  - Layer classes

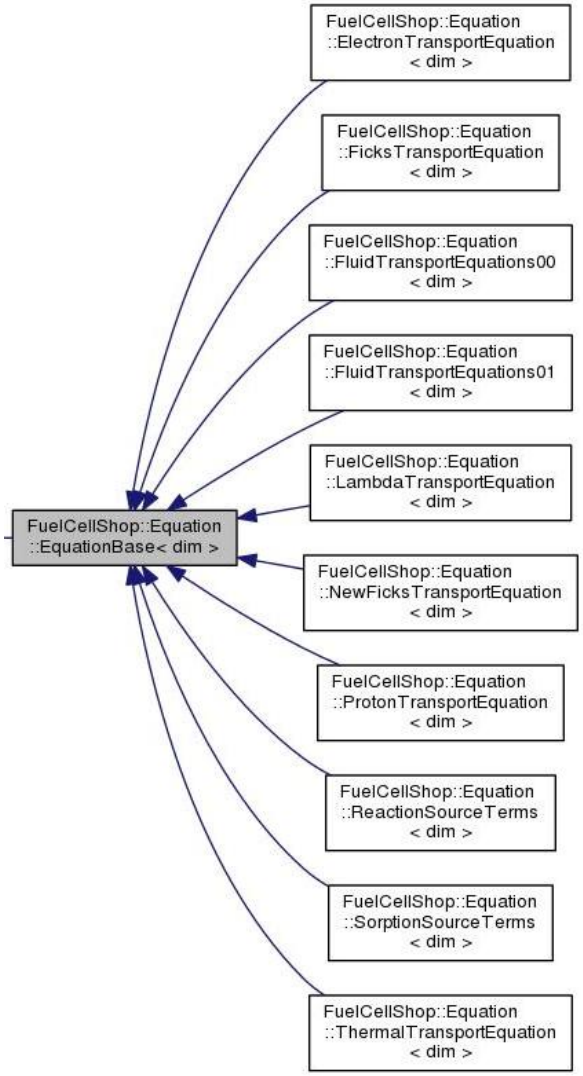
Equation framework

Local CG FEM based assemblers

```
virtual void assemble_cell_matrix (
    AppFrame::MatrixVector &cell_matrices,
    const typename AppFrame::DoFApplication< dim >::CellInfo
    &cell_info,
    FuelCellShop::Layer::BaseLayer< dim > *const layer)

virtual void assemble_cell_residual (
    AppFrame::FEVector &cell_residual,
    const typename AppFrame::DoFApplication< dim >::CellInfo
    &cell_info,
    FuelCellShop::Layer::BaseLayer< dim > *const layer)

virtual void assemble_bdry_matrix (
    AppFrame::MatrixVector &bdry_matrices,
    const typename AppFrame::DoFApplication< dim >::FaceInfo
    &bdry_info,
    FuelCellShop::Layer::BaseLayer< dim > *const layer)

virtual void assemble_bdry_residual (AppFrame::FEVector
    &bdry_residual, const typename AppFrame::DoFApplication<
    dim >::FaceInfo &bdry_info,
    FuelCellShop::Layer::BaseLayer< dim > *const layer)
```

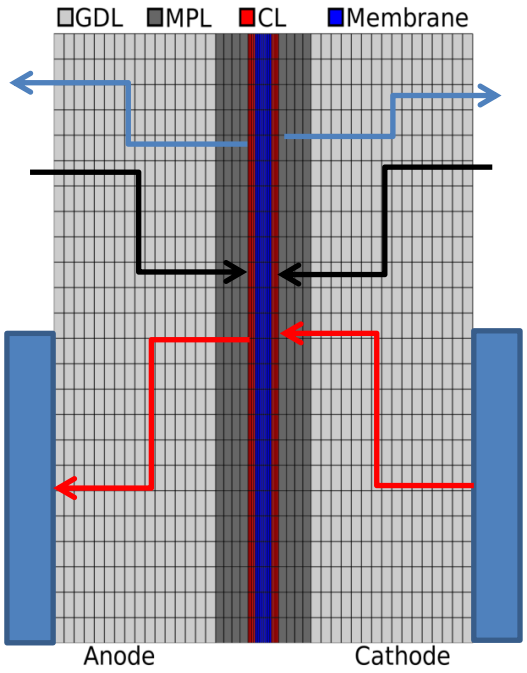
Accessors and info

```
const couplings_map & get_internal_cell_couplings () const
```



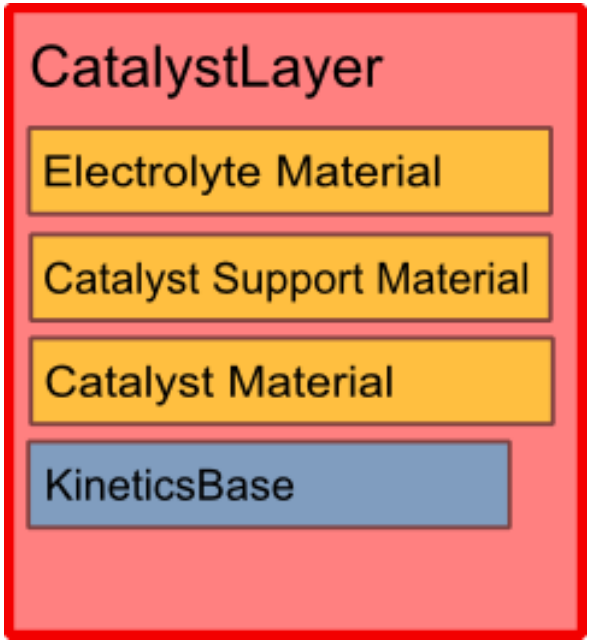
Layer framework

- A fuel cell is composed of about seven distinct layers, i.e. GDL, MPL, CL and PEM.
- Each layer is a porous composite material with at least two materials where at least two phases co-exist
- An interface has been developed for each layer in order to provide
  - Effective transport properties
  - Derivatives of effective transport properties
  - Multi-scale integration requirements



Layer framework

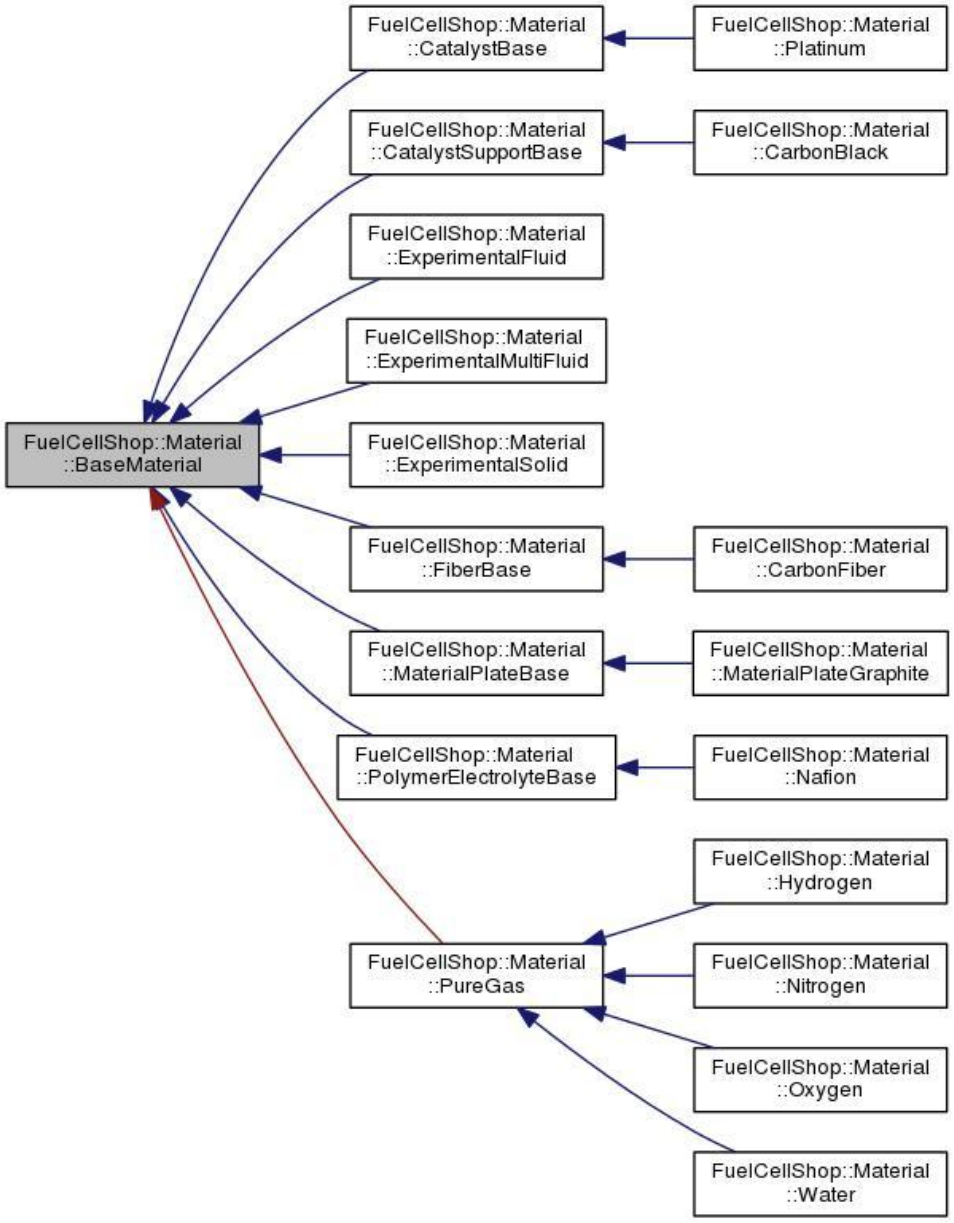
- Layers contain several materials objects which are used, together with effective medium theories such as percolation theory to estimate the effective properties
- If the layer is reactive, i.e. a catalyst layer, then a kinetics object is also stored and used to compute reaction rates in the layer



- In order to allow users to swap layers via the input file, inheritance is used:
  - Application contains only a pointer to the base layer class
  - Base layer contains a map of all children
  - Once the application has read the input file, the child is initialized as appropriate

Material database

- Material database contains information on:
  - Gases: T_c, p_c, enthalpy, entropy, etc.
  - Catalysts: Activation energy, transfer coef., etc.
  - Catalyst supports
  - Fibers (for GDLs)
  - Electrolytes
- Materials of the same family can be swapped at real time



Kinetics framework

- Fuel cell electrochemical reactions are complex, multi-step reactions involving many intermediates
  - To date, a detailed mathematical model does not exist
- An interface to explore novel kinetics has been developed
- Currently it contains:
  - A Tafel equation (simplest electrochemical reaction model)
  - A Butler-Volmer equation
  - A Double-trap multi-step kinetic model for the ORR
  - A Dual-trap multi-step kinetic model for the HOR

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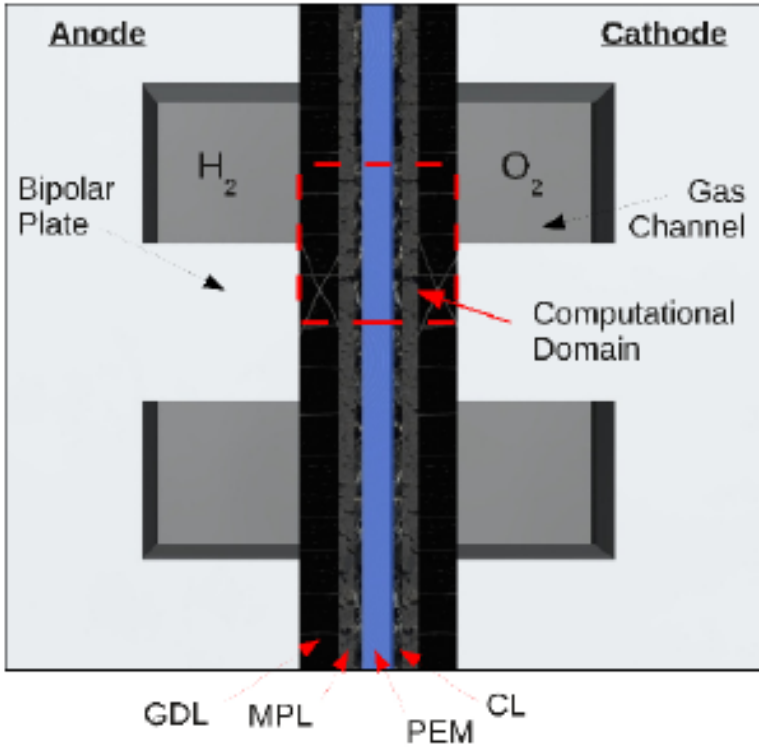
Case study 1:
Membrane electrode assembly with multi-step oxygen reduction reaction kinetics

Mathematical fuel cell model (I)

1. Steady state and isothermal operation.
2. Macro-homogeneous model.
3. Solving for:
   - Oxygen mole fraction, x_{O_2}.
   - Water vapour mole fraction, x_w.
   - Electrolyte potential, ϕ_m.
   - Solid phase potential, ϕ_s.
   - Absorbed water in the ionomer phase, λ.



Mathematical fuel cell model (II)

1. Fick's Law for species transport:

$$\nabla \cdot (c_{total} D_{O_2}^{eff} \nabla x_{O_2}) = \frac{1}{nF} \nabla \cdot i$$

$$\nabla \cdot (c_{total} D_{W}^{eff} \nabla x_{w}) = \frac{1}{nF} \nabla \cdot i + S_\lambda$$

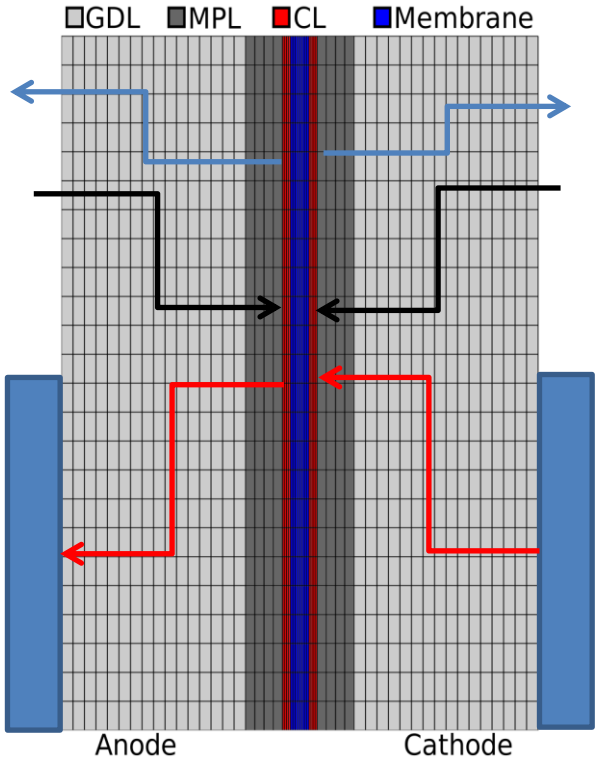
2. Ohm's Law for proton and electron transport:

$$\nabla \cdot (\sigma_m^{eff} \nabla \phi_m) = \nabla \cdot i$$

$$\nabla \cdot (\sigma_s^{eff} \nabla \phi_s) = -\nabla \cdot i$$

3. Springer model for absorbed water transport:

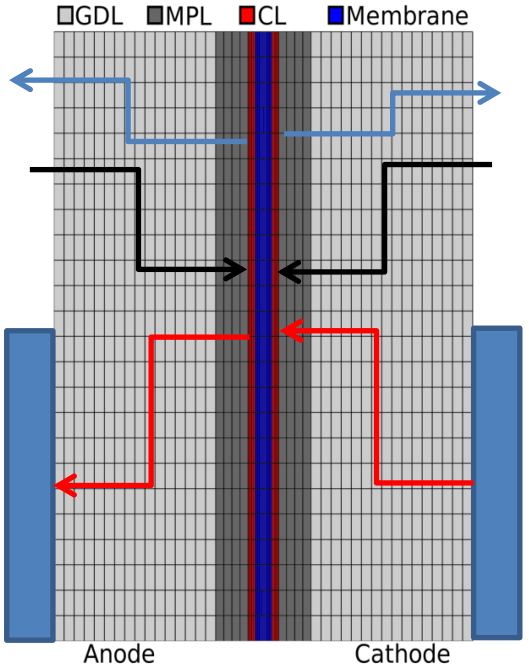
$$\nabla \cdot \left(n_d \frac{\sigma_m}{F} \nabla \phi_m + \frac{\rho_{m,dry}}{EW} D_\lambda \nabla \lambda \right) = -S_\lambda$$



Mathematical fuel cell model (III)

- Solution variables are solved in appropriate domains

Electrode	Anode				Cathode		
Unknown variable	GDL	MPL	CL	PEM	CL	MPL	GDL
O_2 molar fraction, x_{O_2}					X	X	X
H_2O molar fraction, x_{H_2O}	X	X	X		X	X	X
Electron potential, ϕ_s	X	X	X		X	X	X
Proton potential, ϕ_m			X	X	X		
Water absorbed by ionomer, λ			X	X	X		



- Coupling achieved via source terms:
  - Water in ionomer ↔ water mole fraction in gas phase
  - Proton potential ↔ Electron potential

Mathematical model: Electrochemical reactions

- Oxygen reaction given by the following multi-step reaction kinetic pathway
  - Dissociative adsorption (DA): $1/2O_2 \Leftrightarrow O_{ad}$
  - Reductive adsorption (RA): $1/2O_2 + H^+ + e^- \Leftrightarrow OH_{ad}$
  - Reductive transition (RT): $O_{ad} + H^+ + e^- \Leftrightarrow OH_{ad}$
  - Reductive desorption (RD): $OH_{ad} + H^+ + e^- \Leftrightarrow H_2O$

- The source term in the cathode is the:

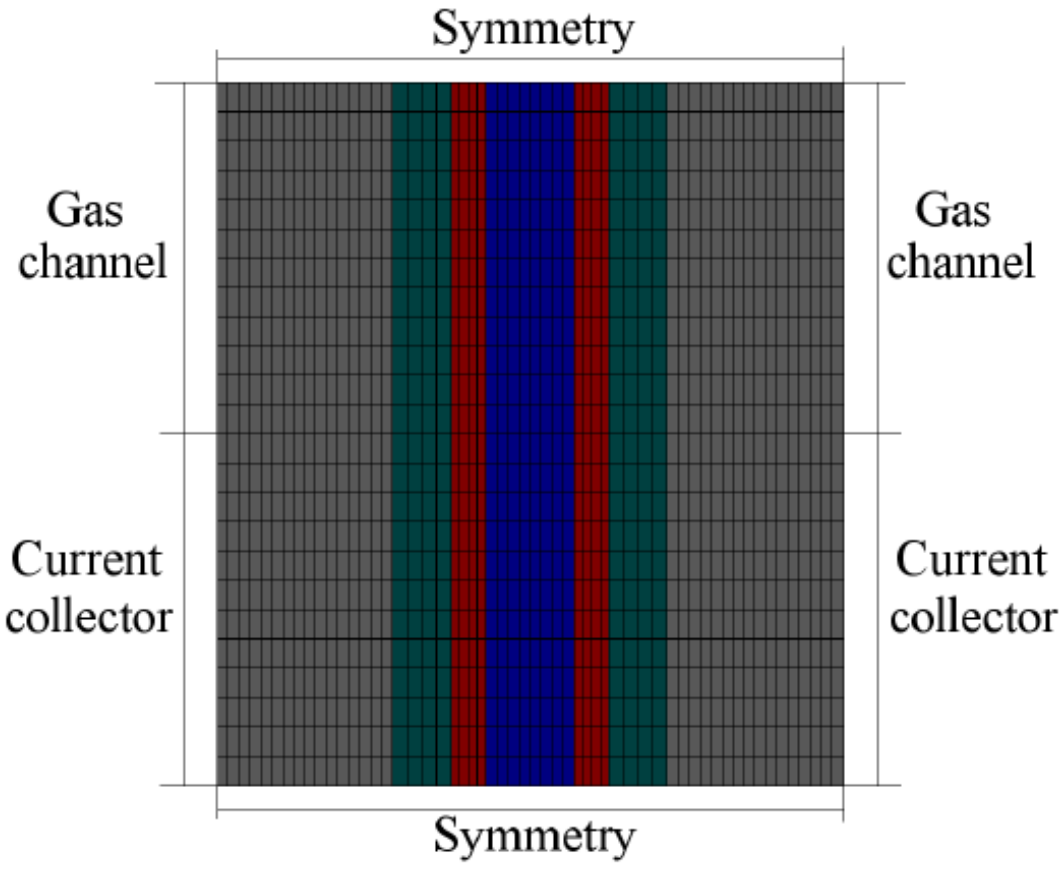
$$\nabla \cdot i = j_k = j^* e^{\frac{-\Delta G^*_{RD}}{kT}} \theta_{OH} - j^* e^{\frac{-\Delta G^*_{-RD}}{kT}} (1 - \theta_O - \theta_{OH})$$

$$\Delta G^*_{RD} = \Delta G^{*0}_{RD} + \beta e \eta \qquad \Delta G^*_{-RD} = \Delta G^{*0}_{RD} + \Delta G^0_{OH} - \beta e \eta$$

where $\eta = \phi_s - \phi_m - E_{eq}$

Mathematical model: Boundary conditions

- Gas channel
  - Concentrations specified for all gases
  - No flux of electrons, protons
- Current collector
  - Electrical potential specified
  - No flux of gases, protons



Nonlinear finite element solution

- Solve system using Newton's method
  - Solve a linearization of the system

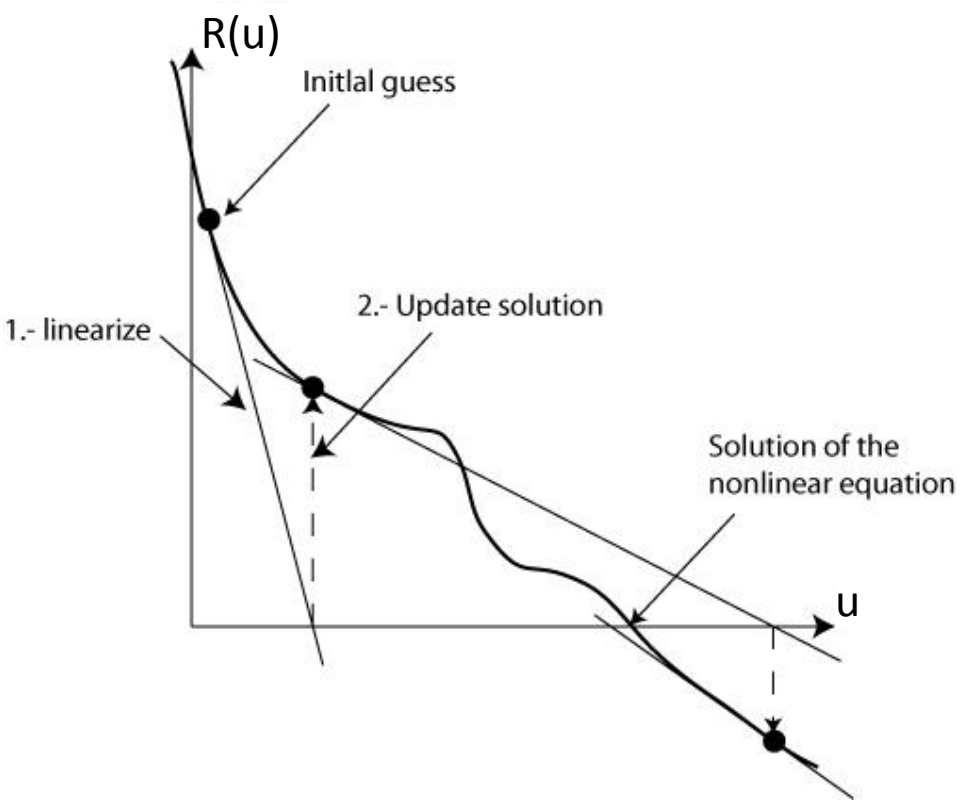
$$\frac{dR(u,\lambda)}{du}\delta u = -R(u,\lambda)$$

  using

    Galerkin finite element method

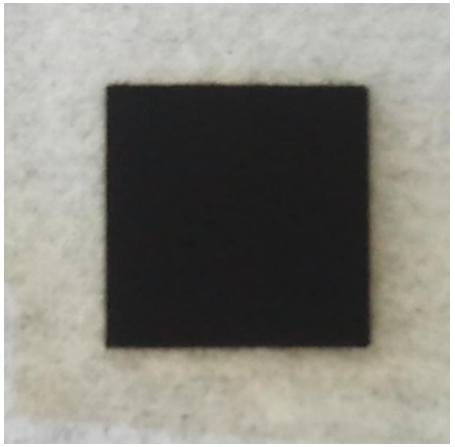
    Second order Lagrange elements

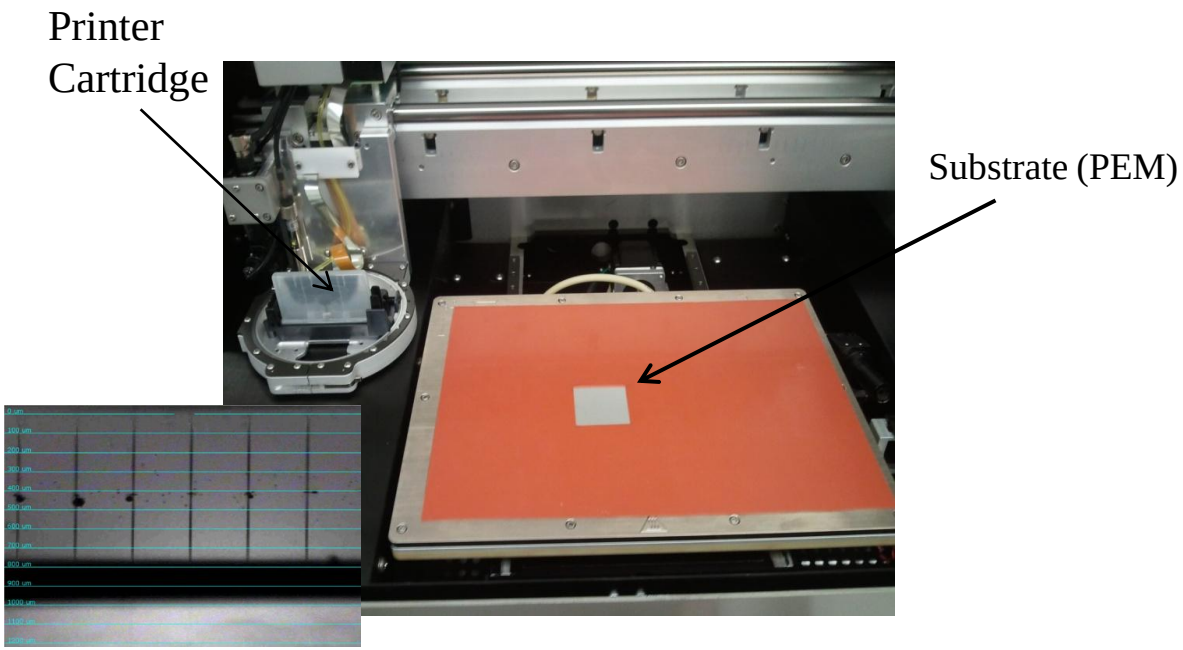
  - Update the solution u
- Adaptive grid refinement

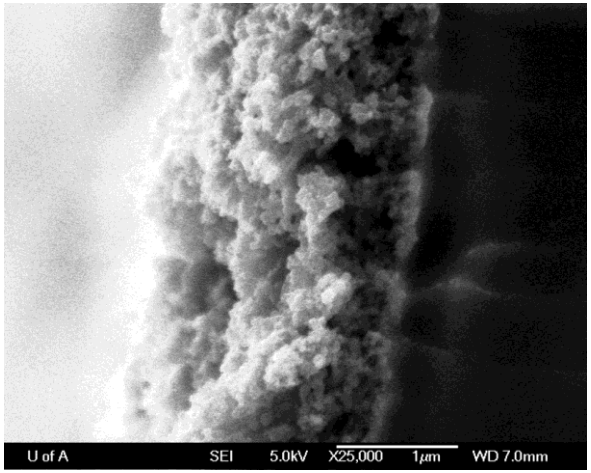


Results for conventional and ultra-thin electrodes

- Used inkjet printing to fabricate ultra-thin electrodes with reduced catalyst loading (0.025mg/cm^2 vs. 0.4mg/cm^2)
- Tested electrodes and compared performance to conventional electrodes



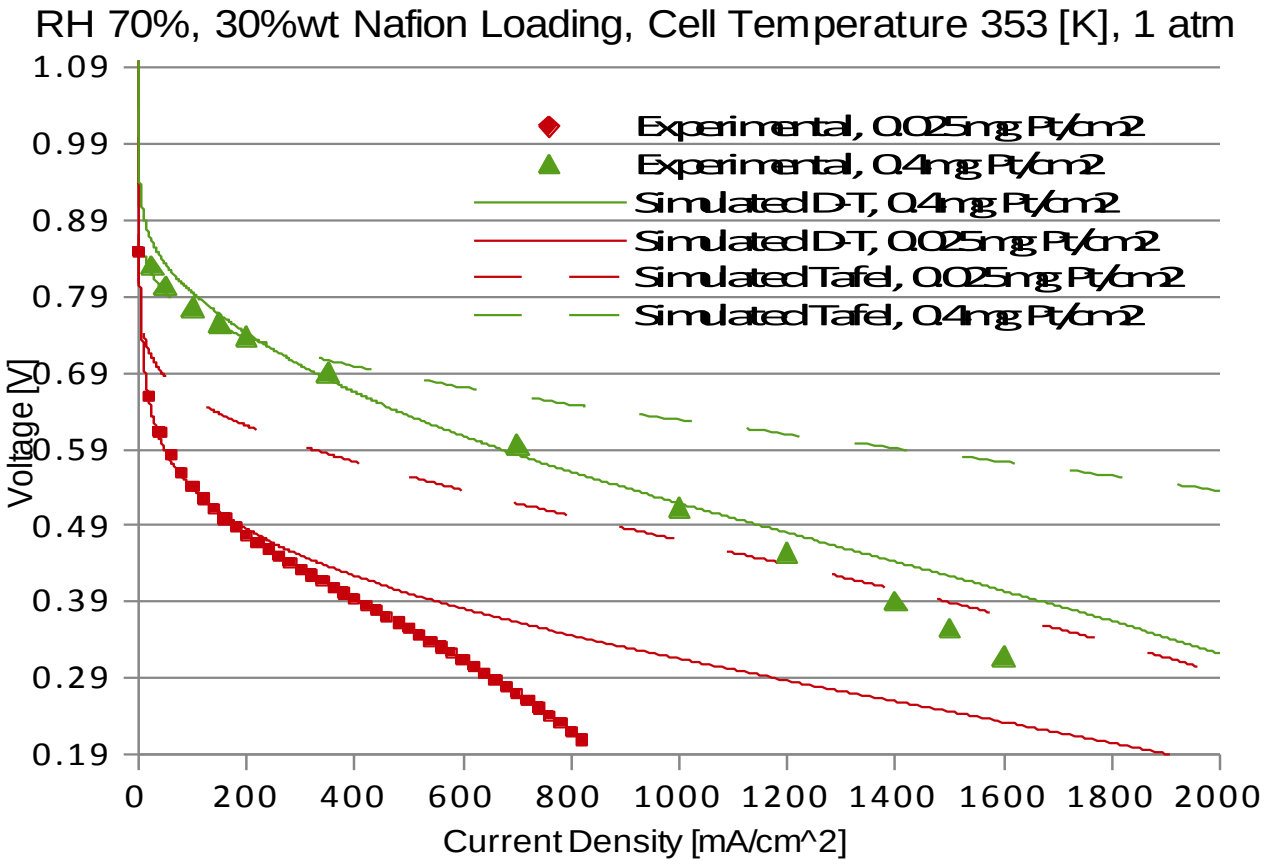




Ink-jet printed Catalyst Coated Membrane (NRE-211)

Results for conventional and ultra-thin electrodes

- Fuel cell models based on semi-empirical reaction models over predict performance for thin electrodes
- Current model is able to predict voltage losses accurately



Case study 2:
Multi-component gas transport model

Motivation

- The classic approach, i.e. “Navier-Stokes + Advection-Diffusion equations”, is only valid if the amount of ”a tracer” in the mixture is small enough such that it does not significantly perturb the flow of the main stream
  - In fuel cells, we have air (O_2 and N_2) and water vapour. At about 1atm and 80-100°C, the vapour, usually a tracer, can be a large part of the molecules in the mixture

- The classic approach includes the coefficients for mixture which are difficult to estimate.

Objective

- Kerkhof and Geboers [1] obtained a new mass and momentum balance for multi-component mixtures as an asymptotic expansion of the Boltzmann equation
- For the non-equilibrium trial functions, the velocity distributions are centered around the averaged velocities of the individual species
  - To date the velocities have been centered around the mass-averaged velocity of the mixture
- The new approach deals with simultaneous equations governing the transport of each individual species and utilizes the pure transport coefficients

[1] Kerkhof and Geboers, Toward a Unified Theory of Isotropic Molecular Transport Phenomena, AIChE Journal, 51(1), p.79-121, January 2005.

Governing equations

- New governing equations

$$\forall i = 1, N :$$

$$\nabla \cdot \mathbf{F}_{mass_i} = 0 \quad \text{in} \quad \Omega$$

$$\nabla \cdot \hat{\mathbf{F}}_{mom_i} = \nabla \cdot \hat{\boldsymbol{\sigma}}_i + (\mathbf{F}_i + \mathbf{D}_i + \rho_i \mathbf{g}) \quad \text{in} \quad \Omega$$

where

$$\mathbf{F}_{mass_i} = \frac{1}{\epsilon} \rho_i \mathbf{u}_i$$

$$\hat{\mathbf{F}}_{mom_i} = \frac{1}{\epsilon^2} \rho_i \mathbf{u}_i \otimes \mathbf{u}_i$$

$$p_i = \rho_i \frac{R}{M_i} T^{\text{const}}_{\text{mixture}}$$

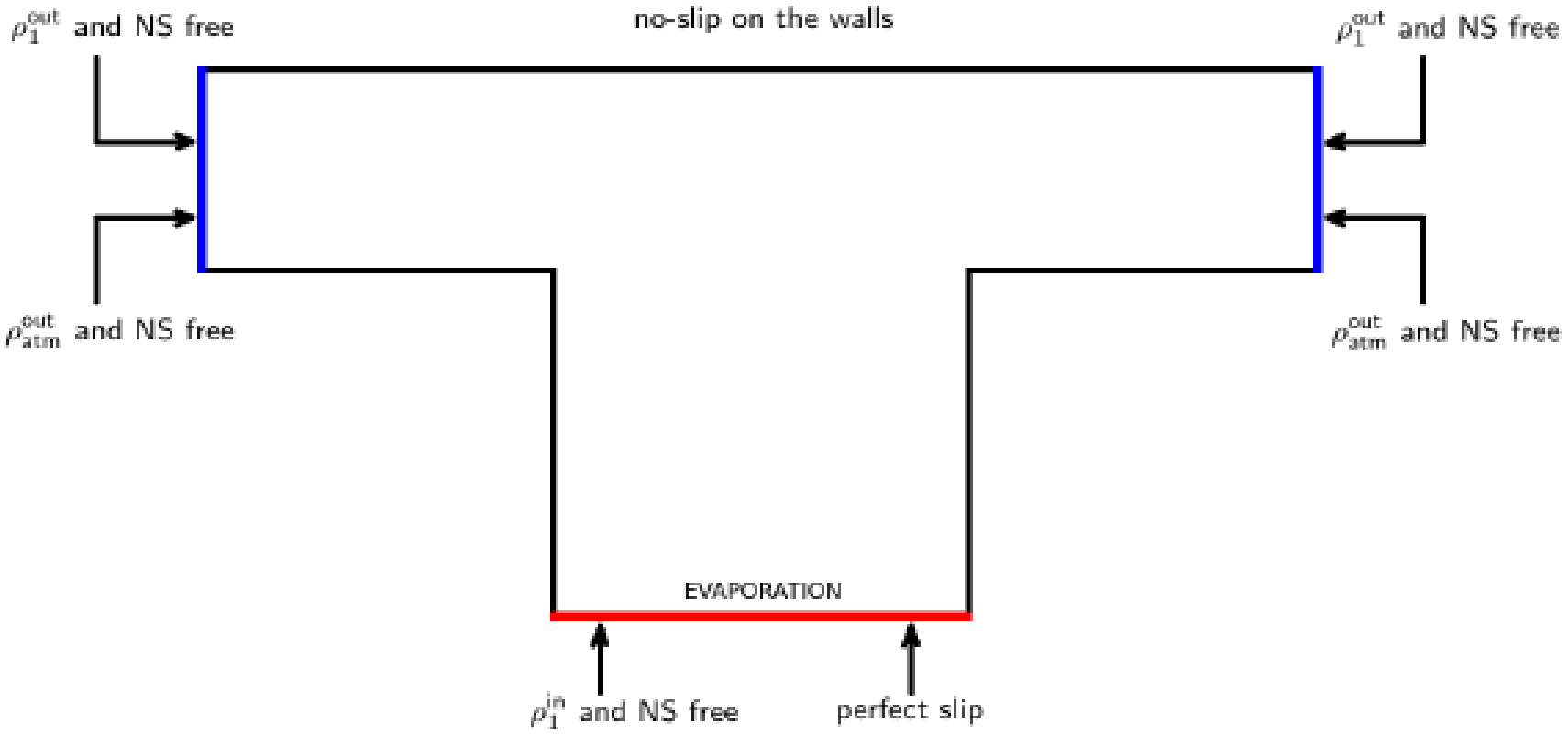
$$\hat{\boldsymbol{\sigma}}_i = -p_i \hat{\mathbf{I}} + 2\mu_i \nabla_s \mathbf{u}_i + \lambda_i (\nabla \cdot \mathbf{u}_i) \hat{\mathbf{I}}$$

$$\mathbf{F}_i = \begin{cases} \mathbf{0} & \text{in} \quad \Omega_c \\ -\mu_i \hat{\mathbf{K}}^{-1} \mathbf{u}_i - C_{\mathsf{F}_i} \hat{\mathbf{K}}^{-1/2} \rho_i \left| \mathbf{u}_i \right| \mathbf{u}_i & \text{in} \quad \Omega_p \end{cases}$$

$$\mathbf{D}_i = \sum_{j=1}^{N} p_i p_j \hat{\mathbf{D}}_{ij}^{-1} (\mathbf{u}_j - \mathbf{u}_i)$$

$$\hat{\mathbf{D}}_{ij}^{-1} = \begin{cases} \left(\sum_{l=1}^{N} p_l \cdot \mathscr{D}_{ij} \right)^{-1} \hat{\mathbf{I}} & \text{in} \quad \Omega_c \\ \left(\epsilon \sum_{l=1}^{N} p_l \cdot \mathscr{D}_{ij} \right)^{-1} \hat{\mathbf{T}} & \text{in} \quad \Omega_p \end{cases}$$

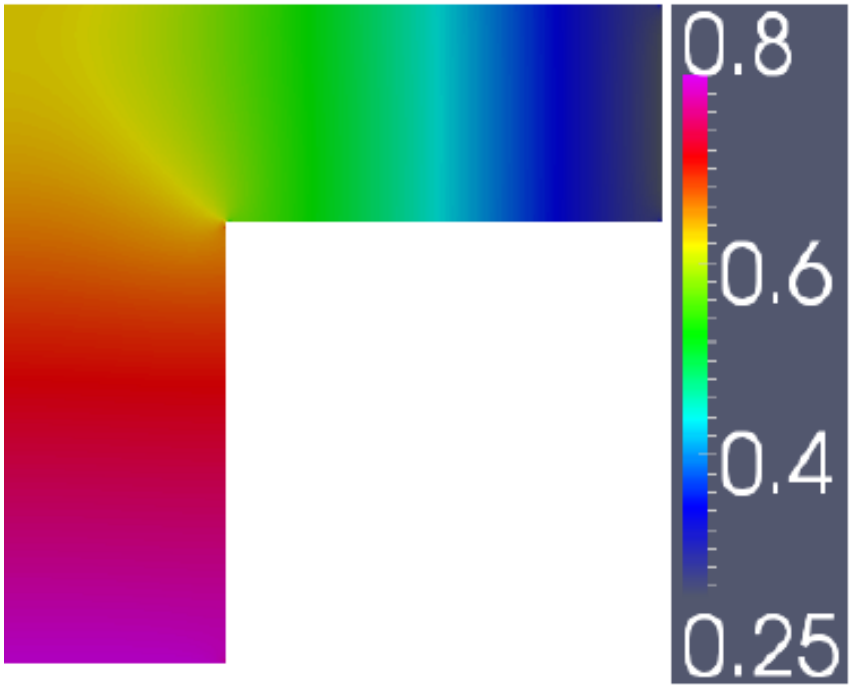
Stefan tube: Boundary conditions



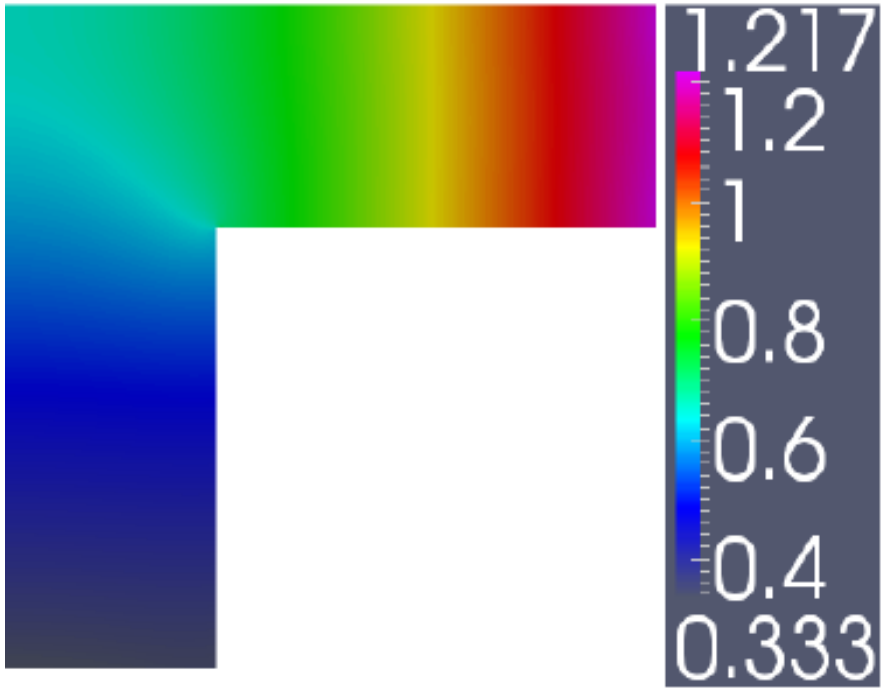
Stefan tube: Density preliminary results

- Species 1 diffuses through the tube
- Species 2, assumed stagnant in most studies, is displaced by spices 1

Density species 1



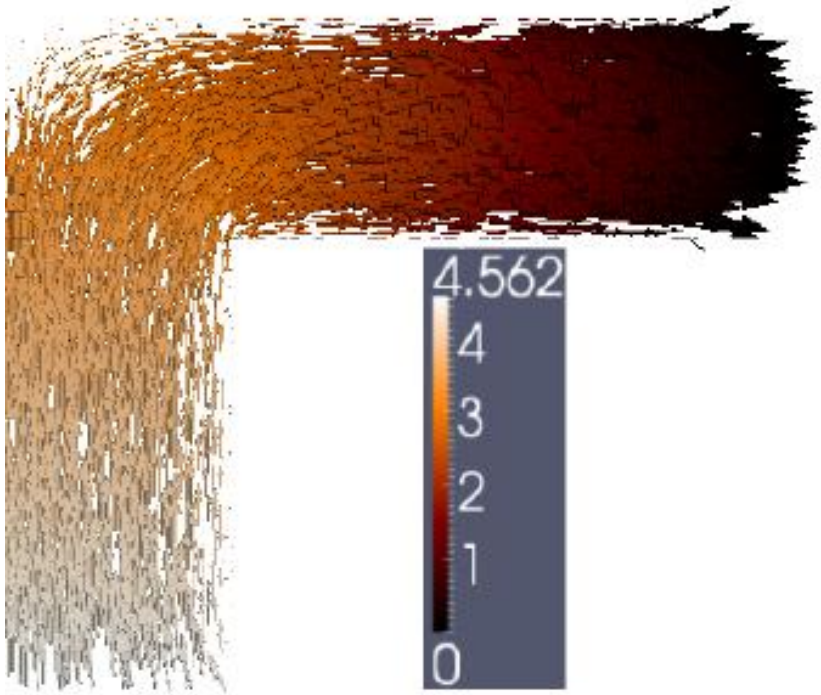
Density species 2



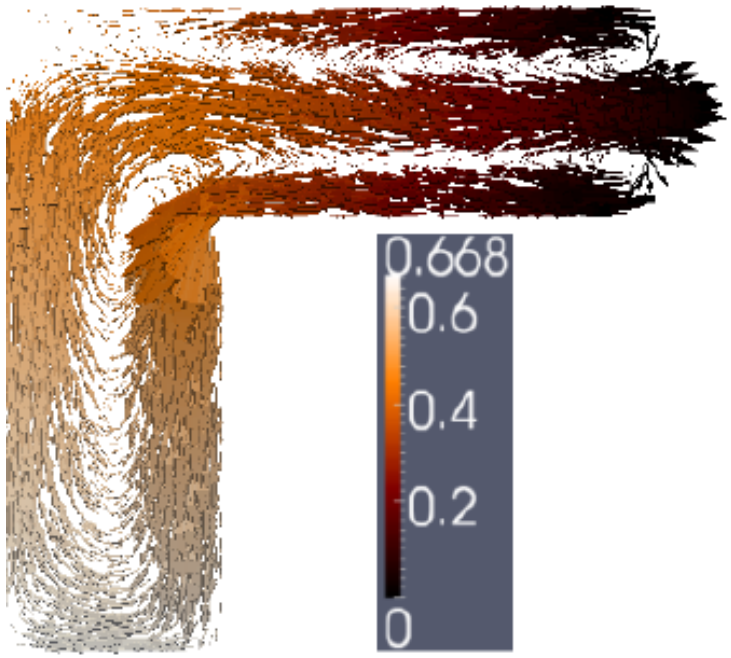
Stefan tube: Velocity preliminary results

- Species 1 is flowing out of the Stefan tube
- Species 2 is recirculating inside the tube. Some of it also leaving due to species 1.

Velocity species 1



Velocity species 2



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  - Membrane electrode assembly model with multi-step reaction kinetics
  - Gas transport model in porous media
  - Mulit-component gas transport model
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Conclusions

- An open-source fuel cell framework was developed using the deal.II libraries and Dakota
- The framework is developed to be able to allow both users and developers to take advantage of the software
  - For users:
    - Interface to open-source pre- and post-processors
    - Text (and soon XML/GUI) based selection of available database objects
  - For developers:
    - Clear and well documented interface to develop new objects of any type, i.e. equations, layers or materials.
- The framework has been successfully applied to analyzing to new fuel cell mathematical models for electrochemical reactions and mass transport

Acknowledgement

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Collaborators:
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Dr. Karan, UofCalgary

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NSERC CRSNG

Alberta Enterprise and Advanced Education

AUTOMOTIVE FUEL CELL COOPERATION

HYDROGENICS
SHIFT POWER | ENERGIZE YOUR WORLD

CaRPE FC
Catalysis Research for Polymer Electrolyte Fuel Cell

CANADA SCHOOL OF ENERGY AND ENVIRONMENT
ÉCOLE DE L'ÉNERGIE ET DE L'ENVIRONNEMENT DU CANADA
Canada

ESDL
ENERGY SYSTEMS DESIGN LABORATORY

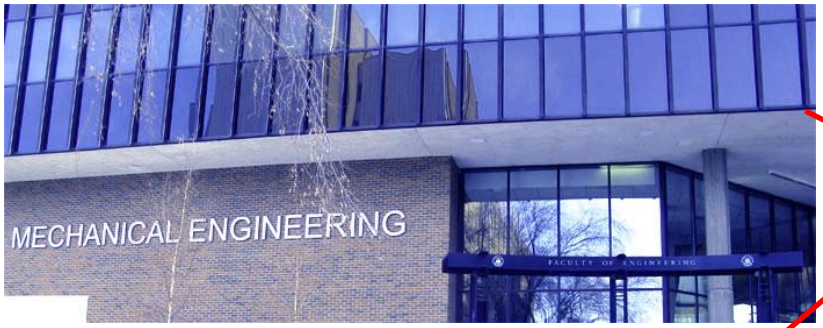
UNIVERSITY OF ALBERTA

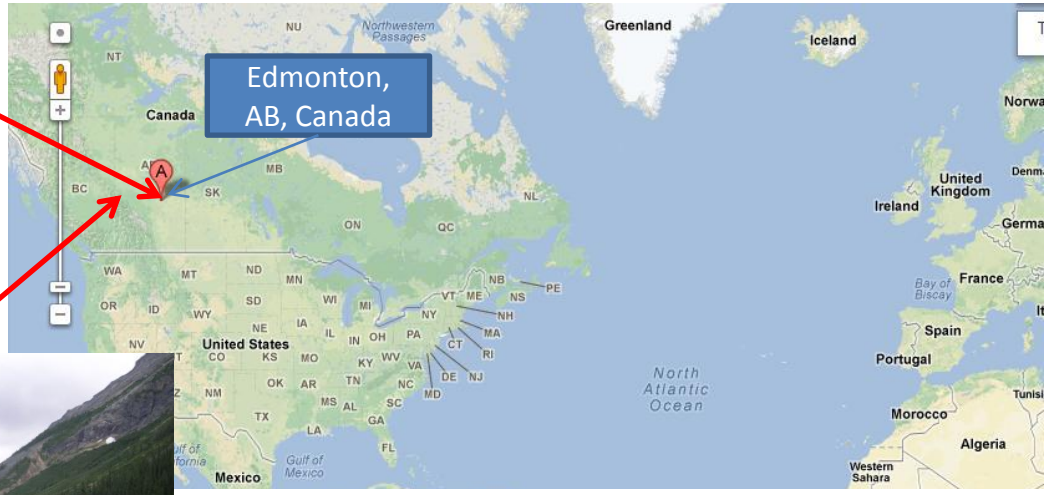
ENGINEERING U of A

THANK YOU

Energy Systems Design Laboratory (ESDLab)

- University of Alberta is located in Edmonton, Alberta
  - One of the top 5 Canadian universities with over 35,000 students (both graduate and undergraduate)
  - Mechanical engineering has over 950 students (700 undergraduate and 250 graduate)

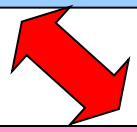




Energy Systems Design Laboratory (ESDLab)

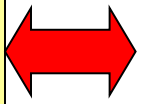
Computational Design and Optimization of Energy Systems

- Multi-disciplinary design optimization and multi-objective optimization
- Fuel cell and fuel cell system analysis and design
- Remote fuel cell power systems
- Flywheel analysis and design
- Hydrogen production systems



Computational Analysis of Energy Systems

- OpenSource PEM fuel cell analysis framework
- Two-phase flow analysis
- Multi-component mass transport analysis in porous media
- Multi-scale modeling in porous media
- Chemical and electrochemical reactions analysis



Experimental Testing of Energy Systems

- Ultra-low platinum loading fuel cell fabrication
- Polymer electrolyte fuel cell fabrication and testing
- Porous material characterization
- Measurements of gas and liquid transport in porous materials

Experimental Facilities

- *Wet laboratory for catalyst layer fabrication*
  - Ultrasonication bath and homogenisers
  - Hot press for decal transfer
  - Automatic film coat applicators
  - Material inkjet printer for CL deposition
  - Access to SEM, TEM, Microfab lab
- *Porous media characterization*
  - *M*ercury/non-mercury intrusion porosimetry
  - Permeability and effective oxygen diffusivity determination setup
  - Liquid permeation and water porosimetry
- *Fuel cell in-situ and ex-situ testing*
  - Fuel cell assembly facilities
  - Fuel cell testing system
  - Potentiostat/Galvanostat
- *Other*
  - Environmental chamber
  - Access to high performance computing