SOFC Model Development and Validation at NETL

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National Energy Technology Laboratory
Goals and Objectives

• SECA Program Goal
  – The Solid State Energy Conversion Alliance (SECA) goal is to create solid oxide fuel cells (3-10 kW) that can be mass produced in modular form
  – Low cost (<$400/kW) and high efficiency SOFC systems for stationary power, transportation, and military applications.

• Project Goals
  – Develop validated modeling tools capable of detailed fuel cell and stack performance analysis
  – Apply these new tools toward development of new/novel fuel cell concepts that can achieve the SECA goals
Technical Challenges

• Complex physics of the fuel cell
  – electrochemical sources and sinks of species and energy
  – chemistry for methane reforming, water-gas shift, catalytic surfaces
  – porous media flow
  – complex geometry, both in the cell and in the stack

• Numerical difficulties in coupled system
  – “stiff” coupling between electrical field and electrochemistry
Technical Challenges - continued

• Ensuring a productive tool given the computational requirements for modeling geometrically-complex cells and FC stacks
  – parallel implementation is needed
  – possible use of simplified submodels for very large scale simulations

• Accurate material properties for the model
  – accurate thermal, electrical, mechanical, and transport properties are needed
Technical Challenges - continued

- Validation Data
  - Much data is proprietary
  - Detailed data is needed for validation
    - local temperature, voltage, current, species, stresses
  - Measurements are difficult to make in SOFC environment
Technical Approach

• Use commercial CFD code as underlying platform for detailed fuel cell model
  
  – FLUENT code is parallel, unstructured mesh, with well-validated models for fluid flow, heat transfer, species transport

  – Working with Fluent Software Developers and Fuel Cell Experts

  – Model will be in public domain
Technical Approach

- NETL SOFC Model Capabilities - Existing, Planned
  - $\text{H}_2$ and CO Electrochemistry
  - Electrical field in conducting regions - current flow, ohmic heat generation
  - Contact resistance for cell/stack components
  - Species diffusion in flow channels and porous media
  - Water-gas shift reaction
  - Internal reforming
  - Parallel Processing
  - Single cells and cell stacks
  - Output compatible with ANSYS
  - Steady and transient analysis
Technical Approach - continued

• Validate NETL SOFC Model
  – NETL Test Facility (SOFCEL) for detailed validation data

NETL SOFC Test Stand
- 300 watt max
- pressure capable
- acquire test cells from partners with range of fundamental properties (component thickness, porosity, etc.)
Technical Approach - continued

• Use temperature field from SOFC model in an FEM model to calculate thermal stresses
  – geometry and temperature field output from SOFC model can be input directly into ANSYS
  – Work closely with ANSYS to make the coupling process easier

Stress in single SOFC cell at 1073K From NETL Model
Technical Approach - continued

• Parallel implementation of the SOFC model for complex cell and stack simulation
  – NETL Computation and Visualization Resources
    • 272 CPUs in 3 PC-Clusters
    • FLUENT license for NETL On-site use for all CPUs
    • 4 terabytes of RAID storage
    • Advanced visualization facilities
    • Multi-wall visualization environment for 3-D visualization
    • Access to Pittsburgh Supercomputer Center Resources
Technical Approach - continued

- Collaborate with other modeling groups and FC developers for model development and validation
  - National Labs
  - SECA participants
  - Other developers
  - Academia

- Publish to disseminate information and for peer review
NETL Fuel Cell Model

Local Species Concentration
Local Temperature
Local Electrical Potential

FLUENT CFD
- Species
- Momentum
- Energy
- Electrical Potential
- Field

Species Source/Sink
Heat Fluxes
Electrical Potential B.C.s

SOFC Model
- Nernst Voltage
- Losses at Electrolyte
- Current Distribution on Electrolyte
- Electrical Potential B.C.s
Model Overview: Physics for Detailed Model

- **Fluid Flow, Heat Transfer, Mass Transfer**
  - Complex Flow Geometry
  - Laminar Flow
    - diffusion is important
  - Porous Media
    - flow and diffusion in porous media

- **Electrical Potential Field**
  - Coupled to Electrochemical Model

- **Electrochemical Submodel**
  - Electrolyte Submodel
  - Electrochemical Losses
  - Stack Logic
Model Overview: Species Diffusion

- No detail today on mass, momentum, energy conservation equations and solution - “standard” FLUENT implementation

- The diffusion coefficient for binary gas mixtures is given by the following equation

\[
\frac{PD_{AB}}{(p_{cA} p_{cB})^{1/3} (T_{cA} T_{cB})^{5/12} \left( \frac{1}{M_A} + \frac{1}{M_B} \right)^{1/2}} = a \left( \frac{T}{\sqrt{T_{cA} T_{cB}}} \right)^b \times 1E-4
\]

- The diffusion coefficient of species \( i \) in the mixture is given by:

\[
D_{i,m} = \frac{1 - x_i}{\sum_{j \neq m} \left( \frac{x_j}{D_{ij}} \right)}
\]

- And effective diffusion coefficient through porous media is given by:

\[
(D_{i,m})_{\text{effective}} = p \cdot D_{i,m}
\]

– where \( p \) is the permeation factor = porosity/tortuosity
Model Overview: Electrical Potential Field

- Submodel solves for the electric potential field throughout all conductive regions
  - porous electrodes, current collectors, interconnects

- Local voltage losses

- Distributed heat generation
  - local ohmic heating in anode, cathode, current collectors, and interconnect regions
Model Overview: Electrical Potential Field

- Basis for the electrical potential equation is obtained from the statement of conservation of charge for steady conditions:

\[ \nabla \cdot \vec{i} = \rho_i \]

- where \( \vec{i} \) is the local current density vector and \( \rho_i \) is the local current source
- for the potential fields we are considering, no local sources of current will be present, so \( \rho_i = 0 \)
- current sources/sinks at the electrolyte-electrode faces are introduced through boundary conditions

- For ohmic materials, the current density vector may be expressed in terms of the gradient of the electrical potential field:

\[ \vec{i} = -\sigma \nabla \phi \]

- where \( \sigma \) is the conductivity of the material and \( \phi \) is the electrical potential
Model Overview: Electrical Potential Field

- The FLUENT capability of solving for transport of a User-Defined Scalar is used where the scalar is electrical potential $\phi$.

- Ohmic heating in the conducting regions can then be calculated from the relation:

$$Q_{genC} = \sigma |\nabla \phi|^2$$

- Boundary conditions are specified as fixed potential on one side and fixed flux on the other.

- This guarantees that the problem is well-posed and that the total current is conserved.
Model Overview: Electrolyte Model

- The electrolyte is always assumed thin for electrochemical modeling purposes
  - It can be represented by a finite thickness region in the FLUENT simulation

- The electrodes can be resolved as porous media zones to capture concentration gradients directly
  - Fickian-based overpotential models are also included to allow the treatment of thin electrodes that are not resolved with mesh

- Models for activation losses are also available for each electrode
  - account for electrochemical kinetics at the electrodes
Model Overview: Electrolyte Model

- Anodic and cathodic faces are associated pair-wise
  - species and temperature information give the local Nernst potential and diffusion and activation overpotentials

- For simple geometries, pair-wise association is not a problem. This is not necessarily true for more complicated designs. This is an area for future development

- The total cell current is specified as user input
Model Overview: Electrolyte Model

- The electrolyte is treated as a parallel resistor-voltage network for n computational cell face pairs on either side of electrolyte.
- Local currents, $I_n$ through each face pair must sum to equal the specified cell current: $I_T = \sum_n I_n$.

Electrolyte

Electrolyte - Anode Face

Electrolyte - Cathode Face

$V_n = \text{Nernst Potential - Overpotentials}$

$R_n = \text{Local Lumped Resistance (ohmic + electrochemical losses)}$
Model Overview: Electrolyte Model

- Corrections to $V_{\text{Nernst}}$ for losses - “Overpotentials”

$$V_n = V_{\text{Nernst}} - \eta_R - \eta_{C,\text{Cath}} - \eta_{C,\text{Anode}} - \eta_{A,\text{Cath}} - \eta_{A,\text{Anode}}$$

Ohmic Loss through Electrolyte:

$$\eta_R = R \cdot i$$

Diffusion Losses through Electrodes:

$$\eta_C = -\frac{R_u T}{nF} \ln\left(1 - \frac{i}{i_L}\right)$$

Chemical Kinetics of Electrode Reactions:

$$i = i_o \left[ \exp\left(\alpha \eta_A F \frac{R_u T}{R T}\right) - \exp\left(\left(1 - \alpha\right) \eta_A F \frac{R_u T}{R T}\right) \right]$$

- where $R$ is the resistance [ohm-m²] of the electrolyte; $i$, local current density; $i_L$, is the diffusion limiting current density; $\alpha$ is the transfer coefficient; and $i_o$ is the exchange current density

- These corrections are made to $V_{\text{Nernst}}$ for each electrolyte face-pair to produce the “local” voltage across the electrolyte
Model Overview: Electrolyte Model

- Using the local current information, the model applies species fluxes to the electrode-electrolyte boundaries in the FLUENT simulation.

- Local current information is also used to account for heat generation in the electrolyte and electrodes as follows:

\[
\Delta \dot{Q}_{\text{total}} \left( \frac{J}{s} \right) = \Delta \dot{Q}_{\text{rev}} + \Delta \dot{Q}_{\text{irrev}} = T \Delta S \cdot \frac{I}{nF} + I^2 R_E + I \eta_{\text{total}}
\]

- Heat generation is calculated for each electrolyte face-pair.
Model Overview: Boundary Conditions

- Specify Uniform Current Density calculated from specified cell current and area
- Specify Uniform Voltage
FY 02 Accomplishments

- **Apply Model to New Fuel Cell Geometry Concepts**
  - Study Delphi proprietary cell geometry
  - Evaluate model in tubular geometry

- **Apply model to NETL Test Facility Designs**
  - Assist in design of NETL test rig
  - Initial investigation of effect of test parameters on cell performance

- **Preliminary Validation with University of Utah Data**
FY 02 Accomplishments

• Compare NETL and PNNL SOFC Models

• Technology Transfer
  – MOU with Siemens-Westinghouse
  – Draft MOU with GE Power Systems

• Developed Stack model
  – “beta” version
Validation Application: Button Cell

- Preliminary Validation with University of Utah Data
  - UU has tested cells and supplied representative performance data

![Graph showing Voltage, Power Density vs Current Density](image)

**NETL Standard Cell**
- Anode Thickness: 1mm
- Temperature: 800°C
Validation Application: Button Cell

- Validate models with experimental data
  - Preliminary Model Validation using University of Utah Data
  - Simulate button cell performance in NETL test rig - compare to U of U data
Validation Application: Button Cell
Validation Application: Button Cell

- Cathode: 50 μm
- Cathode Interlayer: 20 μm
- Electrolyte: 10 μm
- Anode Interlayer: 20 μm
- Anode: 1 mm
- Anode Current Collector: 127 μm
- Cathode Current Collector: 127 μm
Validation Application: Button Cell

Cell Type: Standard Cell, 1mm Anode
Average Current Density: 1A/cm²
10,000A/m²
Cell Temperature: 800°C / 1073K

Current Density on Electrolyte-Anode Face

Current Density (A/m²)

10075.9
10071.5
10067.2
10062.8
10058.4
10054.0
10049.6
10045.2
10040.9
10036.5
10032.1
10027.7
10023.3
10018.9
10014.6
10010.2
10005.8
10001.4
9997.0
9992.6
9988.3
9983.9
9979.5
9975.1
9970.7
9966.4
Validation Application: Button Cell

Cell Type: Standard Cell, 1mm Anode
Average Current Density: 1A/cm²
Cell Temperature: 800°C / 1073K

Cathode Side Temperature

Anode Side Temperature
Validation Application: Button Cell

Cell Type: Standard Cell, 1mm Anode
Average Current Density: 1A/cm²
Cell Temperature: 800°C / 1073K
Validation Application: Button Cell

- Validate models with experimental data
- Data from tests by Virkar et al., May 2002

![Graph showing measured voltage and power density against current density for a Standard Cell: 800C. The graph includes data points for both voltage and power density, illustrating the relationship between current density and power output.]
Validation Application: Button Cell

Standard Cell: 800C

- Measured Voltage
- Predicted Voltage
- Measured Power Density
- Predicted Power Density

Current Density (A/cm²)

Average Cell Voltage (V)

Power Density (W/cm²)
Compare NETL and PNNL Models

- Comparison of NETL SOFC model with PNNL SOFC model was performed
  - Proprietary cell geometry was used for comparison
  - Qualitative agreement was good between the models
  - Models give consistent predictions for planar geometry
Compare NETL and PNNL Models

- **$O_2$ Concentration at electrolyte face**
- **$H_2$O Concentration at electrolyte face**
- **$H_2$ Concentration at electrolyte face**
- **Current Density at electrolyte face**
Model Overview: Extend to Stack

Specify Uniform Voltage

Specify Uniform Current Density calculated from specified cell current and area
Model Overview: Extend to Stack

- Fuel Inlet
- Fuel Outlet
- Air Inlet
- Air Outlet

10 Cell Stack
- Average Current Density: 5000 A/m² (0.5A/cm²)
- Cell Voltage: ~0.5V/cell
- H2 Utilization: 80%
- O2 Utilization: 20%
- 80 W stack
Model Overview: Extend to Stack

Cell based on U of U 10cmx10cm Cell

- Anode Current Collector: Nickel Mesh
- Anode Seal: Mica
- Interconnect: Stainless Steel

- Cathode Current Collector: Silver Mesh
- Cathode Seal: Mica
- Interconnect: Stainless Steel

PEN Assembly

Cell diagram showing layers and materials.
Model Overview: Extend to Stack

Electrolyte Current Density (Amp/m²)
Model Overview: Extend to Stack

- Voltage Field on Bottom Cell (V)
- Voltage Field (V)
Model Overview: Extend to Stack

H₂ Mole Fraction

O₂ Mole Fraction
Collaborations

• Collaborations to date
  – PNNL
    • Comparison of SOFC models
    • Core Technology Plans for Simulation and Modeling
  – Delphi
    • Application of NETL Model to SECA cell geometry
  – University of Utah
    • U of U supplying data and test cells for model validation
  – National Research Council Canada
    • Discussion for potential collaboration on modeling and visualization
FY 03 Work Plans

• NETL SOFCEL Test Facility
  – Facility Startup, Shakedown
  – Button Cell Tests
    • Vary anode, electrolyte, cathode thickness
    • Vary Anode Porosity
    • Measure Sensitivity of data to experimental parameters
  – 10cm x 10cm Cell Testing
FY 03 Work Plans

- Model Development Activities
  - Complete Parallel Version of the NETL Model
  - Add Internal Methane Reforming Chemistry
  - Add Water-Gas Shift
  - Complete Implementation of Stack Model
  - Implement Transient Capability
FY 03 Work Plans

• **Model Validation Activities**
  – Data from NETL Testing and Validation Facility
    • Button cell data
    • 10cm x 10cm cell data
    • Validation data over range of geometry, compositions, operating conditions
  – Data from University of Utah
  – Data from SECA developers
    • Planar data
    • Tube data
FY 03 Work Plans

- Apply the Model - collaborate with SECA development team members
  - Siemens-Westinghouse Power Corporation
  - GE Power Systems

- Link NETL SOFC Model with ANSYS
  - validation
  - application
FY 03 Work Plans

• Work with PNNL, ORNL under Core Technology Program
  – model development
  – material properties
  – technology transfer to SECA developers
  – SECA Core Technology

• Publish