Modeling Tools for SOFC Design and Analysis

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PNNL Modeling Activities: Objectives & Approach

- **Objectives**
  - Develop integrated modeling tools to:
    - Evaluate the tightly coupled multi-physical phenomena in SOFCs
    - Allow SOFC designers to perform numerical experiments for evaluation of stack electrochemical, thermal, and mechanical performance
    - Aid understanding of materials degradation issues
    - Provide wide applicability for industry teams' to solve their challenging design problems
  - Provide technical basis for stack design

- **Approach: Multiphysics-based analysis tools**
  - SOFC-MP: A multi-physics solver for computing the coupled flow-thermal-electrochemical response of multi-cell SOFC stacks
  - Distributed Electrochemistry (DEC) model – Cell level multi-physics model for considering the effects of local properties and conditions on global SOFC performance
  - Targeted evaluation tools for specific cell design challenges
  - Experimental support to provide material property data

PNNL Modeling Tools: Overview

- SOFC-MP
  - 2D and 3D multi-physics stack model
  - 2D model benchmarked with literature data
- Distributed Electrochemistry (DEC) Model
  - 3D multi-physics model of the SOFC electrodes and electrolyte for the investigation of SOFC performance and degradation issues
  - Degradation modeling framework
- Cathode Contact Paste Modeling
  - FEA model for the simulation of densification behavior in cathode contact materials
- Glass Seal Modeling
  - Investigate the behavior of glass seal materials and designs at operating temperatures
- Interconnect Modeling
  - Integrated modeling and experimental approach for prediction on interconnect lifetime

SOFC-MP:
Multi-Physics Stack Modeling Tool

3D Model

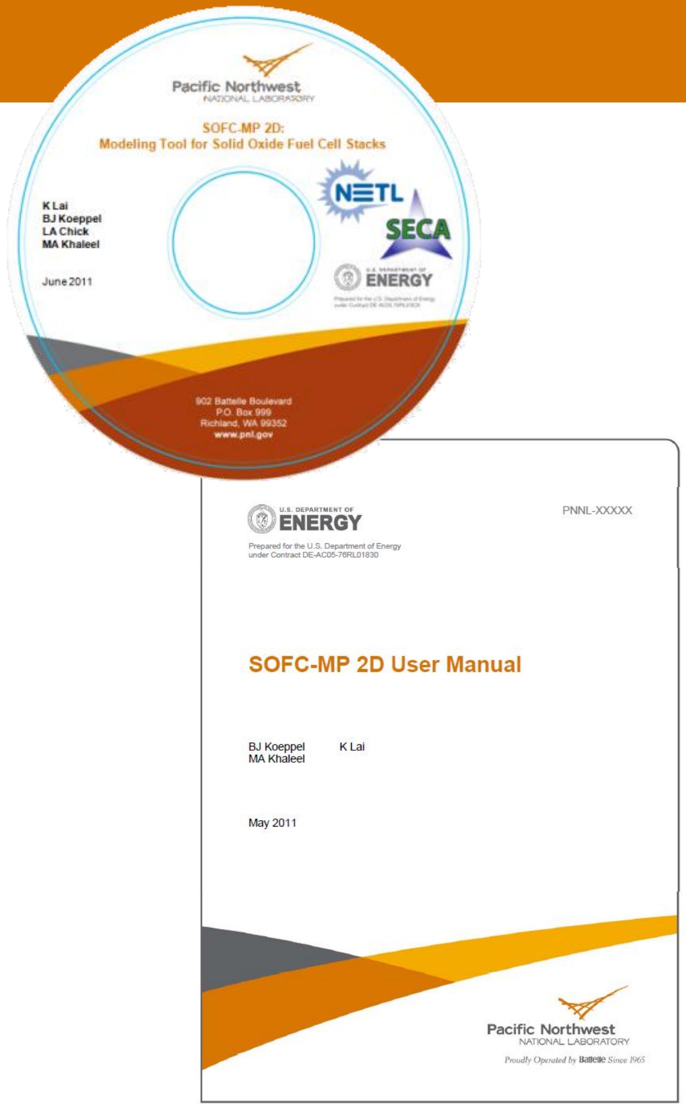
- Usage
  - Detailed 3D distribution for follow-up structural analysis
  - All planar flow configuration including cross-flow
- Computes distributions in entire 3D domain
  - All planar configurations: Co-flow, counter-flow, and Cross-flow
  - Multi-cell configuration (up to 50 cells)
- Computations
  - Current distribution
  - Voltage distribution
  - Thermal distribution
    - Used for FEA stress analysis
  - Species distribution
  - Heat losses

2D Model

- Usage
  - Tall cell stack
  - Fast computation
  - Can be integrated to system tools
- Computes distributions along the symmetric centerline of the stack
  - Co-flow and counter-flow
  - Multi-cell configuration (up to 1000 cells)
- Computations
  - Current distribution
  - Voltage distribution
  - Thermal distribution
  - Species distribution
  - Heat losses

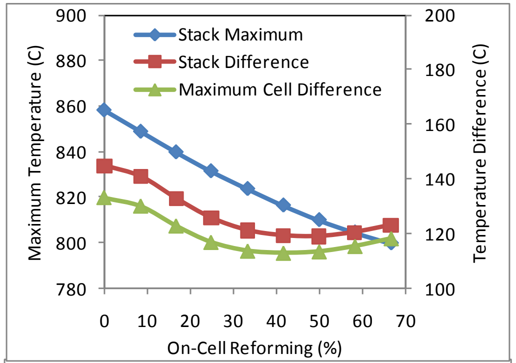
2D SOFC-MP Software Release

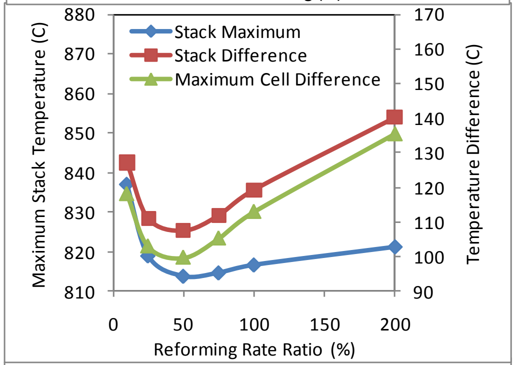
- Official 2D model released
  - NETL can distribute
- User manual for 2D model completed
  - Step-by-step instructions on installation, model simulations, solution, and post-processing procedures
  - Detailed descriptions of sample cases, including in depth explanation of parameters in the input files
- Includes feedback from users
  - Newer version with more robust and faster iteration scheme made available because of request from PNNL users providing vertical team modeling support
- Code usage demonstrated in parametric study on stack temperature uniformity
- * More information available at the poster session

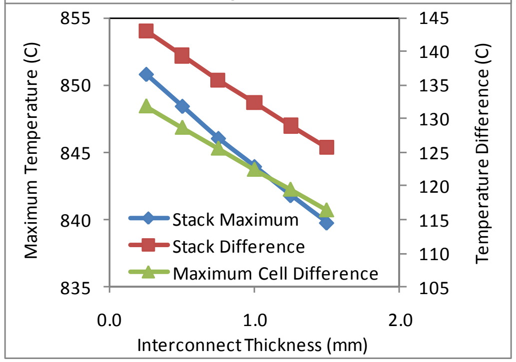


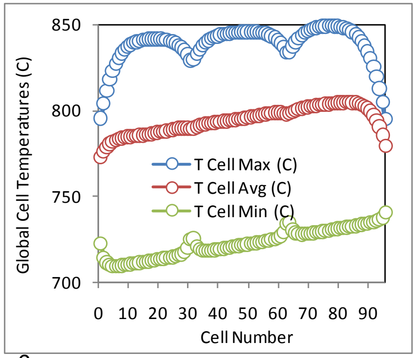
SOFC-MP Rich Features in 2D Model

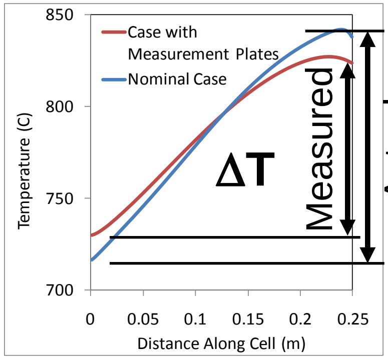
- The module can simulate different flow orientations, cell counts, cell sizes, boundary conditions, fuels, user-defined electrochemistry, reforming, and cell-to-cell variations
  - Effect of amount of on-cell reforming
  - Effect of on-cell reforming rate on temperature
  - Effect of interconnect thickness on temperature
  - Effect of local 25% fuel blockage on single cell (#32) current density
  - Effect of 2 instrumented measurement plates
    - -21% cell ΔT error from measurement plate

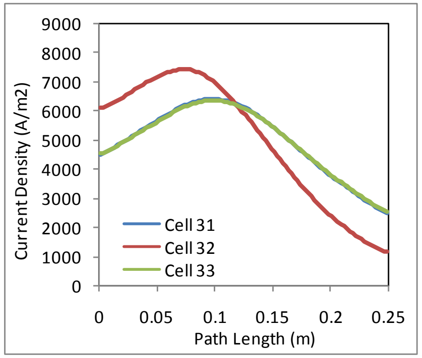












Distributed Electrochemistry (DEC) Model: Modeling SOFC Performance

Technology Challenges

- Understand degradation in the electrodes
  - Resolve local conditions in the cell resulting from various operating conditions
  - Investigate the effect of microstructure on cell performance
  - Confidently predict global cell performance for a range of conditions
- Increase performance by advanced electrode design

Objectives

- Develop a model to predict cell performance based on operating conditions and microstructure
  - Base the performance model on coupled electric potential, charge transfer, and reactive transport
  - Use a modeling approach that enables varying structural parameters
  - Validate the model by comparison with experimental data
  - Simulate microstructural and operational effects on cell performance

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DEC Model: Recent Accomplishments

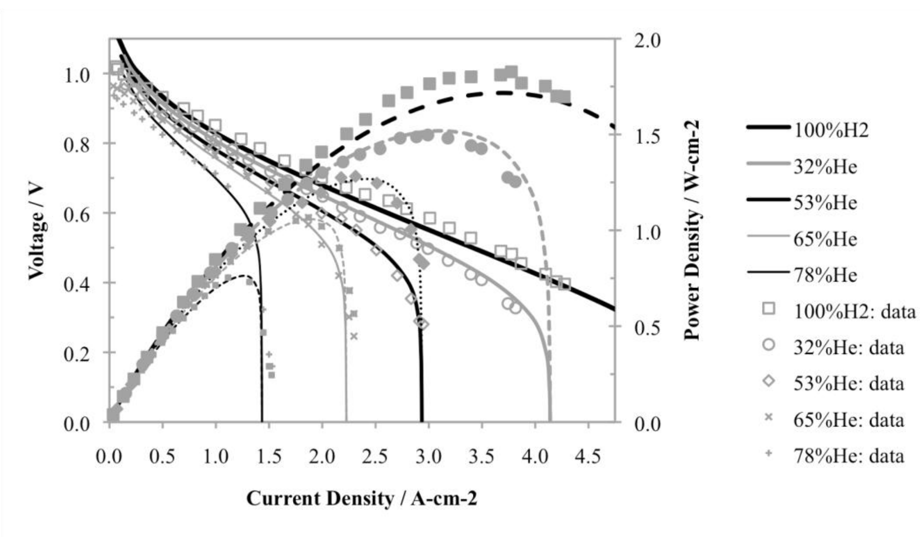
- Developed a 3D multi-physics model of the SOFC electrodes and electrolyte
  - Resolves the electrochemistry throughout the electrode thickness
  - Predicts the global SOFC performance based on local conditions within the electrodes
  - Includes electrode microstructures via an effective properties model
  - Allows for spatially varying microstructural and electrochemical properties
- Validated the DEC model with experimental button cell data at various fuel compositions and operating voltages
- Demonstrated the DEC model's capabilities to investigate the effects of electrode microstructure on SOFC performance
- Developed a degradation modeling framework for considering the effects of local degradation within the electrodes on the overall SOFC performance

- * More information available at poster session

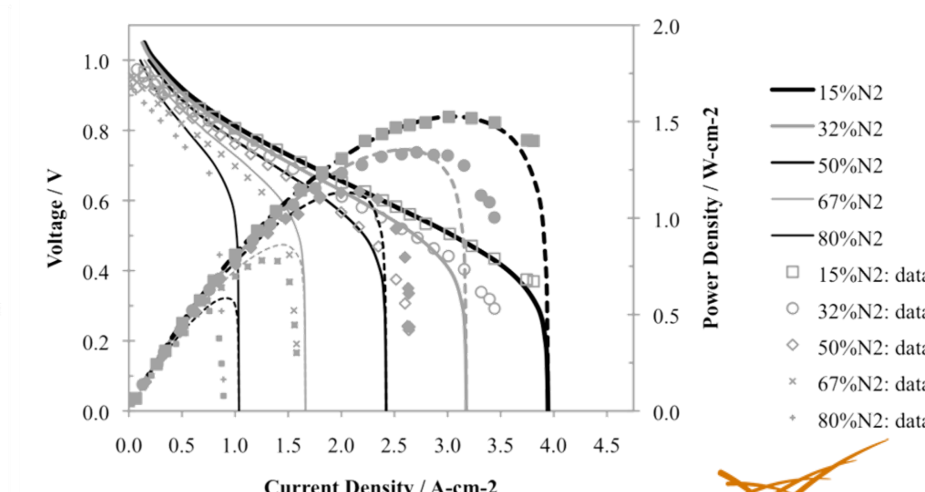
Validation of DEC Model by Comparison with Experimental Button Cell Data

- Experimental data set for binary fuel [Jiang and Virkar 2003]
  - DEC Model shows good agreement with He-H_2 and N_2-H_2 systems
- Highlights model sensitivity to gas diffusion in electrodes
- Good predictions of limiting currents and peak power

Binary Fuel	Maximum Difference: DEC model to Experimental Data	
	Peak Power	Limiting Current
He-H_2	7%	6.5%
N_2-H_2	12%	11%



He-H_2 fueling



N_2-H_2 fueling

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Degradation and Life Prediction of Coated Metallic Interconnects Summary

- Motivation: Ensure IC life meets the SECA life time requirement
- Goal: Use modeling to predict interconnect life with and without spinel coating under isothermal cooling and thermal cycling
- Technical Approach: Develop a combined modeling/experimental approach to enhance spallation resistance; use finite element based modeling tools to evaluate various design issues on spallation driving forces and determine the main factors influencing IC degradation in terms of spallation; and evaluate IC candidate materials

- Accomplishments:
  - Developed an integrated modeling and experimental approach for IC life prediction:
    - Identified and quantified spallation driving forces
    - Quantified interfacial strength
    - IC life prediction for coated and uncoated Crofer 22
    - Interfacial strength quantification for as-received and surface modified SS441

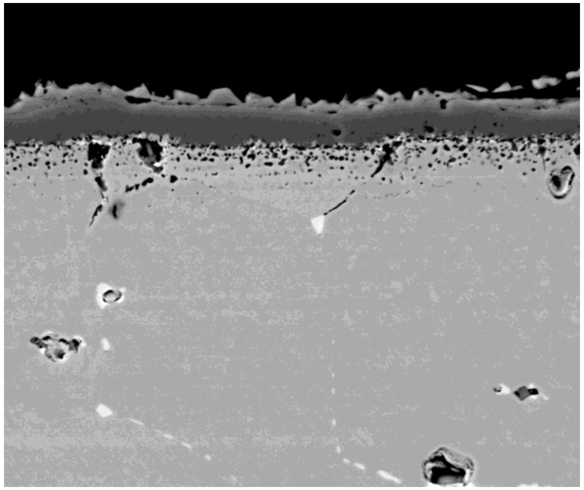
Doping Coating with Rare Earth Improves Spallation Resistance

- Model determined increase in interfacial strength between oxide scale and substrate when spinel coating was doped with Ce
  - Improvement of adhesion also observed experimentally (SECA CTP Materials team)

	Crofer 22 (0.5 mm thick)	**441 Substrate (1.5 mm thick)**		
	Bare	Bare	Mn-Co Spinel	Ce-doped Spinel
Coating thickness (µm)	NA	NA	15	14
Scale thickness (µm)	2.41	3.82	3.87	1.49
Strength (MPa)	**395**	**324**	**324**	**403**

Surface Modification Increases Adhesion Strength – Mechanical Polishing

- Initial surface modification studies began with the effects of mechanical polishing on scale spallation of bare specimens
- Model determined that surface quality influenced the interfacial strength
- Bear in mind: this technique increases both interfacial strength and spallation driving force
- Polishing substrate surface became a common practice prior to applying coating on specimens for experimental studies



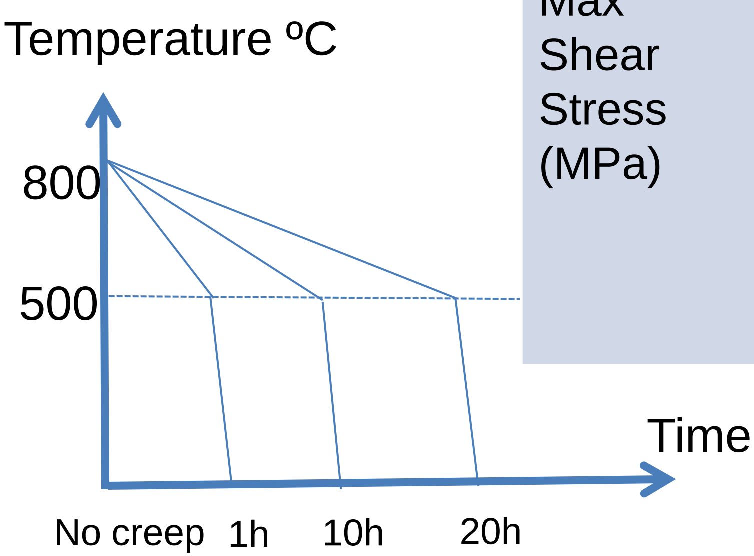
Surface modified (polished) specimen exposed to 850C for 900h.

	441 Substrate (1.5 mm thick)		
	As-received	Surface Modified	Surface Modified
Surface Roughness (Ra)	0.7	0.25	0.02
Scale thickness (µm)	3.82	3.87	5.61
Strength (MPa)	**324**	**394**	**384**

Optimizing Cooling Profile to Reduce Spallation Driving Force

- Optimization of cooling profile helps to reduce the interfacial stress, only to a certain extent
- Benefit plateaus to about 6.8% stress reduction

		500ºC		600ºC		No Creep
Max Shear Stress (MPa)	10min	362	2.1%			370
	1 h	357	3.5%			
	10 h	348.3	5.8%	347.9	6.1%	
	20 h	345.3	6.7%	345.2	6.7%	



Cooling profile schematic

Reducing IC Substrate Thickness to Reduce Driving Force

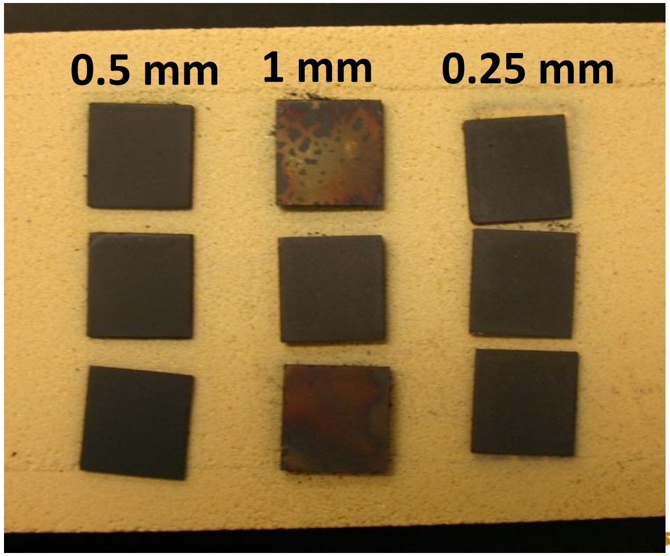
- Interfacial failure driving force can be reduced by reducing the bulk thickness of SS441
  - The thicker the substrate, the higher the driving force for spallation

Model Predictions

Substrate thickness	1.6 mm	0.5 mm
Coating thickness	10 um	10 um
Scale (um)	Scale/441	Scale/441
2	441 MPa	361 MPa
5	487 MPa	410 MPa
10	489 MPa	463 MPa
15	485 MPa	479 MPa

*Liu et al., *Journal of Power Sources* 189 (2009) 1044–1050

Experimental Validation



Results provided by Materials team

Contact Modeling Task Summary

- Motivation: Cathode contact layer is weak and must meet multiple design criteria
- Goal: Use modeling to understand *in situ* low temperature formation of the cathode contact layer and its influence on the stack's mechanical reliability
- Technical Approach: Develop FEA modeling approach to simulate the densification behavior of cathode contact materials and determine their influence on the stack thermal-mechanical stress state during formation

- Task Accomplishments:
  - Determined expected stress levels for contact layer in the cell
  - Verified reduced seal loads by load path modification
  - Implemented constitutive model for constrained sintering
  - Supported test cell development
  - Developed method to extract model input parameters from Task 1 material experiments
  - Simulated contact materials and effects of design parameters on densification in stacks

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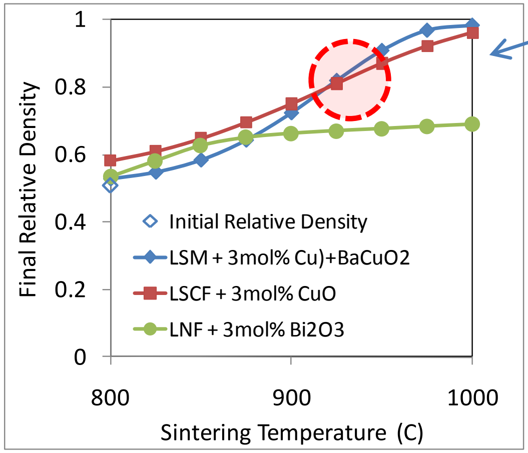
Contact Modeling: Realistic Multi-Cell Stack Geometry

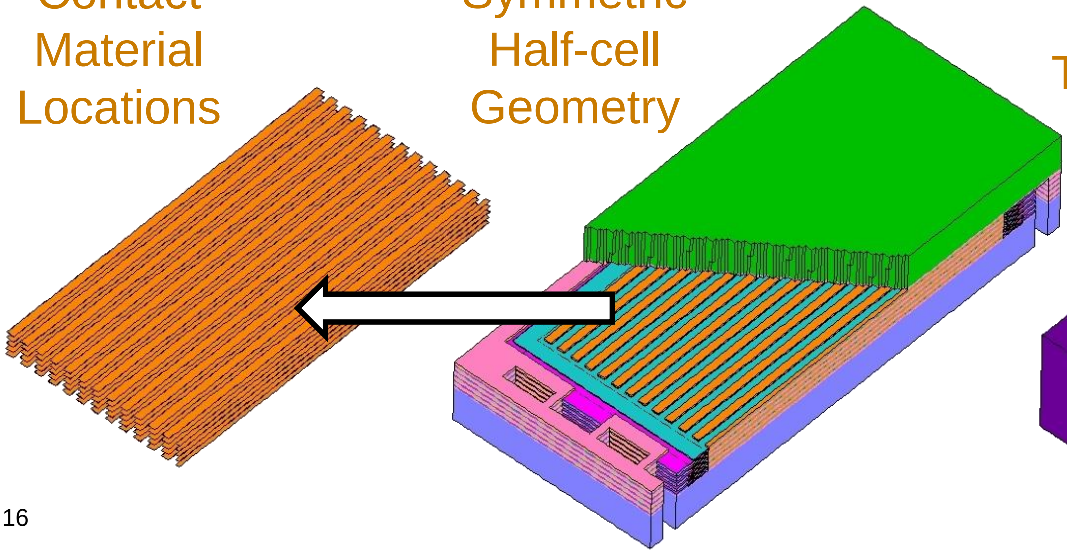
2-hr Free Sintering



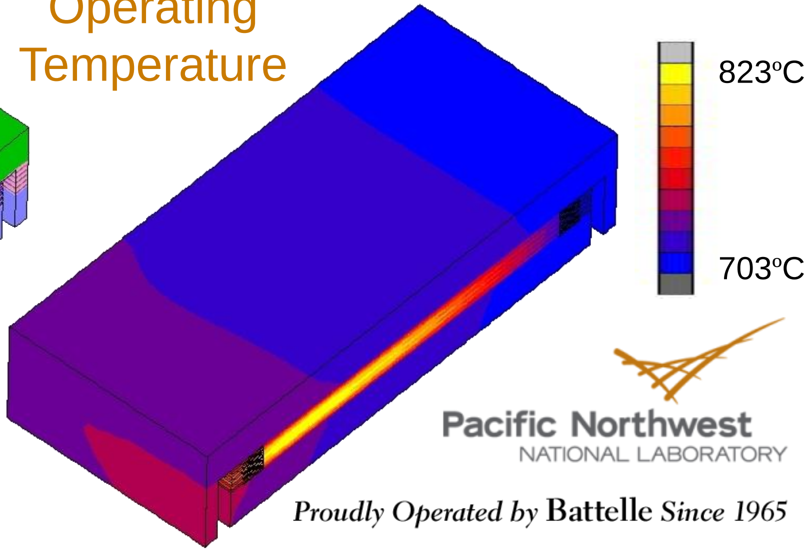
- LSCF-based contact material
- Heat treatment schedule modeled:
  - 2 hr @ 930°C
  - Operating temperature distribution
  - Shutdown to 25°C
- Stack operating conditions:
  - 400 mA/cm^2
  - 97% H_2 fuel
  - 80% UF
  - 12% UA
  - 700°C furnace
- Stack temperature 703°C to 823°C with 120°C ΔT

Contact Material Locations

Symmetric Half-cell Geometry



Operating Temperature



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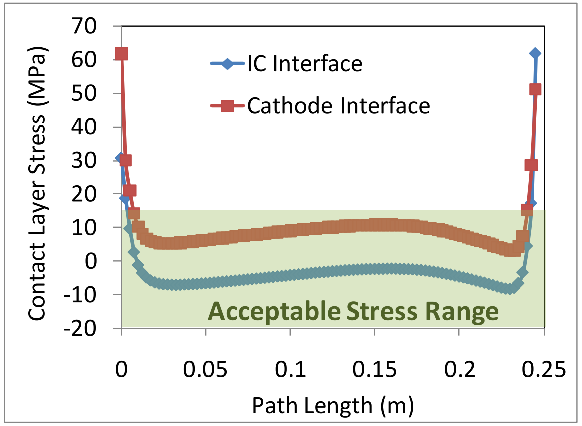
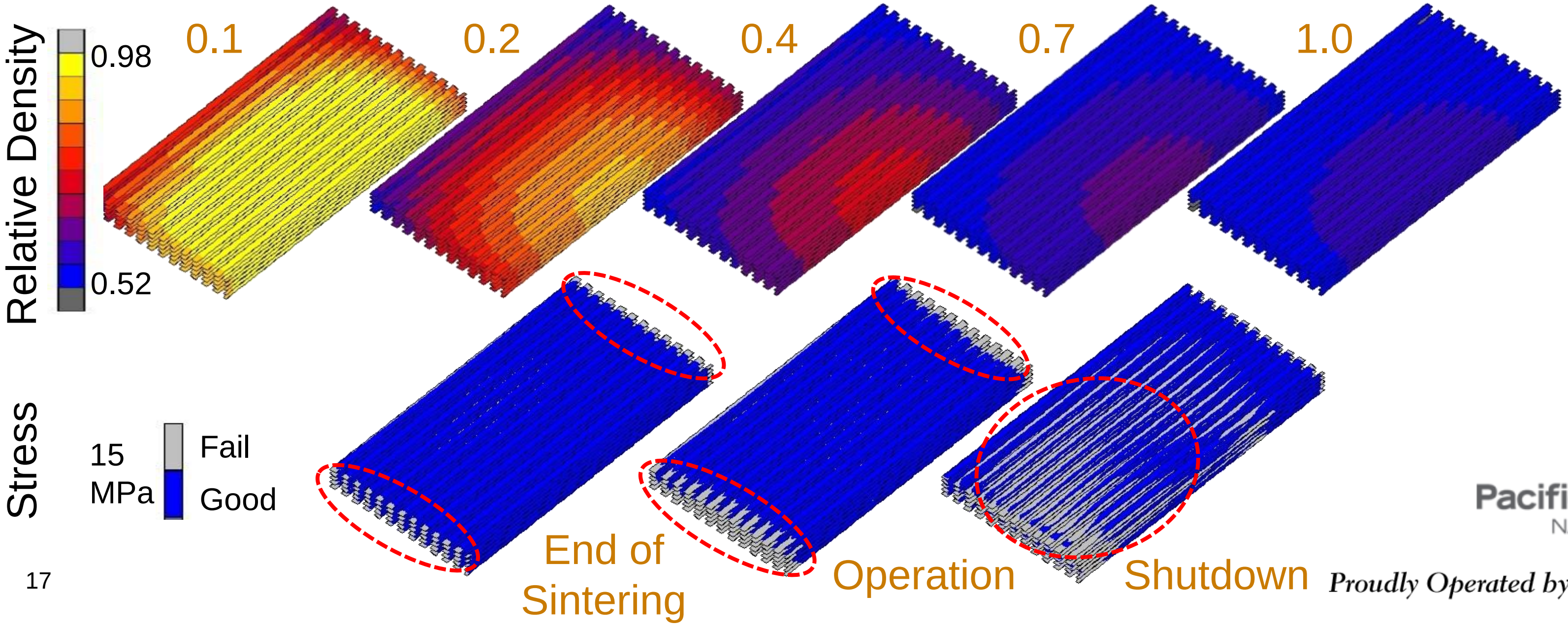
Contact Modeling:
Smaller Initial Grain Size Improves Density

- Densification with nominal 0.4 μm grain inadequate
  - Maximum density only 72%
  - Distribution non-uniform across the cell
    - Corners and edges restricted by stiff surrounding frame; relative spring stiffness between frame and active area will be critical design parameter
- Grain size < 0.5 μm substantially improved density but geometry influence remained

Contact Interface Stress





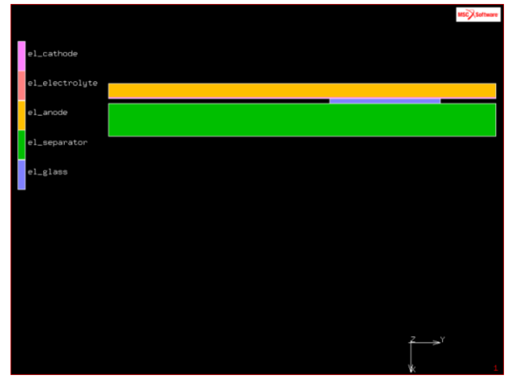
Initial Grain Size (μm)	Max Final Density
1.0	59%
0.7	63%
0.4	**72%**
0.2	91%
0.1	98%

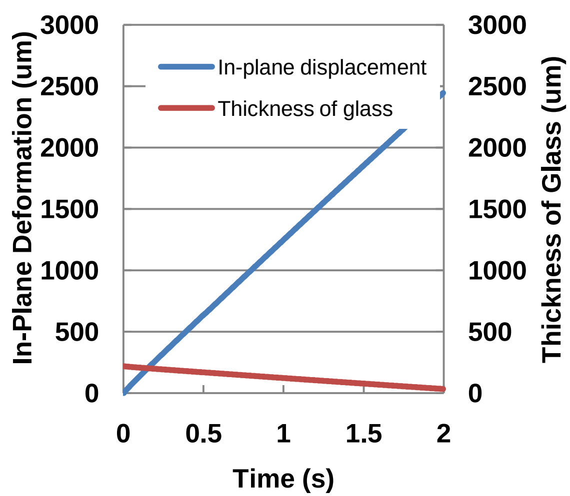
Seal Materials Modeling: Task Summary

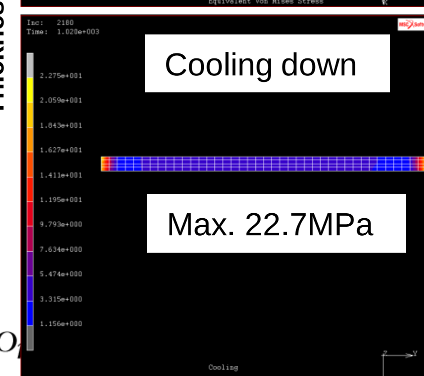
- Motivation: To explore the behavior of seal materials at operating temperatures and better understand the interplay of material microstructure and properties on stresses and degradation
- Goal: Use modeling to assist the development of a reliable sealing system to achieve the stack-level design requirements
- Technical Approach: Develop time and temperature dependent seal models to use as building blocks in stack-level seal performance simulations and investigate the effects of various design parameters on multi-cell stacks

Seal Modeling: Predicted In-Stack Behavior of Pure Glass Sealants

- Pure glass sealant will flow out without stopper.
- The stress in the glass and its interface with IC and PEN are very small at the working temperature.
- Cooling-induced stress in the glass and its interface with IC and PEN are relative large, and potential interfacial damage will occur.
- Gap between ceramic stopper and glass will reduce the stress in the glass and interface of glass and IC/PEN.
- Scale up of cell will increase the stress level in glass as well as its interfaces with IC and PEN

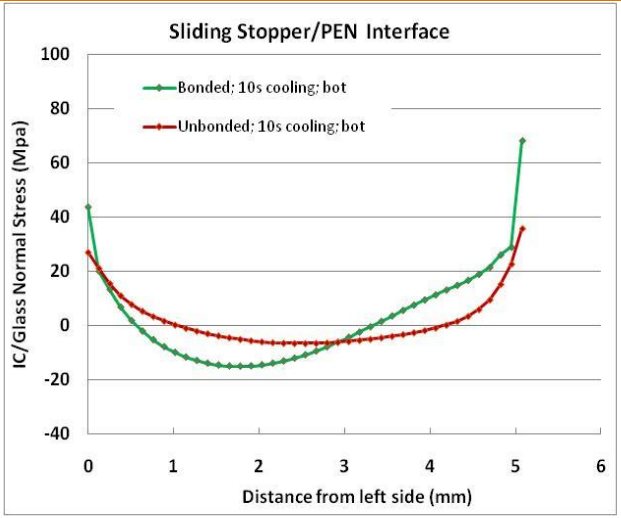


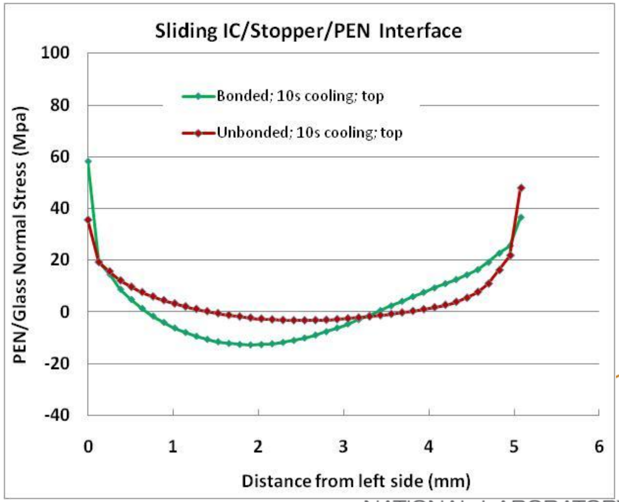




Seal Modeling: Predicted In-Stack Behavior of Pure Glass Sealants with Ceramic Stoppers

- Studied the effects of ceramic stoppers on the geometric stability of the self-healing seals in a simulated stack environment using creep analysis:
  - Stoppers will help the glass seal to maintain geometry during operation
- Studied effects of various interfaces of PEN/Stopper, IC/Stopper, and Stopper/glass on the interfacial stresses upon cooling:
  - Weak interfaces between stopper and glass always lead to lower stresses on glass/PEN and glass/IC interfaces.
  - In most cases, localized high stress regions are predicted for the edge of the glass seal: possible localized failure.





PNNL Modeling Summary

- SOFC-MP: 2D and 3D multi-physics stack models
  - 2D Software and manual released
- DEC Model: 3D multi-physics cell model
  - Resolves the local conditions within the cell and predicts SOFC global performance from cell level electrochemistry
- Contact Modeling: Continuum sintering model suitable for stack modeling
  - Good free sintering densification of candidate materials possible for T<1000°C
  - Reduced initial grain size improved densification with only a small negative impact on predicted stresses at operation and shutdown
- Interconnect Modeling and Experimentation: Integrated modeling and experimental approach for IC life prediction
  - Spallation resistance can be improved by increasing the interfacial strength between the oxide scale and substrate
  - Spallation driving force can be reduced through cooling profile optimization and by reducing IC thickness
- Seal Modeling:
  - Predicted the outflow pattern for pure glass seal with different initial glass height (volume) in PNNL leak test setup
  - Studied the possible self-healing mechanisms/driving forces for glass seal:
    - Role of pressure on crack healing rate
    - Role of gravity on healing rate for thin glass seal

Current/Future Modeling Activities

- Simulation of long-term and transient degradation behaviors
  - DEC Model: Implement secondary reactions (degradation)
  - SOFC-MP: Include transient degradation of state variables & coupling to DEC model
- Improved accessibility to software tools
  - Transfer DEC model to open source tool (e.g. OpenFoam)
  - Transfer SOFC-MP to a more flexible framework for interface with FEA solvers beyond MSC MARC
- Contact Modeling:
  - Identify/test a specimen configuration for validation of constrained sintering simulations
  - Adapt model to other volumetric behaviors in the stack (e.g. anode reduction, seal formation, re-oxidation tolerance)
- Interconnect Modeling:
  - Quantification of interfacial strength for varying surface modified SS441
  - Life prediction for coated and surface modified SS441
- Seal Modeling:
  - Continue to quantify the self-healing mechanisms for SCN glass and develop temperature dependent constitutive models for SCN glass considering aging/crystallization
  - Use modeling tools to virtually examine the various concepts of engineering seal design with glass/stopper sealing system in multi-cell stack