SECA Core Program– Recent Development of Modeling Activities at PNNL

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R&D Objectives & Approach

- **Objective**: Develop integrated modeling tools to:
  - Evaluate the tightly coupled multi-physical phenomena in SOFCs
  - Allow SOFC manufacturers to numerically test changes in stack design and performance to meet DOE technical targets
- **Approach**: Finite element-based analysis tools:
  - Mentat-FC: Easy-to-use pre- and post-processor to construct a complete analytical model from generic geometry or templates
  - SOFC-MP: A multi-physics solver that quickly computes the coupled flow-thermal-electrochemical response for multi-cell SOFC stacks
  - Probabilistic-based design methodology to assess system performance and component reliability against DOE technical targets
  - Targeted evaluation tools for eminent engineering challenges:
    - Interface and coating durability
    - Reliable sealing
    - On-cell reformation for thermal management
    - Structural integrity under thermal cycling
    - Time dependent material degradation

Accomplishments

- **Stack Design Tool Available**: PNNL and MSC-Software combined efforts to develop and release a user-friendly electrochemical-thermal-structural stack design software package (consortium available). Design tool capability includes import of planar and non-planar SOFC stack designs
- **Probabilistic-Based Design Methodology**: Methodology developed in which probability of failure of stack components can be made uniform for a proposed stack design
- **Glass-Ceramic Seal Damage Characterized**: Experimentally-based model enables prediction of damage accumulation and failure in steady and thermally cycled stacks
- **Characterization of On-Cell Reformation in Stacks**: Experimentally- based reformation model enables prediction of the effects of on-cell steam-methane reformation under variable stack operating conditions
- **Experiments Provide Critical Properties**: Testing has provided fundamental material properties enabling model development

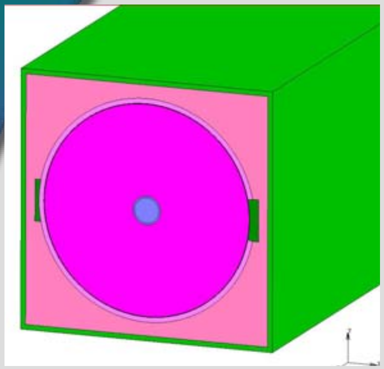
Teaming and Collaborations

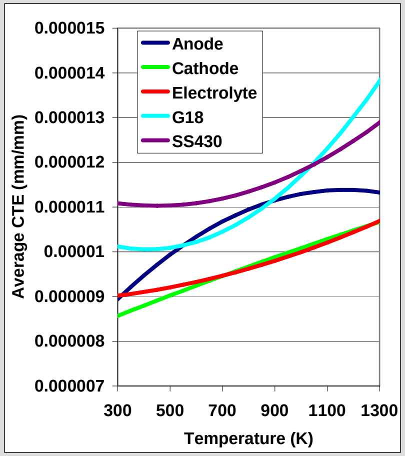
- Industry
  - Modeling tool training
    - GE
    - Delphi
    - Acumentrics
    - Siemens
- University and National Labs:
  - Georgia Tech
  - ORNL
  - U CONN

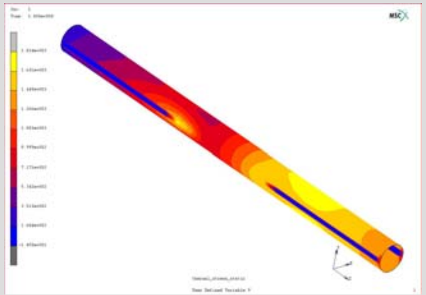
Results to Date

Mentat-FC & SOFC-MP Tools
On-Cell Reforming
Coarse Methodology
Seal Damage and Thermal Cycling
Experimental Support of Modeling

SOFC Analysis Overview







MENTAT-FC MODEL GENERATION

Model Dimensions
Boundary Conditions
Material Properties
Fuel Cell Operating Parameters

↓

SOFC-MP™
FUEL CELL ELECTROCHEMICAL, FLOW, AND HEAT TRANSFER ANALYSIS

↑↓

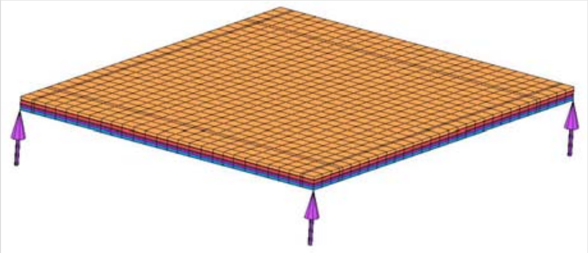
MSC-MARC

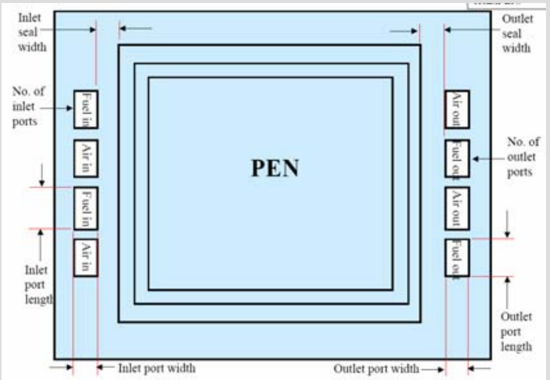
Thermal/Structural Analysis

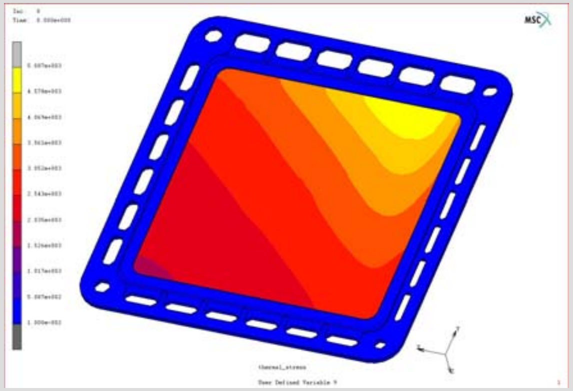
↓

MENTAT-FC RESULTS VIEWER

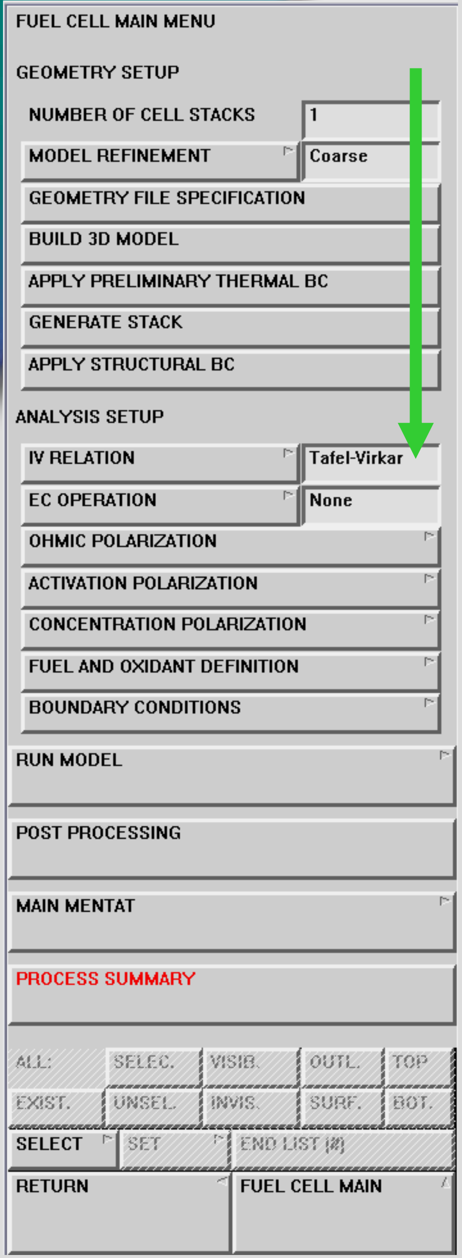
Stresses
Deformations
Temperatures
Gas Species



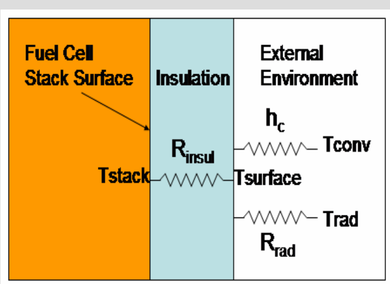




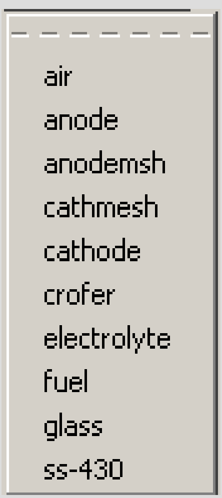
Mentat-FC Model Generation



- GUI guides user through entire analysis
- Geometry
  - Generic CAD (ACIS format)
  - Planar Template (co-, counter-, cross-flow)
  - Tubular
- SOFC operating parameters
  - I-V relation
  - Fuel utilization, total voltage, total voltage options
  - Fuel/oxidant inlet concentrations/rates
  - Polarizations
- Boundary conditions
  - Generic thermal losses from stack

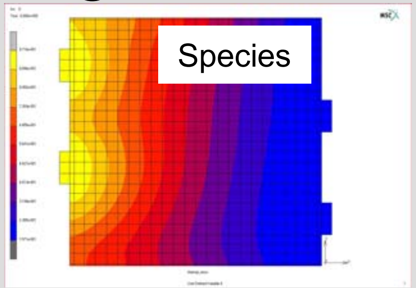


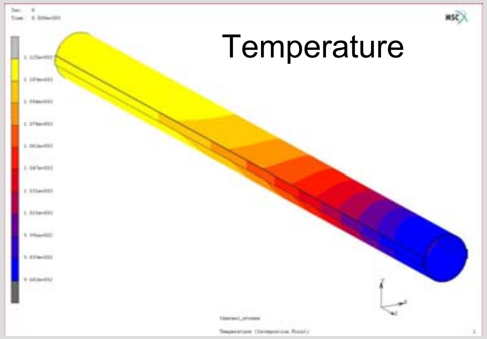
- Material properties
  - Pre-populated database
  - User-defined

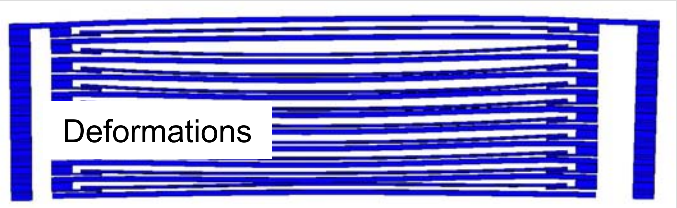


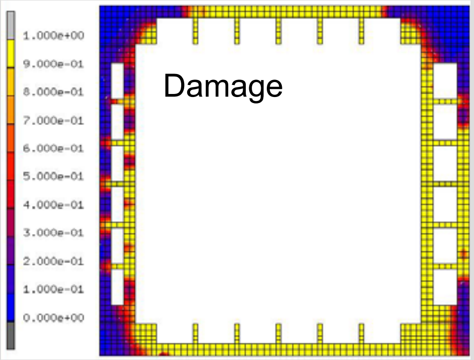
Mentat-FC Analysis and Results

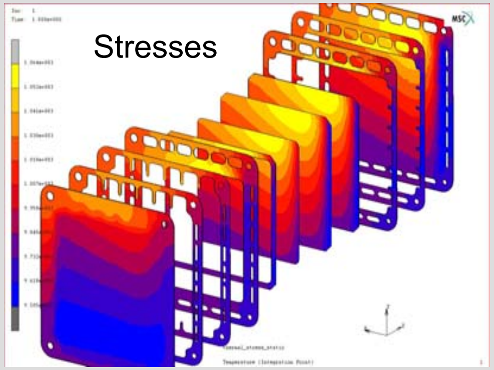
- Automated post-processing
  - Power output
  - Species depletion
  - Thermal distribution
  - Deformation and stresses
- Customized evaluation tools
  - On-cell reformation
  - Seal damage
  - Creep
  - Thermal cycling
  - Leak







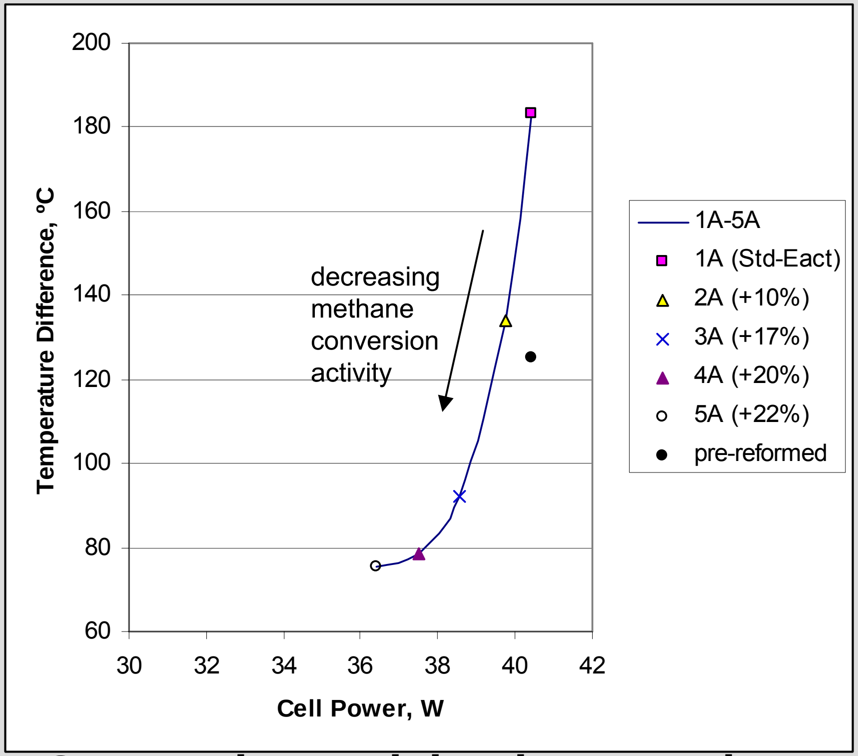




On-Cell Reforming: Manipulation of Conversion Activity

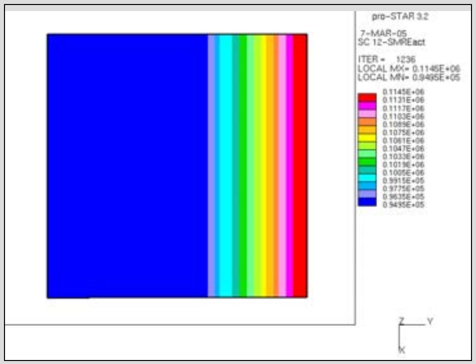
- PNNL experimentalists are developing modified anode materials to slow methane conversion
- The modeling tool can be exercised to simulate the effect of possible anode material manipulations
- Model predictions show temperature difference benefit resulting from decreased conversion activity uniformly on cell area:
  - 57% decrease in cell temperature difference (4A)
  - 7% decrease in gross power (4A)
- Temperature difference benefit created by decreased methane conversion is limited as hydrogen formation decreases

Case Study: 110.24 cm^2 cross-flow cell, 750°C, 0.7 Volts, (0.53 A/cm^2 baseline)

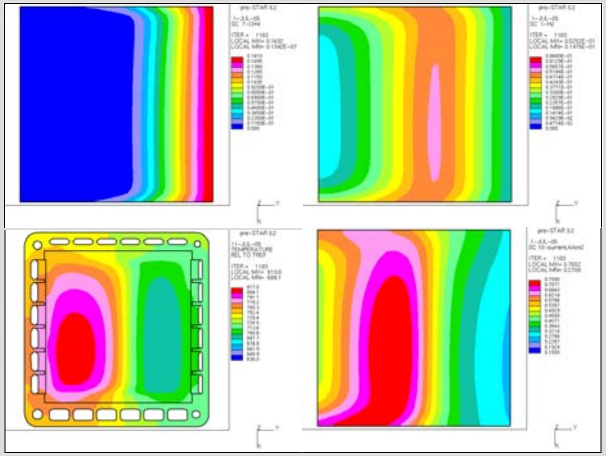


Conversion activity decreased uniformly on the cell area

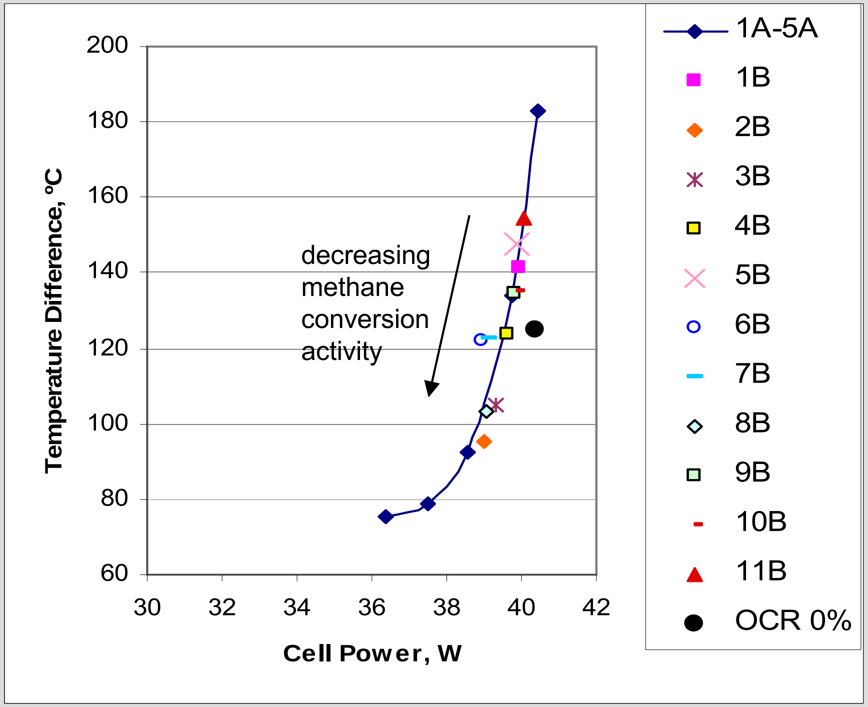
On-Cell Reforming: Non-Uniform Conversion Activity:



Nonuniform Activation Energy Distribution of Case 5B in J/mol



Nonuniform Activation Energy Case 5B Distributions (from left to right and top to bottom: methane partial pressure, hydrogen partial pressure, temperature, and current density);the fuel flows from right to left and air from top to bottom

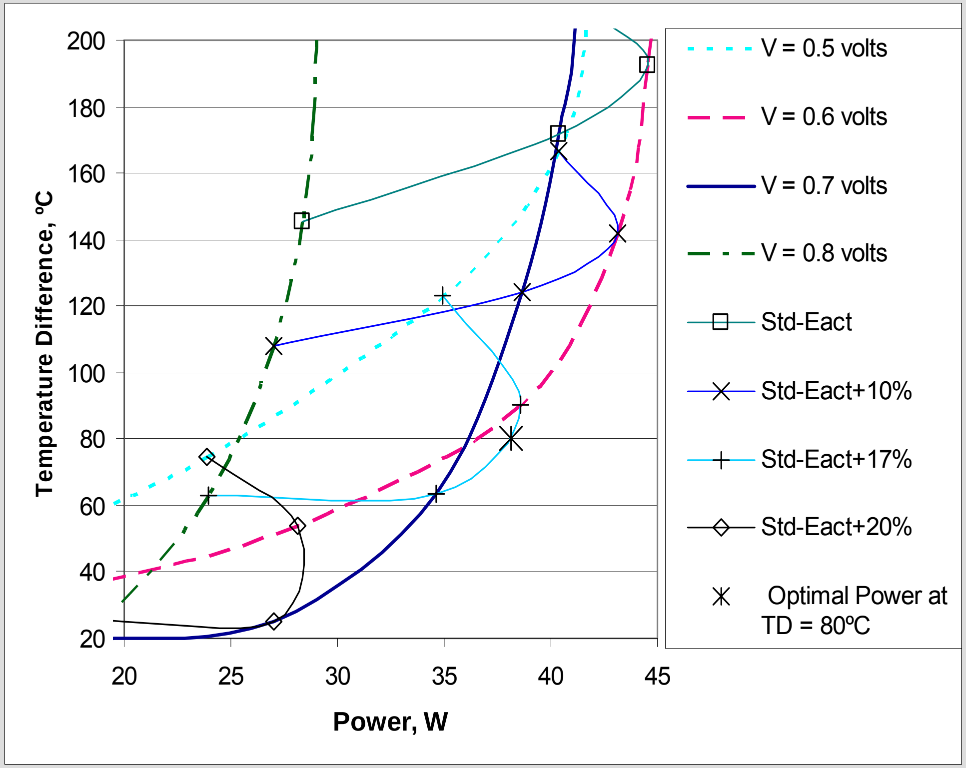


Non-uniform manipulations of conversion activity show no marked benefit compared to uniform activity changes.

Case Study: 110.24 cm^2 cross-flow cell, 750°C, 0.7 Volts, (0.53 A/cm^2 baseline)

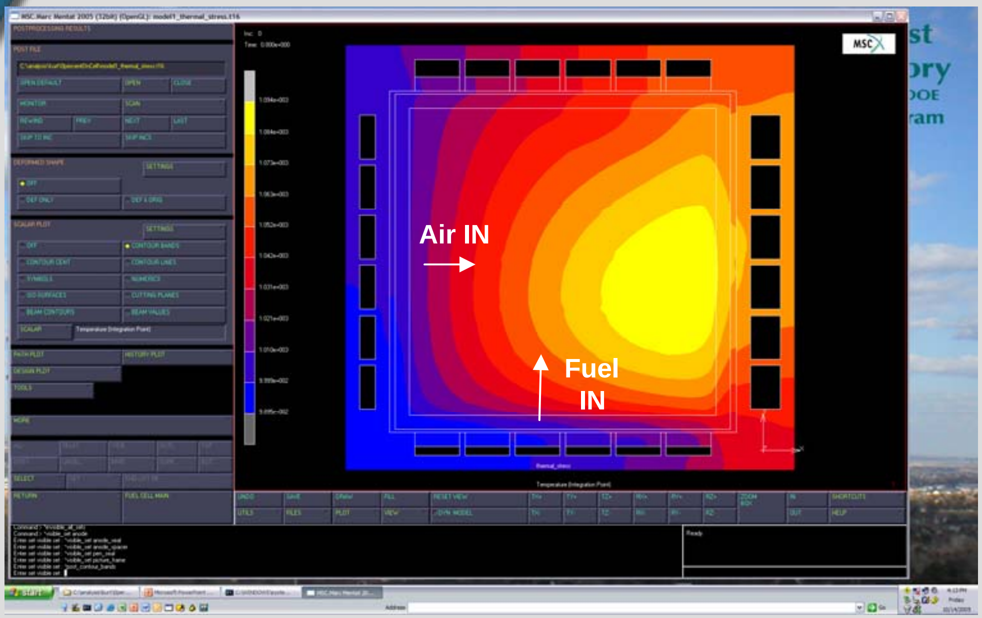
Maximizing Power: Cell Voltage and Uniform Activity

- Cases expanded to include range of cell voltages
- At each voltage, the cell temperature difference decreases with methane conversion
- For a chosen acceptable temperature difference, the power can be maximized by proper choice of voltage and conversion activity

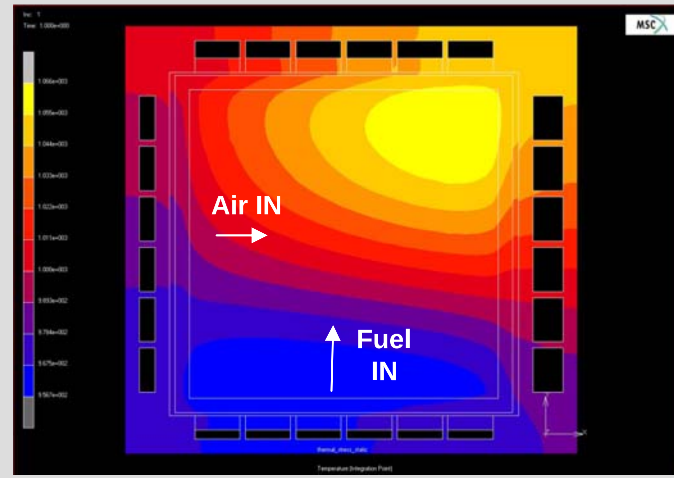


Case Study: 110.24 cm^2 cross-flow cell, 750°C, 0.7 Volts, (0.53 A/cm^2 baseline)

On-Cell Reforming: Simulations in Mentat-FC



1 - H_2 fuel (No CH_4)



2 - Standard Rate On-Cell Reforming

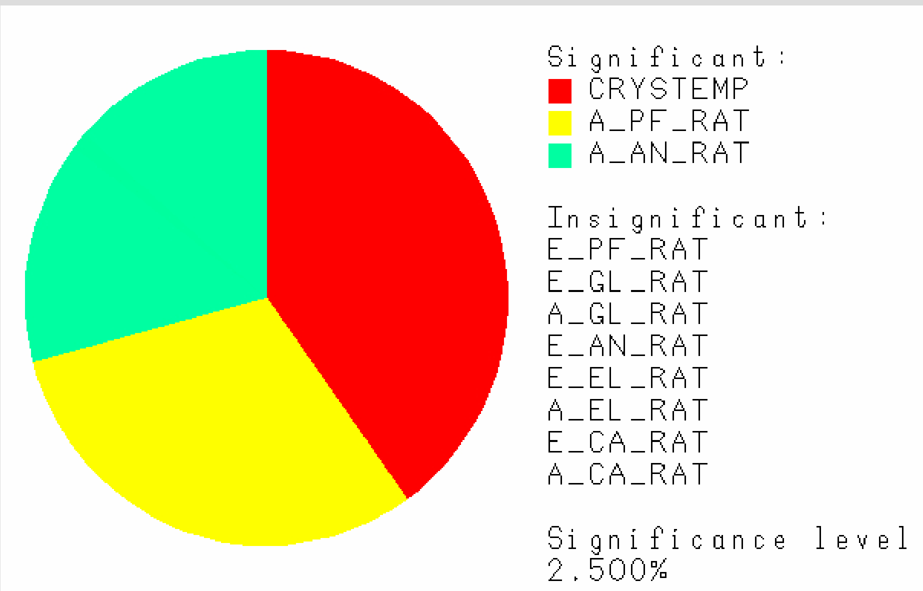
Case	Temperature, °C Min	Temperature, °C Max	$S1_{max}$, MPa Anode	$S1_{max}$, MPa Seal	$S1_{max}$, MPa Picture Frame
1 – No CH_4	720	821	9.7	6.6	98.1
2 – Standard Rate	684	793	5.0	7.6	99.7

Probabilistic Based 'Coarse Design Methodology' for SOFC Stacks

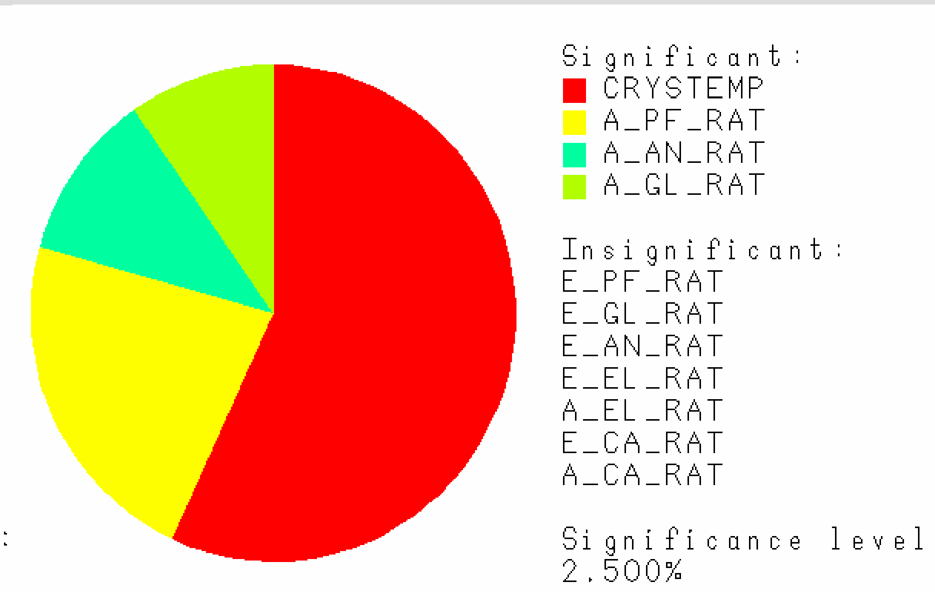
- FY05 Accomplishments
  - Performed cell maximum principal stress sensitivity study under start-up/cool-down condition and operating condition.
  - A probabilistic-based component design methodology is developed for solid oxide fuel cell (SOFC) stack.
  - Component failure probabilities for any particular design can be calculated as a function of operating conditions.
  - Procedures for calculating the safety indices for anode and seal have been demonstrated such that uniform failure probability of the components can be achieved.
  - Documented analyses results and procedure in PNNL Topical Report.

Component Level Sensitivity Study
- Isothermal Start-up and Cool-down

Maximum principal stress in anode

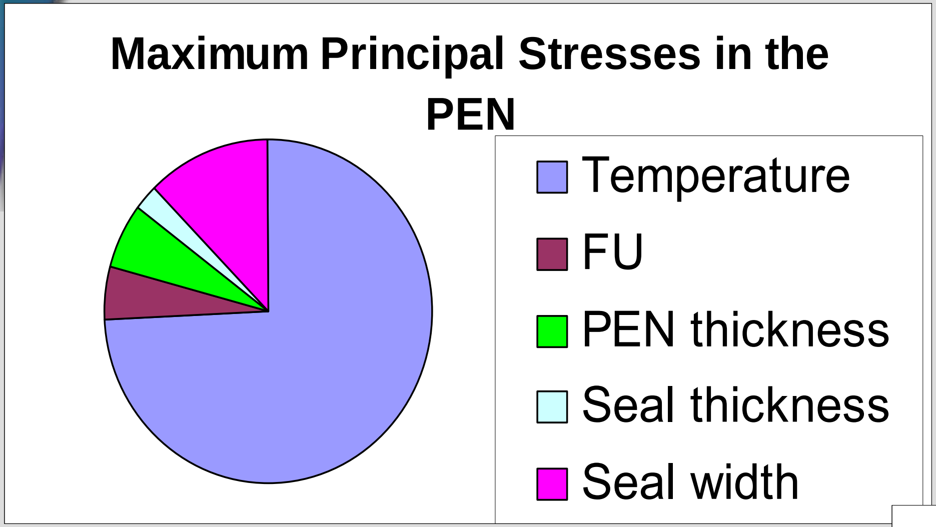


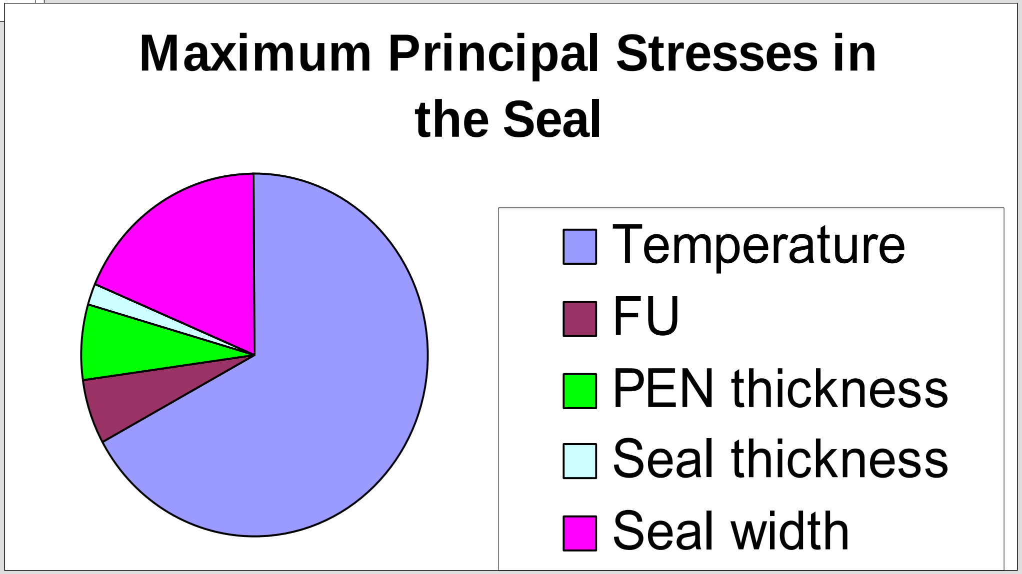
Start-up (Operating temperature)



Cool-down (Room Temperature)

Component Level Sensitivity Study
- Operating Condition, example 1





Component Level Sensitivity Study
- Operating Condition, example 2

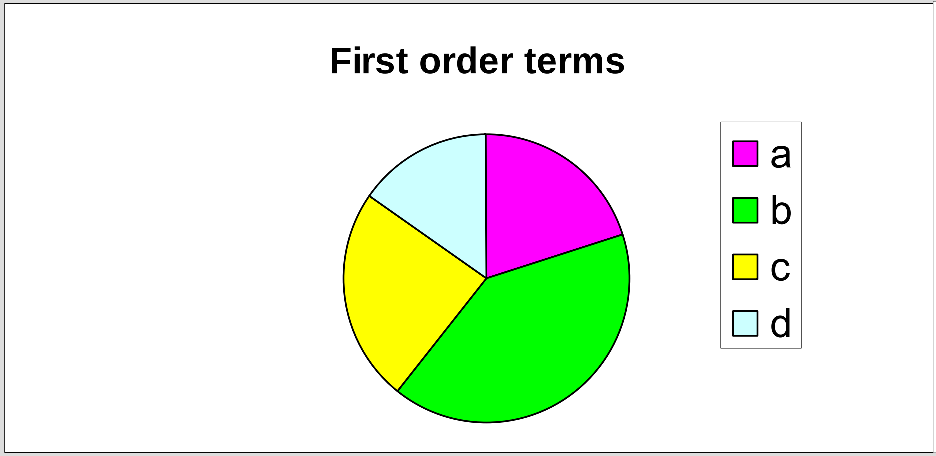
Design variables considered:

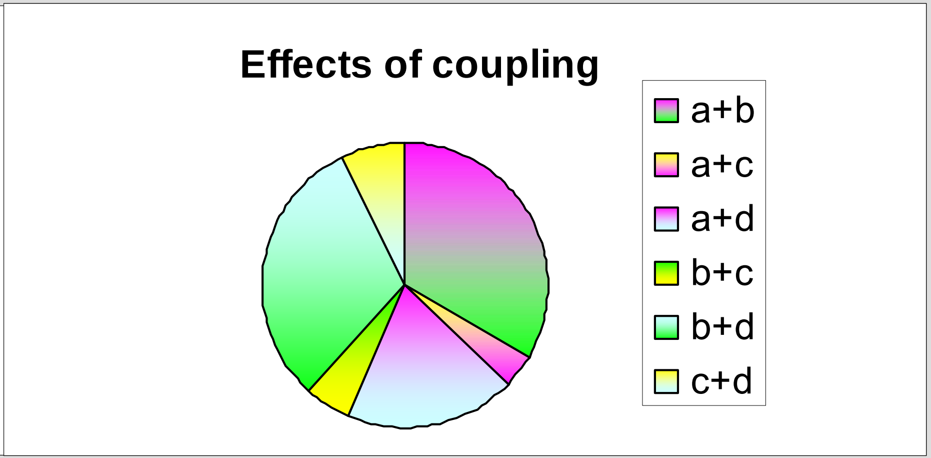
(a) Increase seal width from 0.5mm to 0.55mm.

(b) Increase all PEN layer thicknesses by 10%.

(c) Decrease stainless steel CTE to the weighted average of the PEN layer CTEs.

(d) Increase width of the cell active area by 10%.





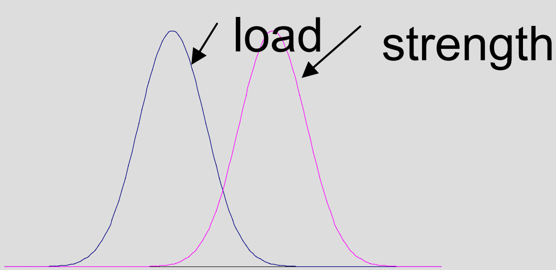
Influence of different parameters on anode maximum principal stress

Probabilistic Based 'Coarse Design Methodology'

$$Design\ goal:\quad stress < \frac{strength}{\theta},\quad \theta: equivalent\ safety\ factor$$

Example design target: uniform component failure probability Pf=0.0014, safe index β=3.

$$\beta = \frac{\bar{R}_i - \bar{S}_i}{\sqrt{\sigma_{R_i}^2 + \sigma_{S_i}^2}}$$



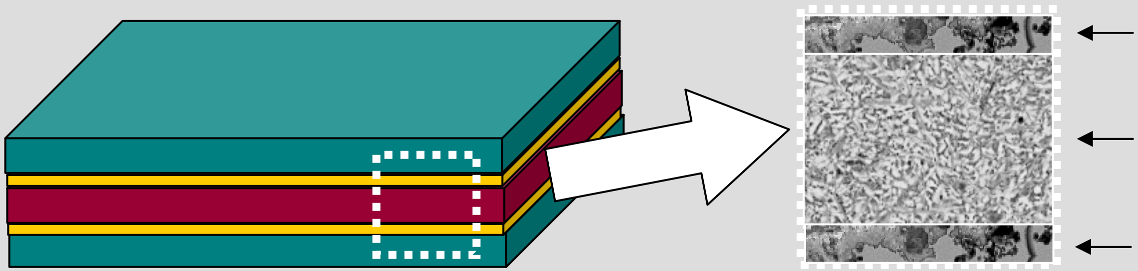
Safety factors helps guide future design by:

- Changing material strength
- Changing operating conditions
- Changing design parameters

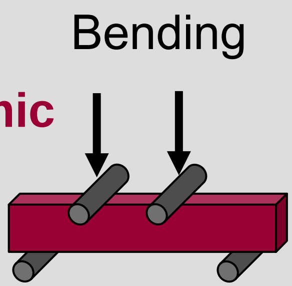
Operating condition	Equivalent safety factors	
	θ_{anode}	θ_{seal}
T=700°C, FU=45%	1.85	1.52
T=700°C, FU=70%	1.89	1.51
T=700°C, FU=90%	1.85	1.51
T=750°C, FU=45%	1.66	1.53
T=750°C, FU=70%	1.58	1.53
T=750°C, FU=90%	1.54	1.51
T=800°C, FU=45%	1.47	1.62
T=800°C, FU=70%	1.43	1.6
T=800°C, FU=90%	1.42	1.51

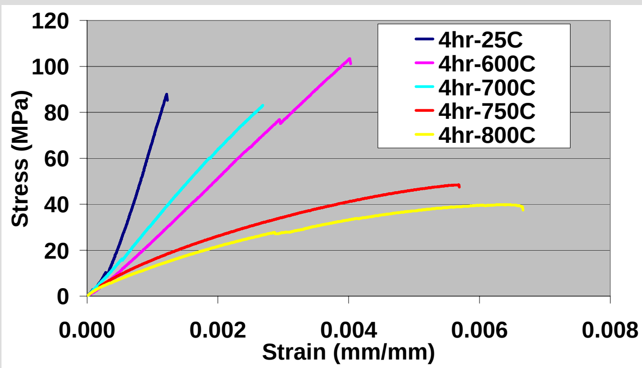
Seal Damage Modeling
Mechanical Testing for Material Response

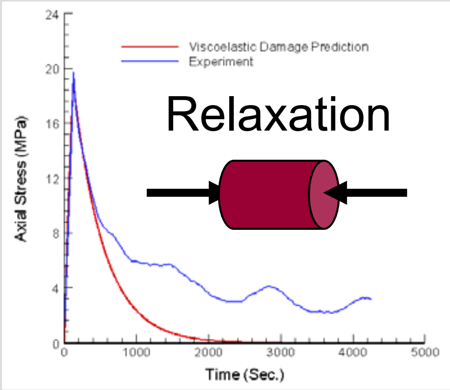
Characteristic Stack Seal Assembly



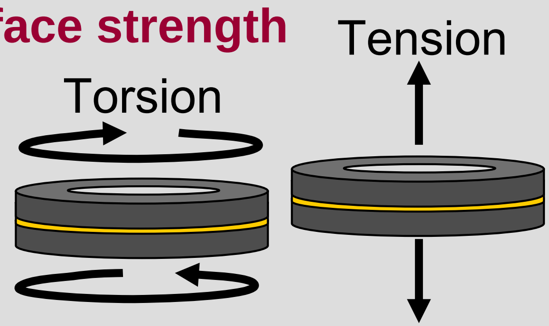
1. Test the glass-ceramic material strength

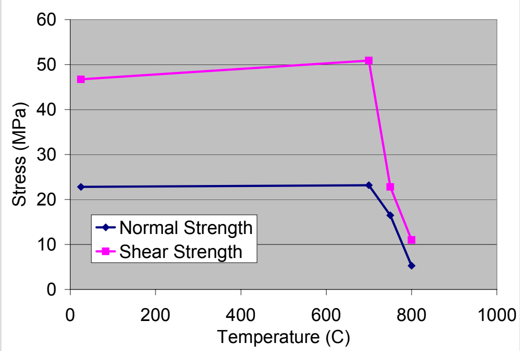






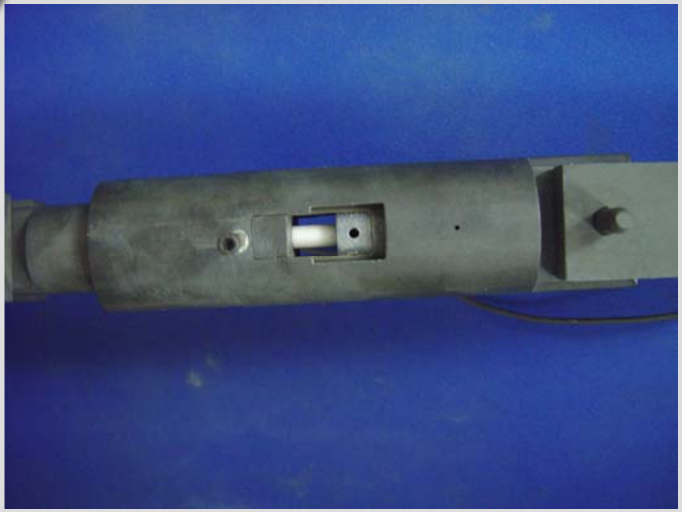
2. Test the weaker interface strength





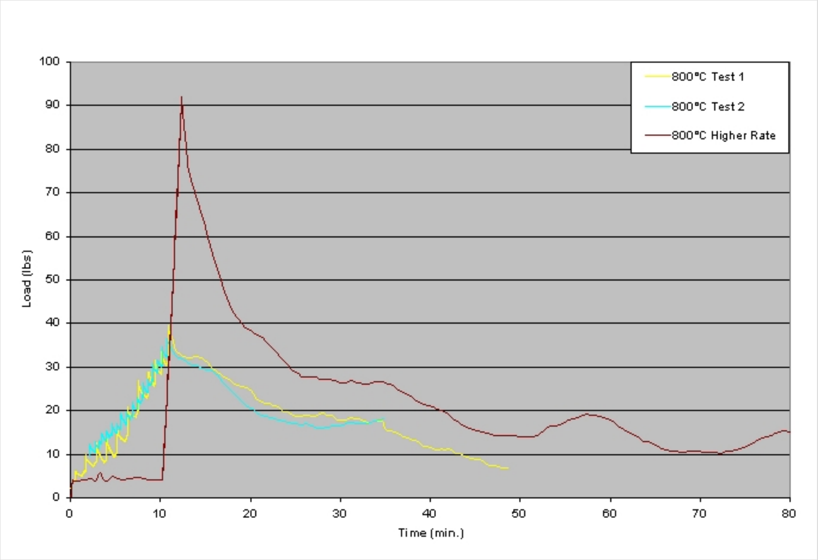
3. Obtain elastic moduli and the coefficient of thermal expansion (CTE)

Time-Dependent Behavior of G18 Glass



Samples are 5mm diameter, 10mm high, right-circular cylinders

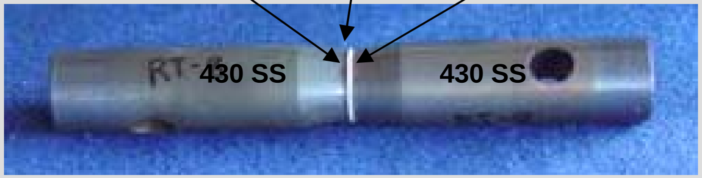
Deformation at $1 \times 10^{-5}\ s^{-1}$ and $1 \times 10^{-4}\ s^{-1}$ to approximately 0.5% compressive strain, then allowed to relax. This simulates strains created during heat-up of stack and relaxation at high temperatures. Sample viscosity can be measured and high-temperature deformation modeled.



Data From Seal Assembly Analogs

0.020” Crofer 22 APU washer (Ni brazed to 430) on both sides

Dispensed Glass



Tension

Torsion

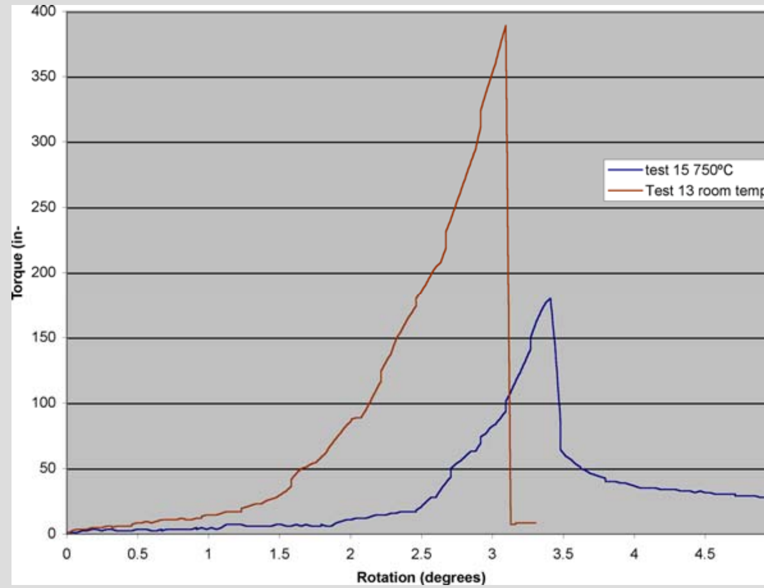
Testing Method	Test Temperature (°C)	Mean Failure Stress (MPa)	Number of Samples
Tension	25	22.8	2
	700	23.7	5
	750	16.5	6
	800	5	6
Torsion	25	23.4	6
	700	25.5	6
	750	11.4	6
	800	5.5	6

Thin-film analogs to test the entire seal assembly. These complement the previous tests in bulk glass.

Failure is generally interfacial rather than in the glass itself indicating that the interface needs further development.

Mica/Glass Hybrid Seals

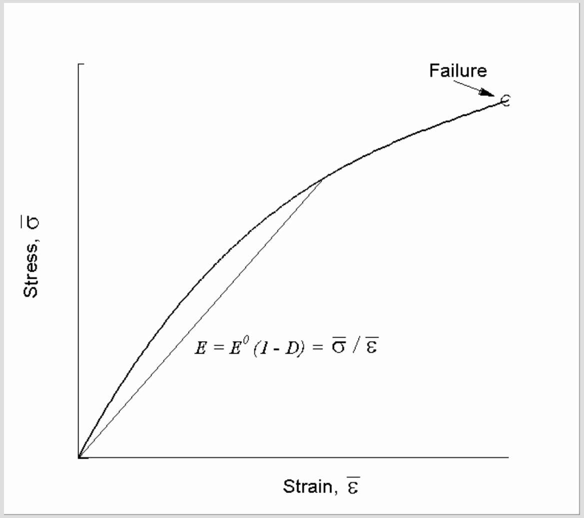
Mica/glass hybrid seals are proposed for use at the ends of the stack where shear stresses are higher.



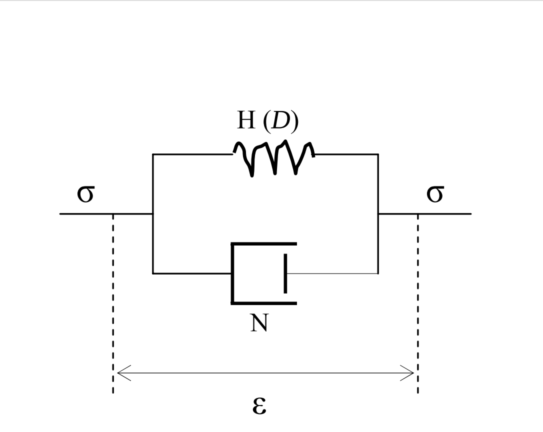
At RT the glass broke along the glass-Crofer interface but at 800°C the mica deformed. This behavior is reflected in the torque-rotation graph where the RT test shows a drop-off to zero torque but the 800°C test loaded, then dropped to a roughly steady-state rotational stress.

Seal Damage Modeling
Constitutive Models for Observed Behavior

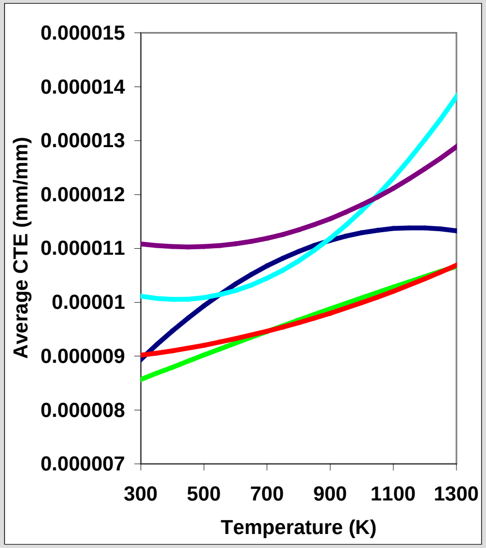
1. Develop a continuum damage model to study accumulated damage in the seal and interface which results in cracking and leakage



2. Extend the damage model to include viscoelastic response of the glass matrix to model creep and relaxation in transient stack operation

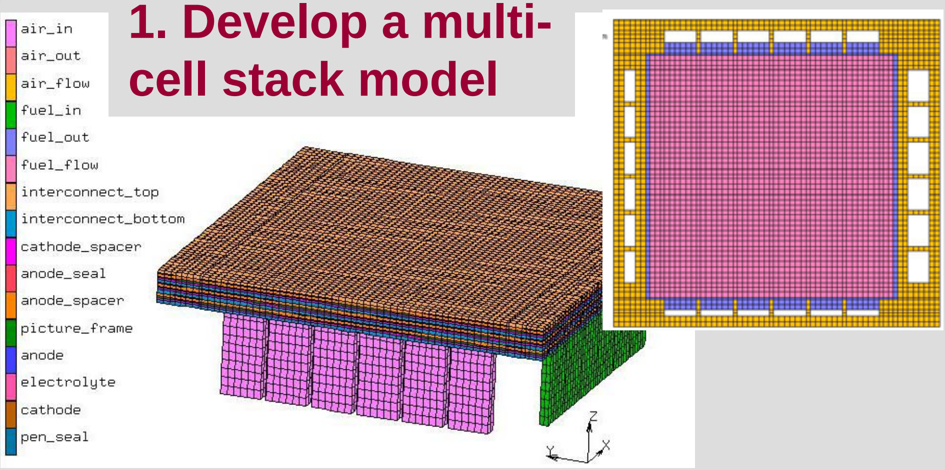


3. Apply temperature-dependent coefficient of thermal expansion to accurately capture the thermal mismatch stresses of the stack components

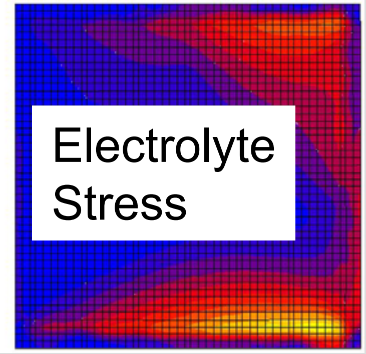


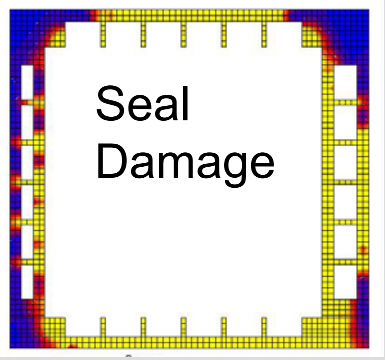
Seal Damage Modeling
Stack Stress Analysis

1. Develop a multi-cell stack model

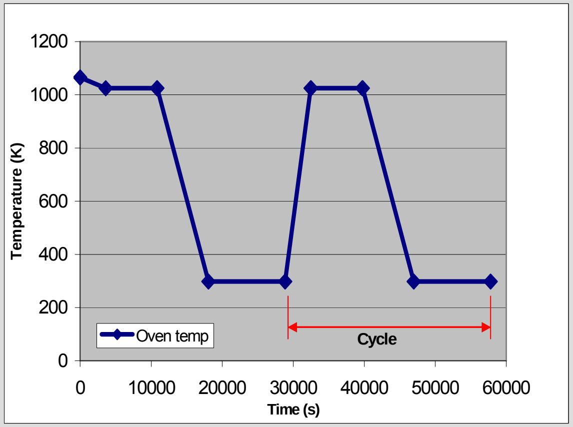


3. Track stresses and damage during the thermal transient





2. Impose a desired thermal load cycle on the stack via temperature history of inlet flows and surroundings. Include EC heat generation during "operation"



Conclusions

- Damage begins in first cycle
- Bottom seal fails first

Activities for the Next 6 Months

- Model improvement/calibration:
  - Interface modeling
  - Time dependent property incorporation
  - Viscoelastic damage modeling of seals
- Parametric studies on material properties and design parameters to guide material development activities
- Electrochemical degradation modeling
- Effects of on-cell reformation on stack thermal and electrochemical performance
- Automation of the reliability-based design framework for easy execution
- Measurement of mechanical degradation of seals and other interfaces

Looking Forward- Phase II

- Degradation modeling and life prediction
  - Seal
  - Interconnect
  - Cell
  - Interfaces
- Scale up within SECA goal
  - Virtual feasibility study on
    - Stack EC performance
    - Stack structural reliability
- System integration
  - Stack thermal management and cell thermal profiles
  - Integration with other components
- Validation