Modeling Tools for SOFC Design and Analysis: Recent PNNL Progress

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PNNL Modeling Focus

1. Develop stack modeling tools
   - SOFC-MP 2D/3D: Multi-physics solver for computing the coupled flow-thermal-electrochemical response of multi-cell SOFC stacks
   - SOFC-ROM: Creation of high fidelity reduced order model (ROM) for use in system-level studies
   - User Interface: Make these tools accessible to stack designers

2. Develop models to improve component durability
   - Compliant Seals: Damage-healing constitutive model for study of seal designs
   - Interconnects: Model to evaluate effects of surface modifications and coatings on durability and lifetime

Summary of Accomplishments

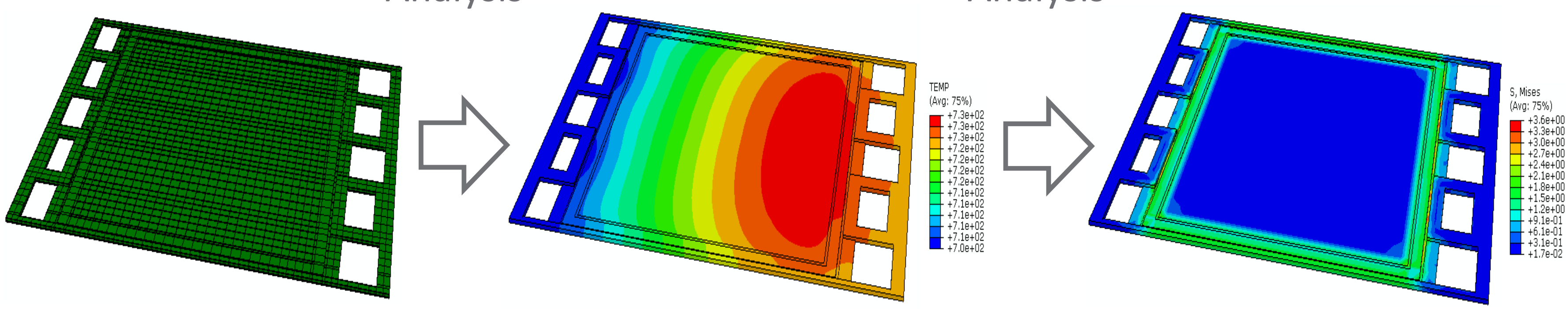
- Stack Modeling Tools
  - Coupled 3D SOFC-MP tool with ANSYS and ABAQUS finite element codes for structural analysis
  - Created a graphical user interface (GUI) for pre- and post-processing of 2D and 3D SOFC-MP models
  - Completed reduced order modeling (ROM) tool
- Compliant Seals
  - Evaluated compliant seal performance and damage-healing evolution in multi-cell stack simulations under thermal cycling
- Metallic Interconnects
  - Developed an experimental-modeling approach for prediction of interconnect lifetime using interfacial indentation tests of surface-modified, coated interconnects

SOFC-MP 3D Integration with FEM Software

- Integration with ABAQUS and ANSYS FEM models implemented
  - Models created in the FEM environment per guidelines and exported
  - Mesh is read by SOFC-MP and the electrochemical model is solved
  - Thermal distributions from the 3D simulation exported
  - Structural analysis run in the commercial FEM code using the exported temperature field data

3D SOFC-MP Analysis

FEA Analysis



FEM Geometry/Mesh

Temperature Field

Thermal Stress Distribution

Example: SOFC-MP/ANSYS FEA Model

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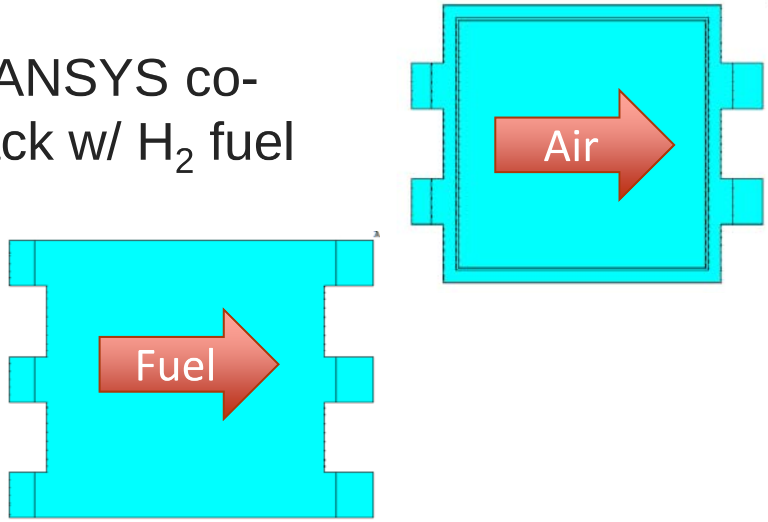
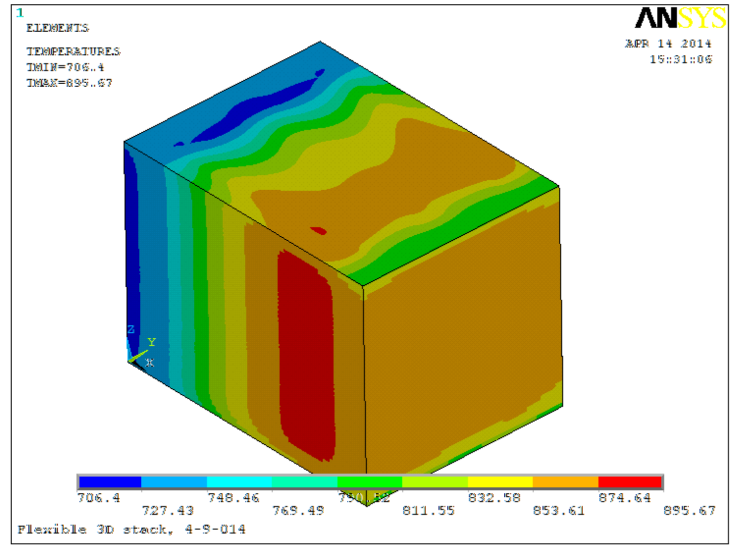
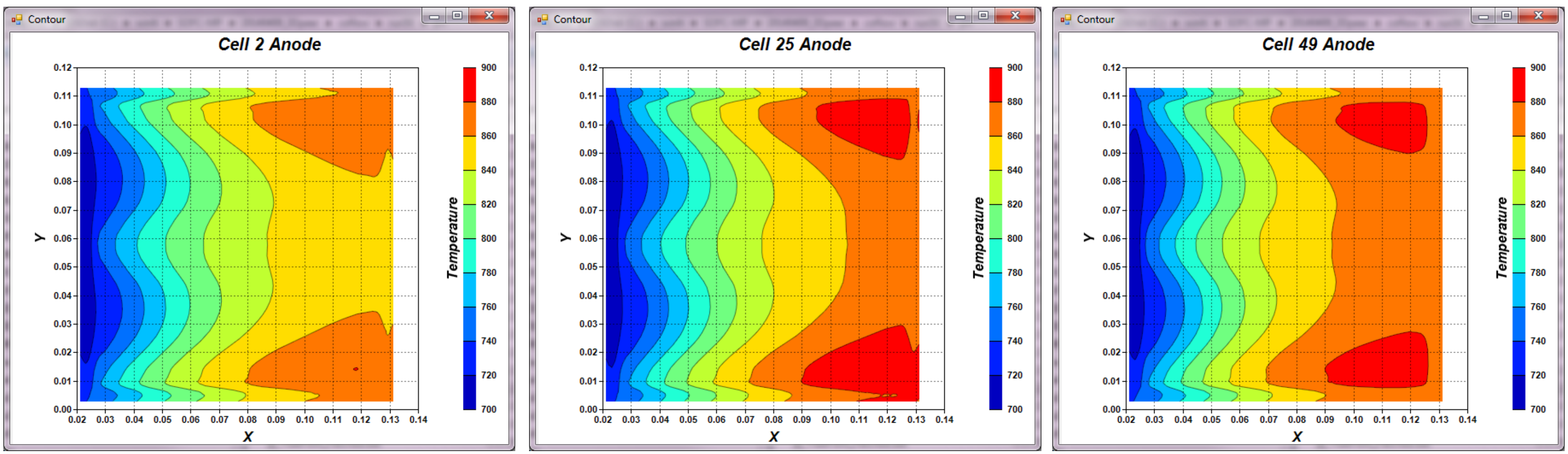
- 50-cell ANSYS co-flow stack w/ H_2 fuel

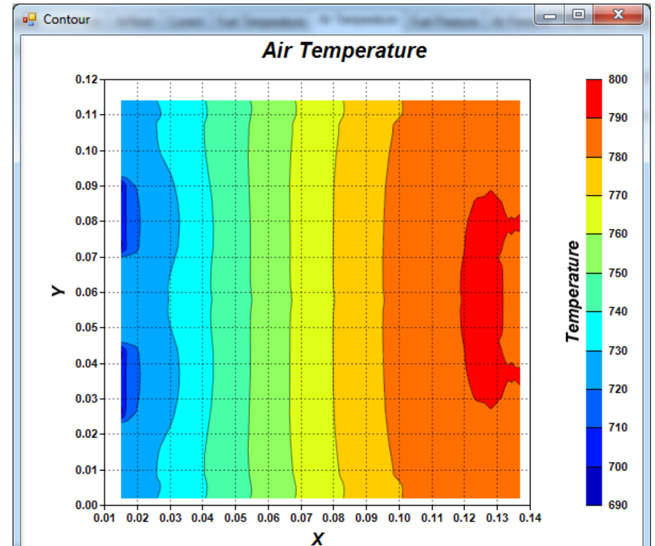




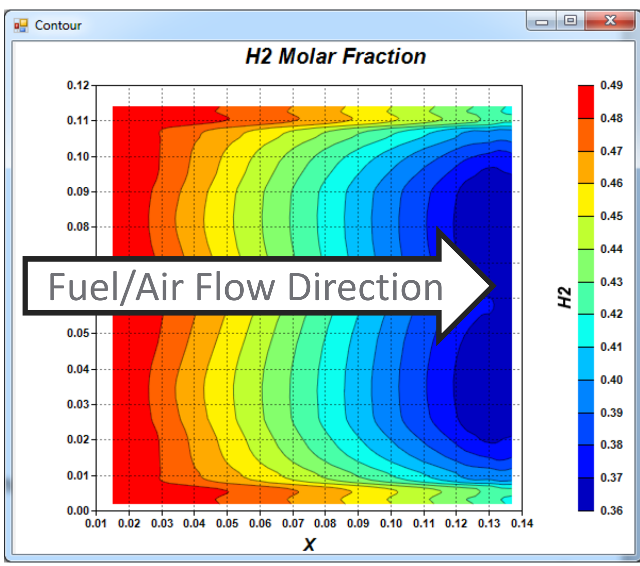


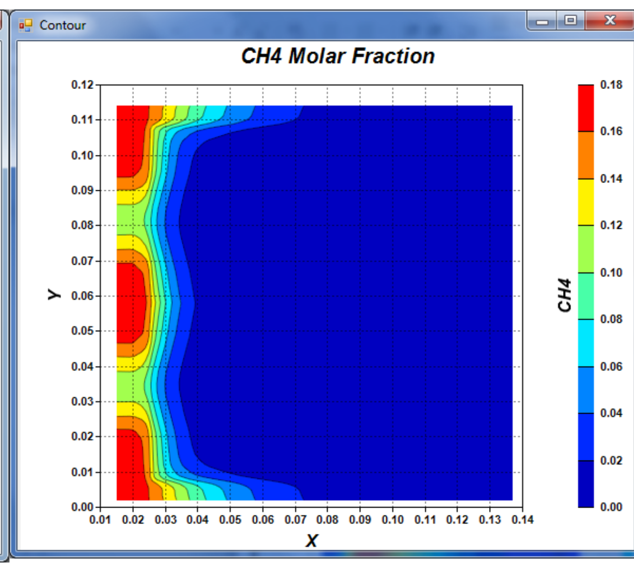
SOFC-MP GUI

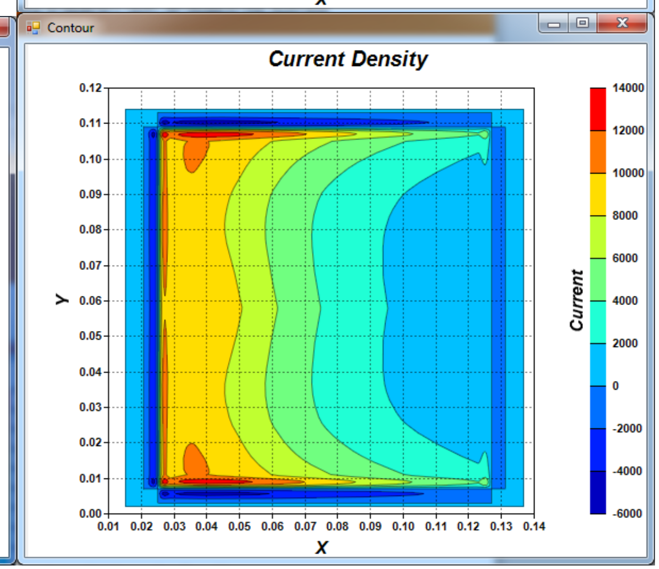
- Graphical user interface replaces legacy MSC-MARC tool
- Tabular menu structure provides pre-processing, job submission, job monitoring, and post-processing capabilities for both 2D and 3D SOFC-MP simulations



3D SOFC-MP planar contours for cells



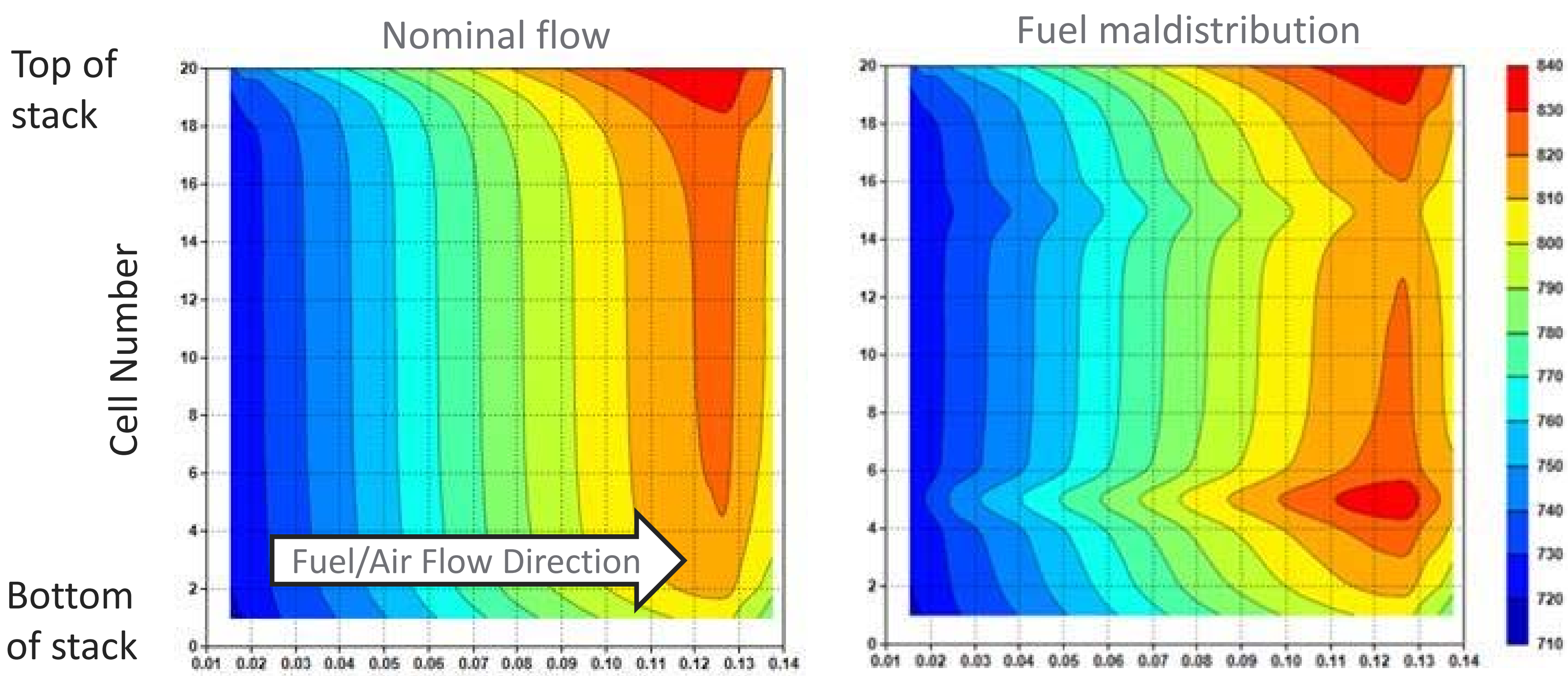




SOFC-MP GUI (cont'd)

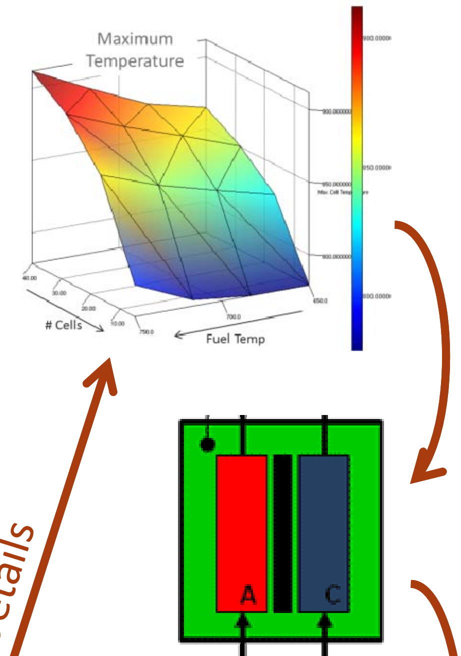
- Cross-section plot of fuel temperature for 20-cell 2D model
  - Effect of different fuel utilizations for cells #5 and #15 show higher peak temperature and gradient

2D SOFC-MP cross-section contours for stack



System Design Challenge

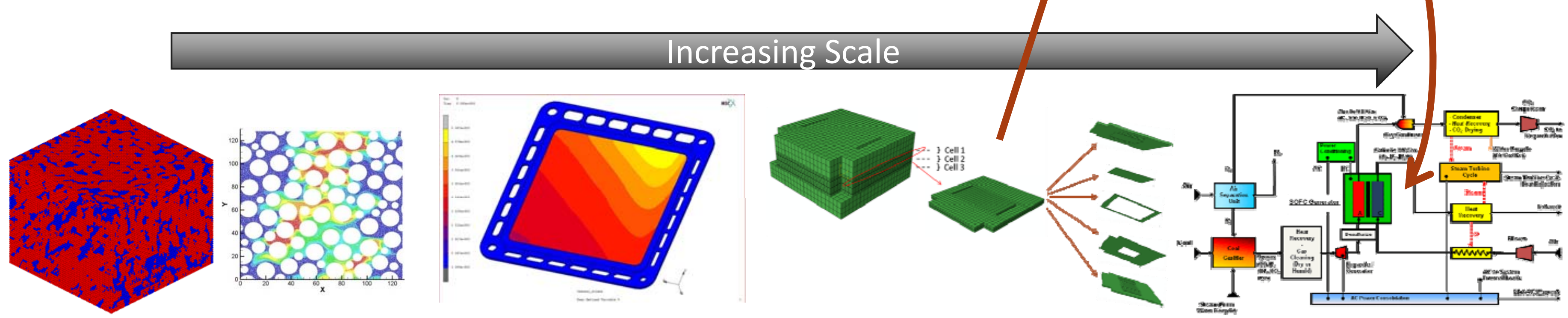
- Ultimately, need design/engineering at several scales for successful SOFC commercialization
- Modeling is practically limited by required computation times
  - Lower scales require smaller spatial resolution
  - Higher scales sacrifice detail or accuracy
- Utilizing more details from the cell/stack level models to inform the system level model will allow more insight necessary for optimizing the BOP components and control strategies



Response Surface Analysis

Reduced Order Model (ROM)

Details



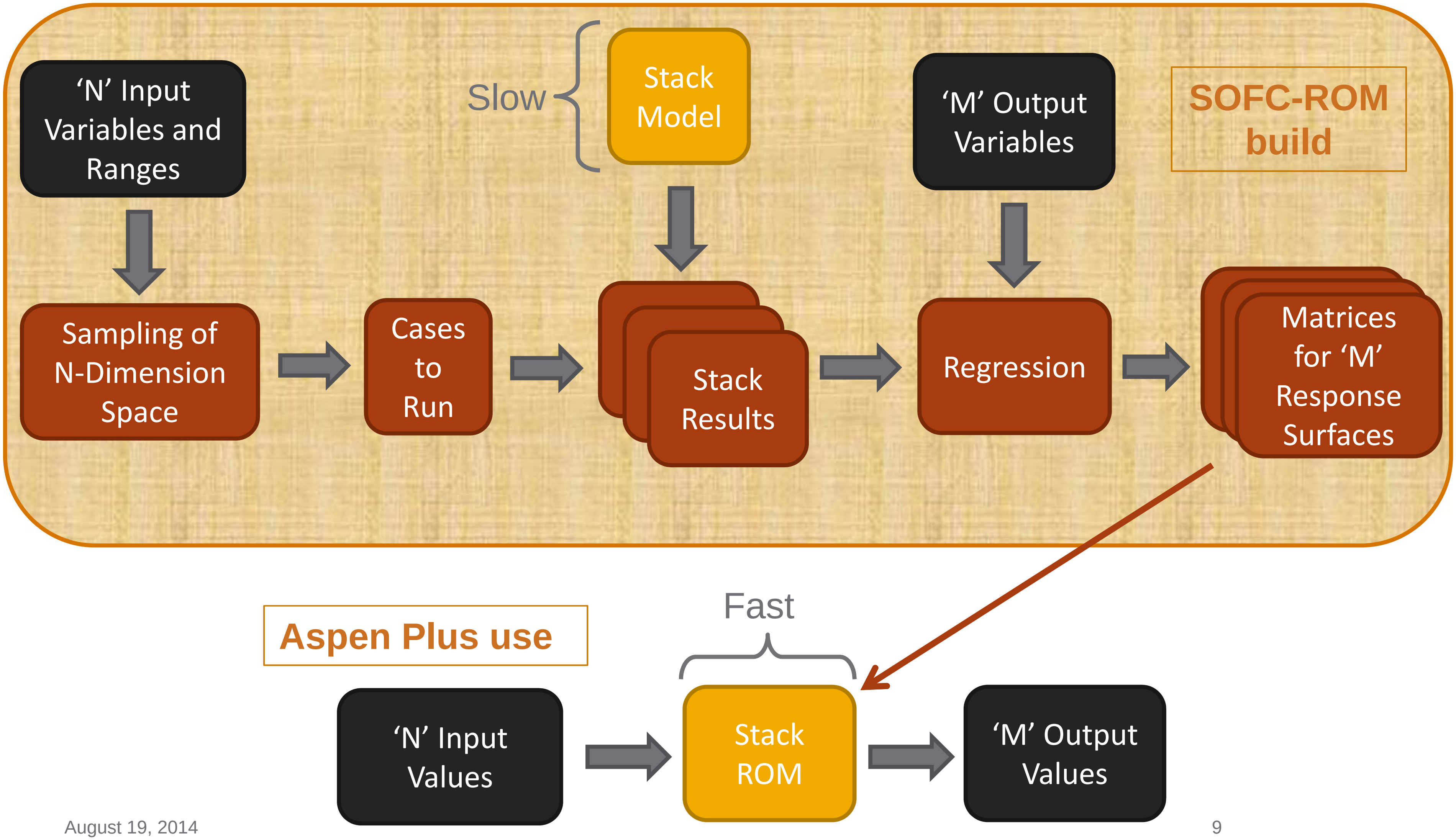
Electrode Microstructure

Single Cell

Multi-Cell Stack

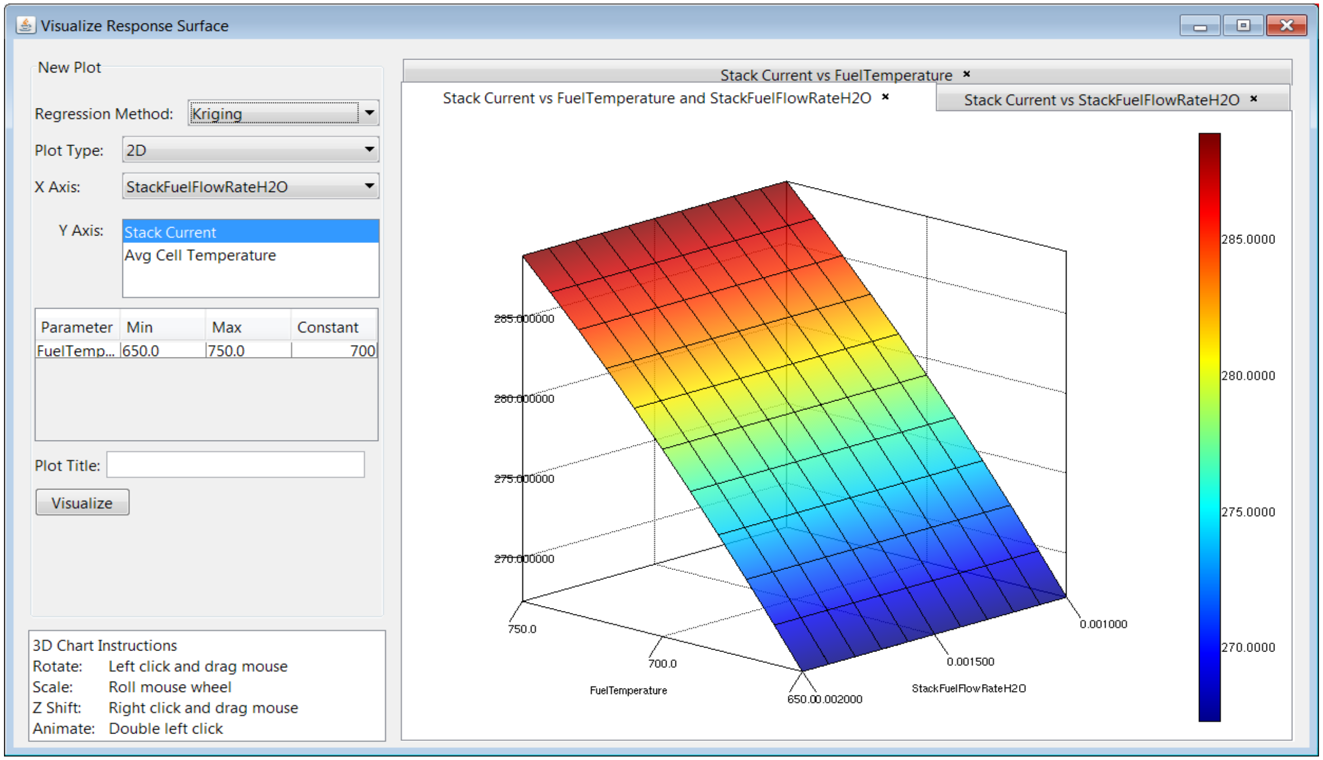
IGFC Power System

Reduced Order Model (ROM) Approach for SOFC Stacks



SOFC-ROM: Visual Analysis Tools

- Rich menu-driven plotting capabilities to aid understanding
  - visualize 2D-3D response surfaces
  - plot sampling space
  - actual vs. predicted values
  - output vs. input values
  - output vs. output values
  - sensitivity charts
  - error histograms
  - results as a function of S/C ratio



Example: Amount of On-Cell Reforming

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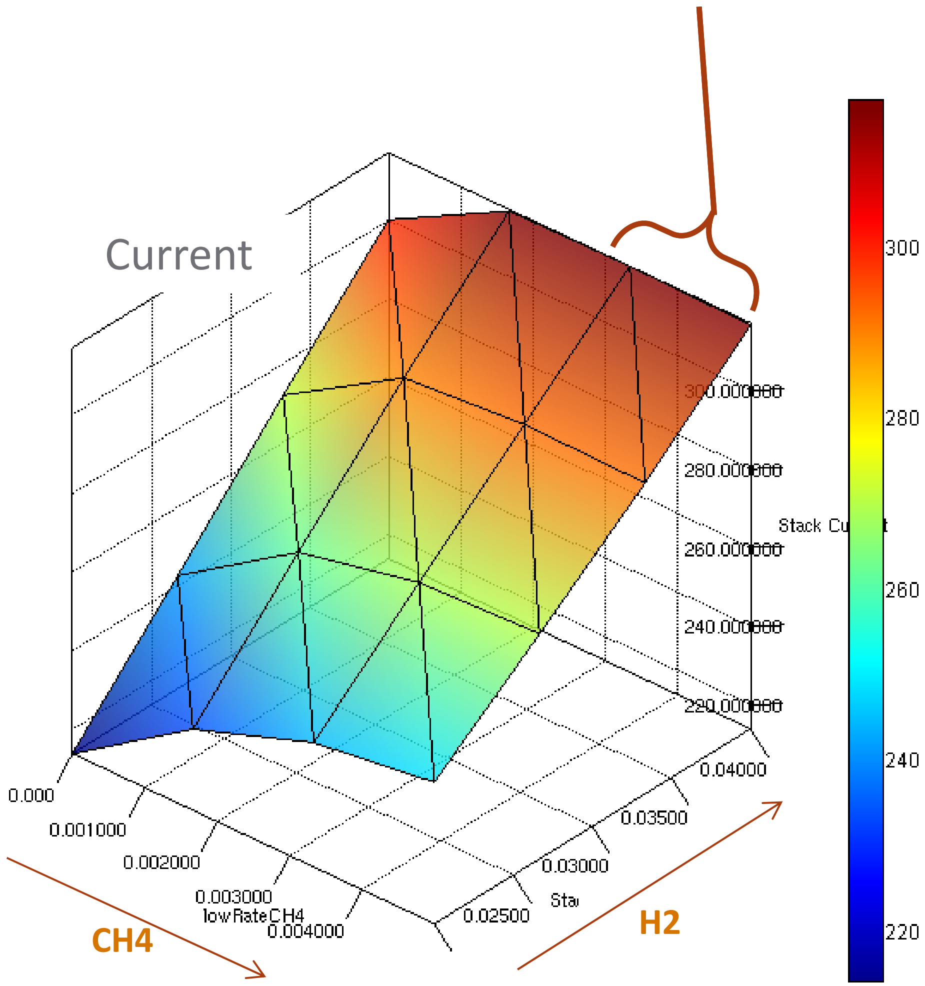
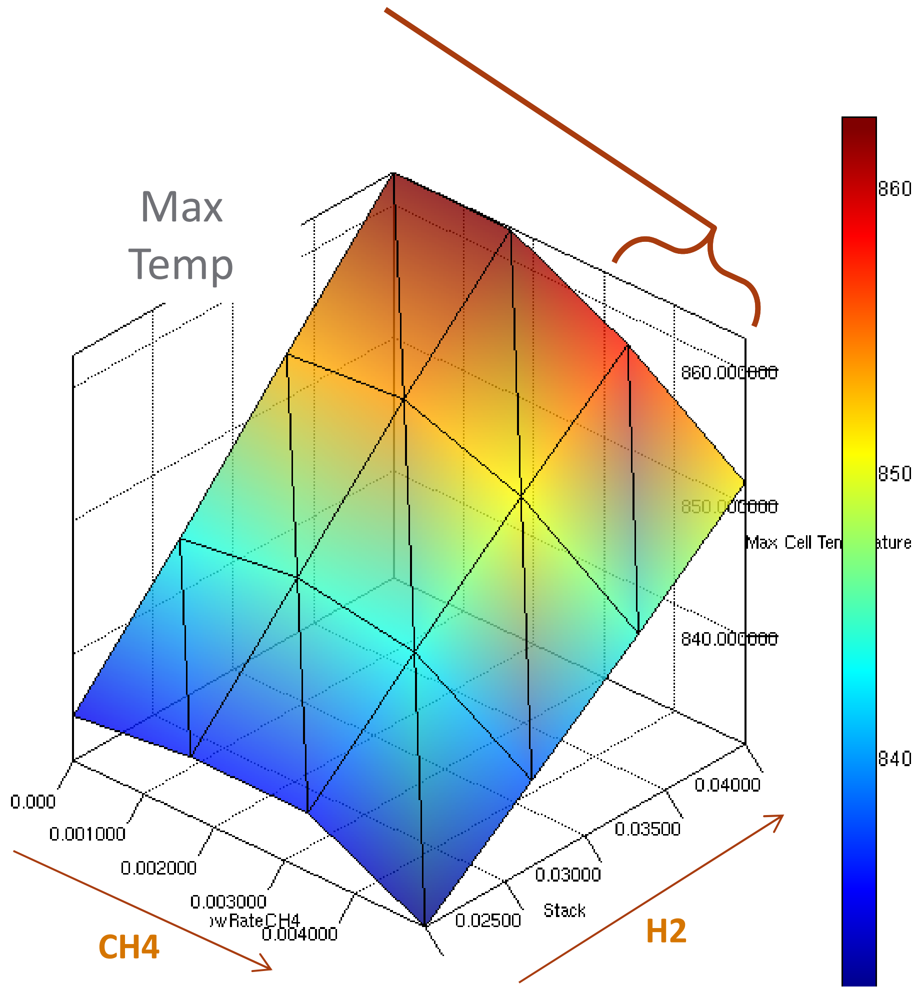
- Region with high current density and reduced temperature identified





Example: Stack Peak Temperature Control

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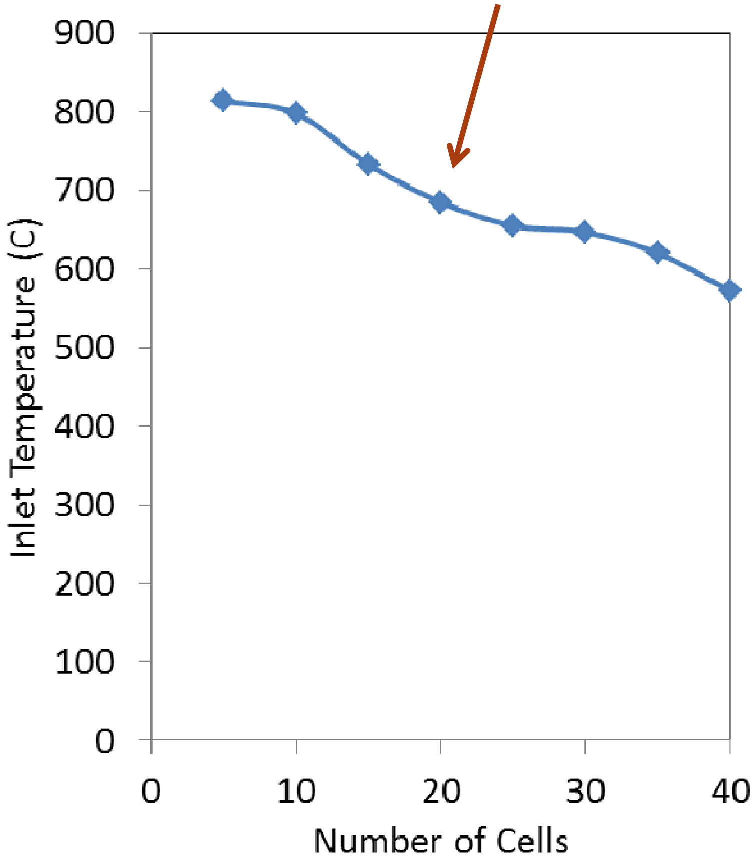
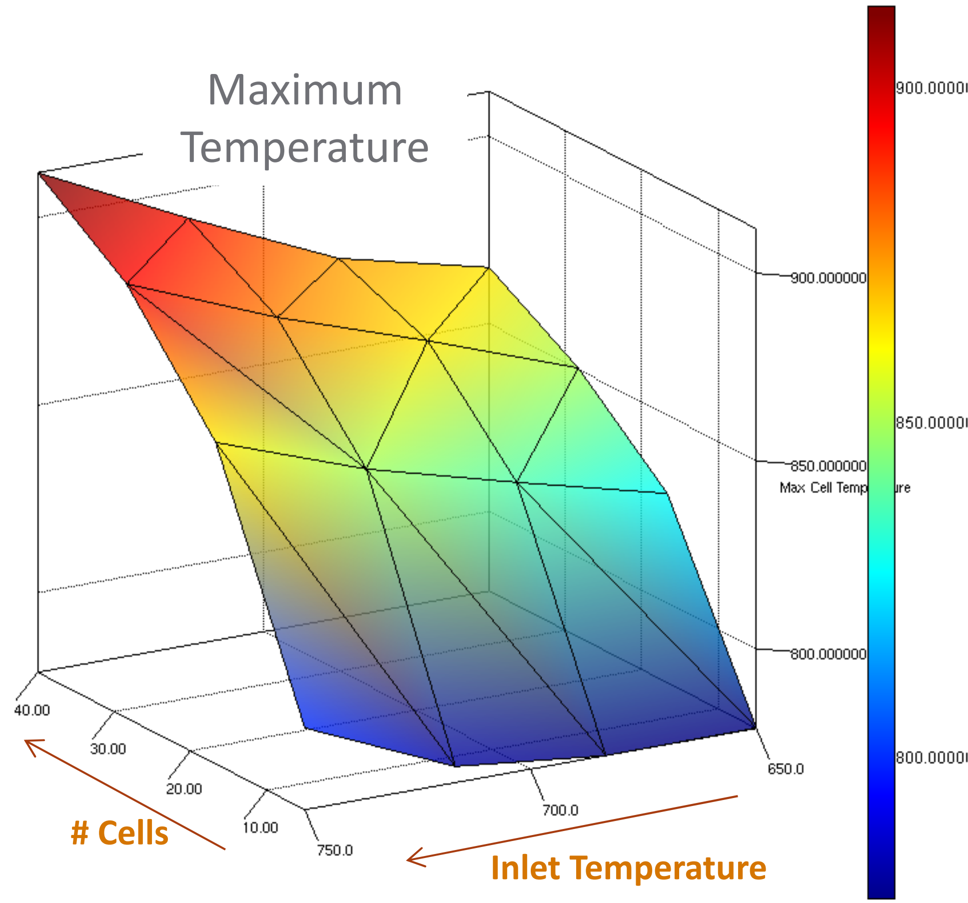
- Evaluate stack maximum temperature as the stack size increases
- E.g., identify required inlet temperature to ensure T_{max} < 850°C
  - Use exported ROM





Summary for SOFC-ROM

- Implemented data validation tools
  - Checks predicted values from a created ROM against actual values to ensure ROM is suitable
- Implemented scalable error estimation
  - Previous 1-off estimation unsuitable for large number of sampled cases and replaced with 20/80 cross validation approach to test multiple parameter simultaneously
- Implemented scalable visualization
  - Pre-computation of data for rapid plotting replaced with dynamic generation for 2D or 3D graphs
- Planned initial implementation is complete

- Have initiated collaborative testing with BAH systems modelers to demonstrate utility for modeling of SOTA SOFC-based power systems

Seal Modeling Task Overview

- **Challenge**:
  - Seals must remain hermetic for stack operating lifetime
- **Goal**:
  - Develop quantitative models to capture the thermo-mechanical behaviors of the sealant glass materials and examine the durability of the compliant sealants under SOFC stack operation conditions
- **Technical Approach**:
  - Develop constitutive models to resolve the thermo-visco-elastic-damage-healing material behaviors of the sealant glass
  - Use multi-scale modeling approaches to bridge the intrinsic material characteristics of the glass and its thermo-mechanical properties
- **Accomplishment**:
  - Completed model development and stack simulations to evaluate sealing performance under different operating conditions including thermal cycling

Modeling of the compliant SCN-1 glass

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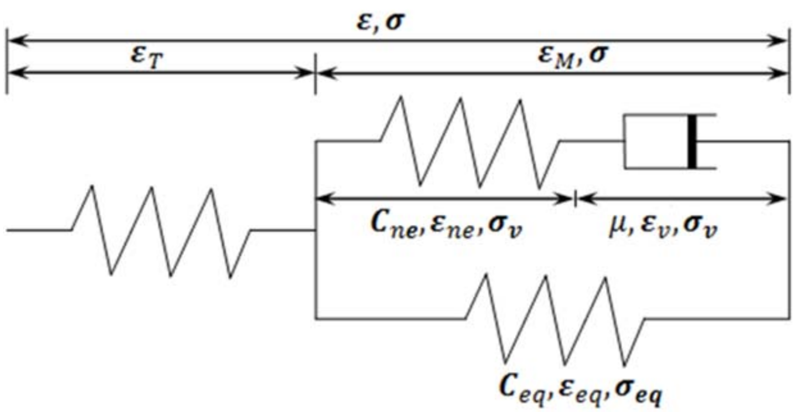
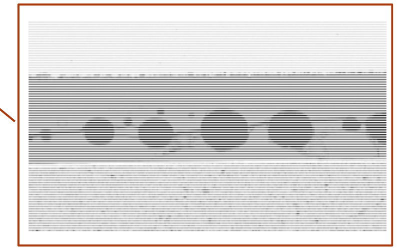
- Constitutive thermo-visco-elastic-damage-healing model

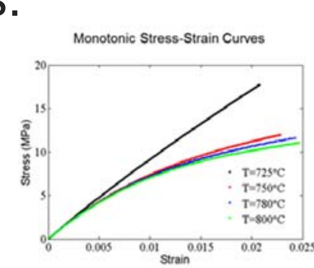


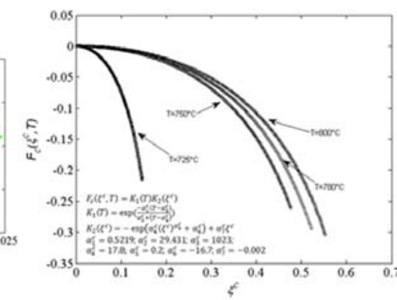
"damage" is considered to be the fractional reduction in elastic modulus and formulated as $\dot{\varphi} = \dot{\varphi}_c + \dot{\varphi}_p$

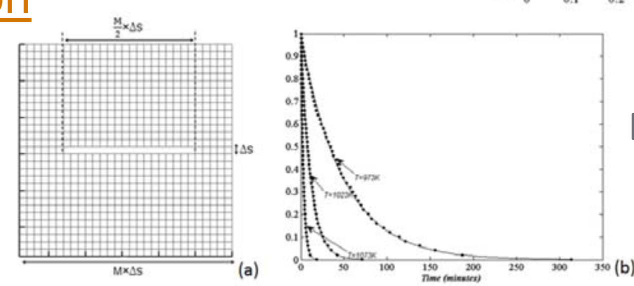


- Physically-driven damage and healing kinetics determined through lower-length scale simulations and experiments:
  - Pressure driven crack nucleation
  - Energy driven crack growth
  - Thermal diffusional crack healing
  - Hydrostatic-stress induced pore nucleation
  - Inelastic flow induced pore growth

Crack propagation







Diffusional healing

Xu W., X Sun, BJ Koeppel, H Zbib 2014. International Journal of Plasticity doi:10.1016/j.ijplas.2014.06.011

Finite Element Model of SOFC stack

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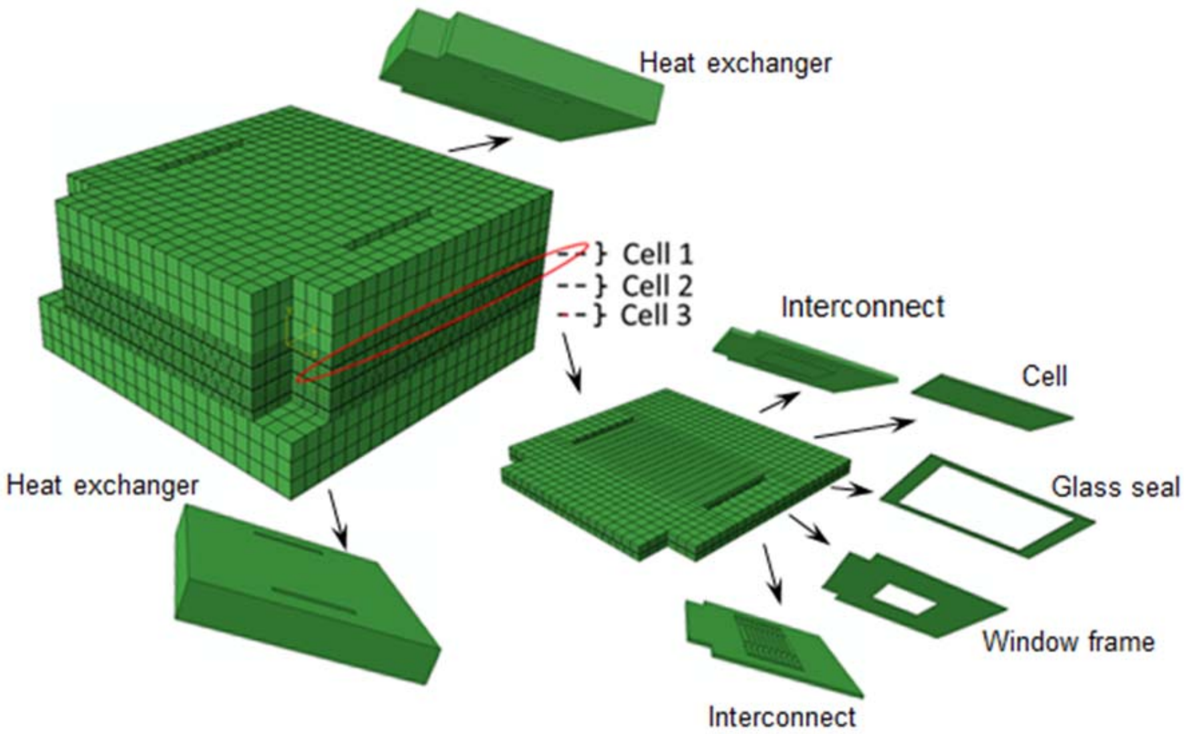
- PNNL's SECA Core Technology Program stack test fixture



- Finite element analyses performed using ABAQUS
- Compliant seal bonds the cell to the metallic frame
- Compliant glass material model implemented in the stack simulation through subroutines

- Similar finite element models have also been created for larger planar designs that more closely resemble full-sized SECA cells and stacks.

Reliability of Multi-Cell SOFC Stack During Multiple Thermal Cycles

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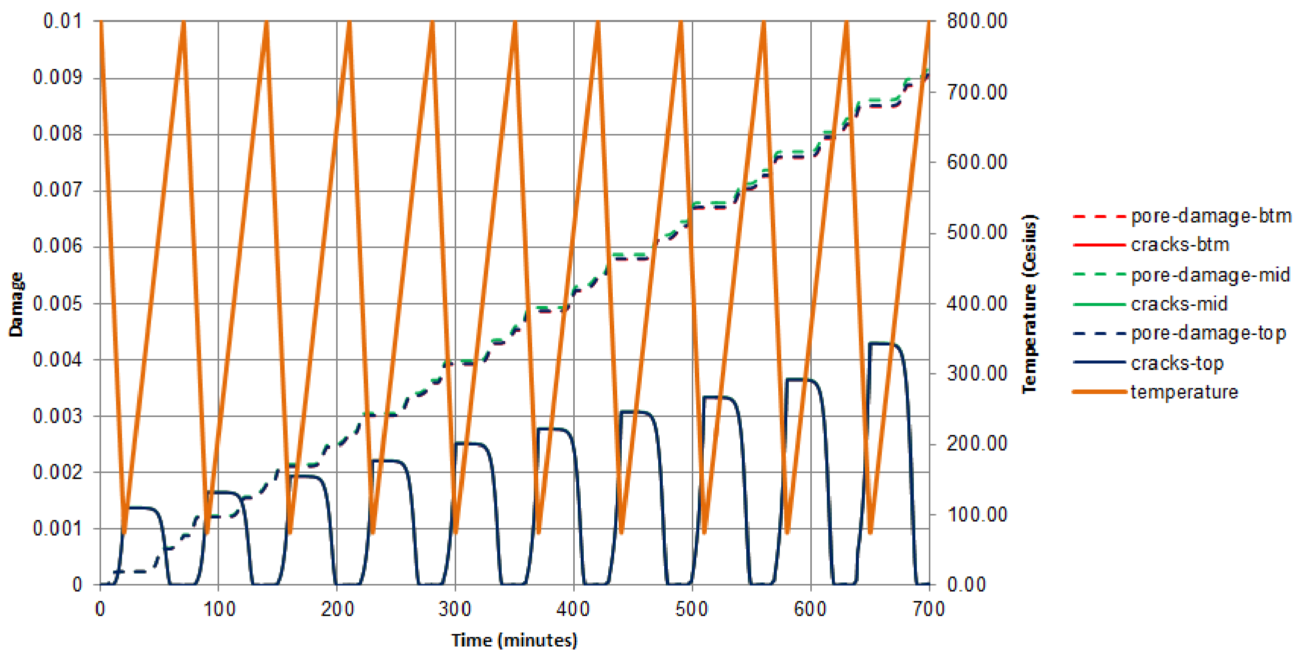
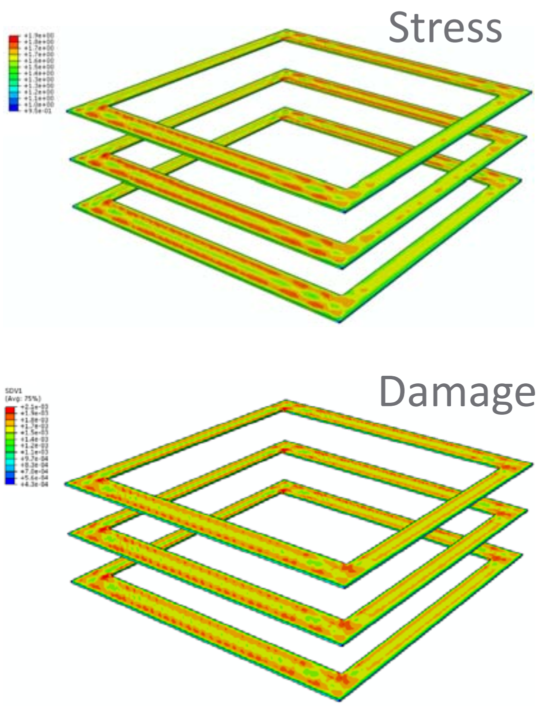
- Mechanical integrity of the glass seal is minimally impacted by 10 deep and rapid thermal cycles





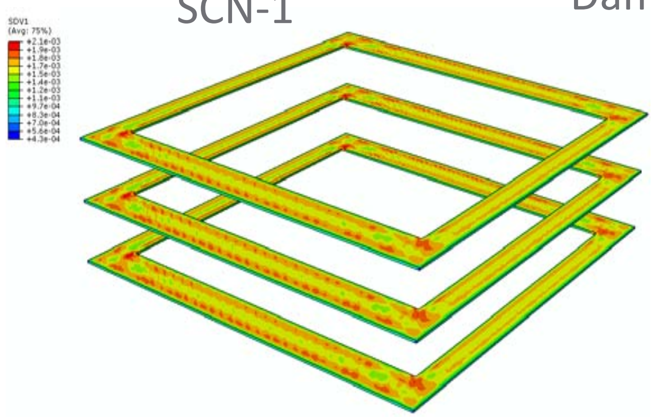
- Stress and damage distributions within the three seals (from top to bottom) are almost the same; crack initiation is increased by accumulated pore-related damage which appears to saturate over time

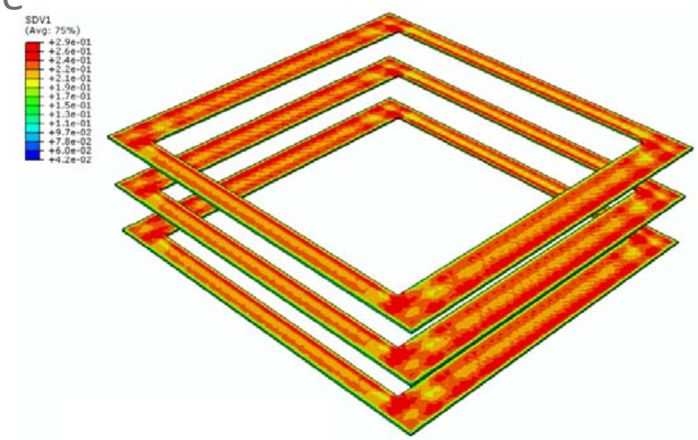
Comparison of Mechanical Responses Between Sealant Glass Materials

- Compliant SCN-1 glass accumulates much less damage because of timely stress relaxation and its healing capability upon heating

SCN-1

Damage



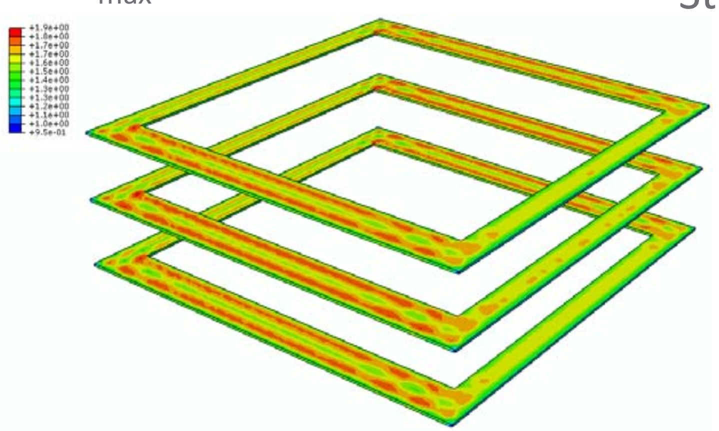


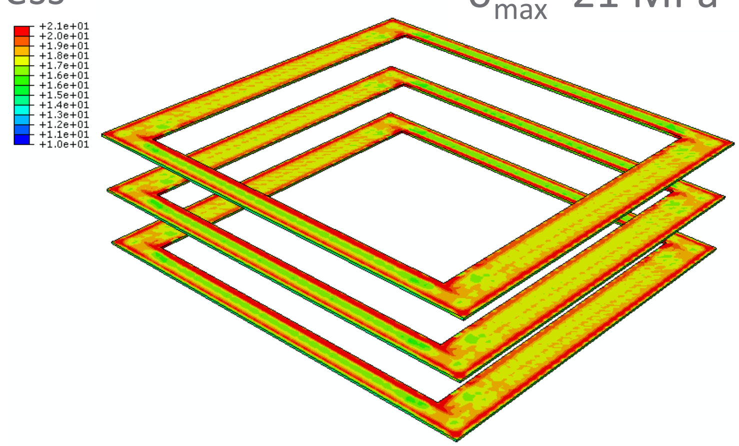
G18: A state-of-the-art devitrifying glass (Nguyen et al., 2006 and Stephens et al., 2009)

σ_{max}=1.9 MPa

Stress

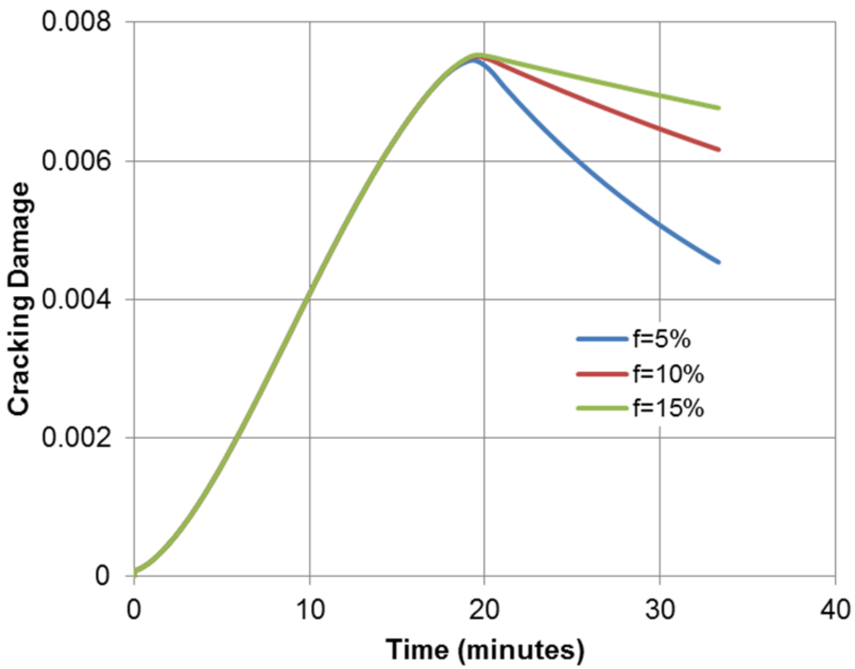
σ_{max}=21 MPa





Effects of Ceramic Fillers on Healing (Stack Simulation)

- Healing time will increase with fiber volume fraction



$$\dot{\varphi}_h = -h_1 \exp(-h_2 f) \left(\frac{T}{T_g}\right)^{h_3} \varphi_h$$

h_1=4E-4 h_2=0.2 h_3=28.5

- Required cooling/heating rates and durations during thermal cycling to sufficiently heal cracking damage can be determined

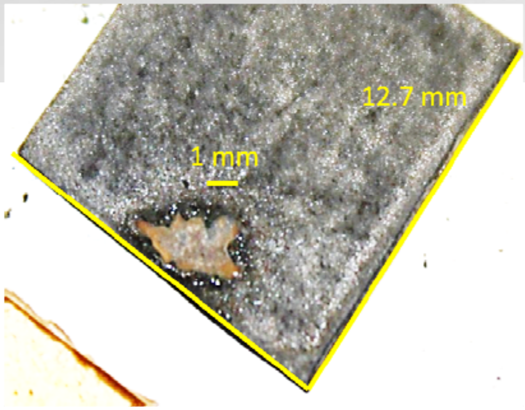
Note: here the existence of ceramic fillers is effectively considered through the activation energy of the healing probability function. In order to explicitly resolve the interaction between the reinforcement phase and the glass matrix as well as to establish a more generic description/prediction of the influence of the fillers, high resolution lower-length scale model is needed.

Summary for Seal Modeling

- Simulation of the compliant seal material suggests that it a viable design option to consider for stack sealing
  - Cracking damage from thermal-cycling can be completely healed
  - Pore damage occurs but is small and can be mitigated with fillers

- Description and capabilities of the model developed in this task are described in a recent summary report and journal article
  - YS Chou, JP Choi, W Xu, EV Stephens, BJ Koeppel, JW Stevenson, E Lara-Curzio (2014) Compliant Glass Seals for SOFC Stacks, PNNL-23397.
  - W Xu, X Sun, E Stephens, I Mastorakos, MA Khaleel, HM Zbib (2012). “A Mechanistic-Based Healing Model for Self-healing Glass Seals Used in Solid Oxide Fuel Cells,” *Journal of Power Sources* 218:445-454.
  - W Xu, X Sun, BJ Koeppel, HM Zbib (2014). “A Continuum Thermo-Inelastic Model for Damage and Healing in Self-Healing Glass Materials,” *International Journal of Plasticity* (in press).

Mechanical Reliability and Life Prediction of Coated Metallic Interconnects



- **Challenge**:
  - IC must meet SECA lifetime requirement
- **Goal**:
  - Use modeling to predict interconnect life under isothermal cooling and quantitatively estimate the effect of materials and geometry parameters on the interconnect life
- **Technical Approach**:
  - Develop a combined analytical/numerical approach based on the theory of bucking driven blistering to related the interface strength to the life prediction
- **Accomplishment**:
  - Evaluated the predicted statistical lifetime as a function of scatter in the strength measurements

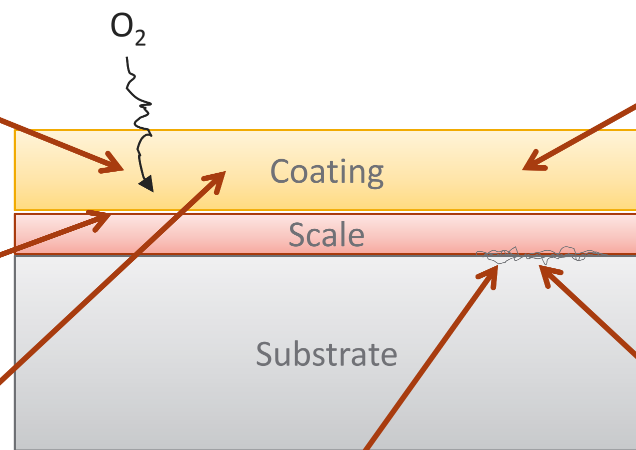
Contributions to Long IC Lifetime

- Different design features contribute jointly to mitigate degradation mechanisms and ensure long lifetime of the IC protection system

Coating **restricts O_2 diffusion** to reduce scale growth rate

Coating **compatibility** provides very strong coating-scale interface bond

Coating **CTE matched** with substrate to minimize thermal stresses



Coating stiffness provides **mechanical support** to hinder blister buckling

Substrate surface modifications increase **interfacial strength** and **crack resistance** to prevent scale-substrate delamination

Substrate surface modifications increase scale adhesion post-delamination by **mechanical interlocking**

IC Lifetime Analysis Methodology

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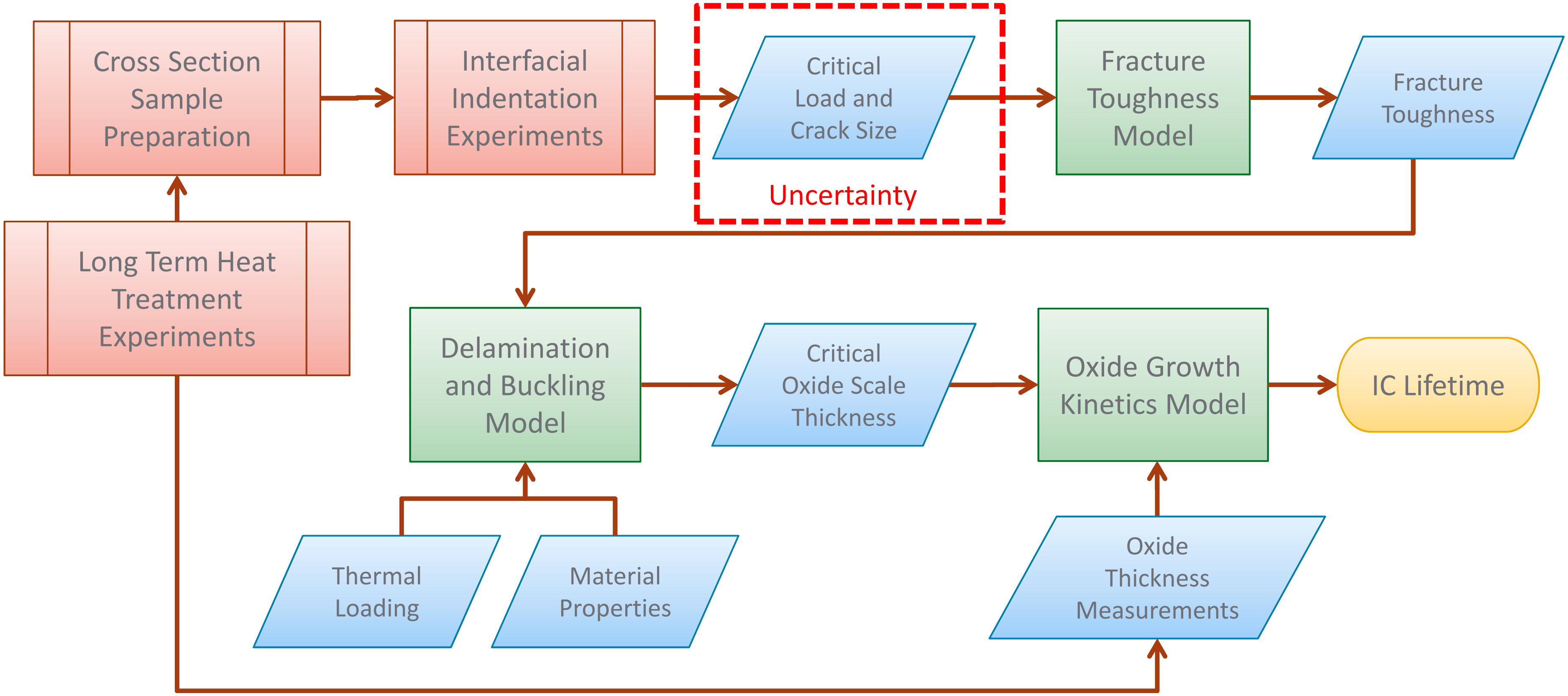
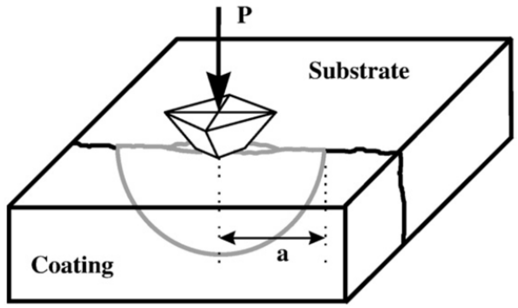
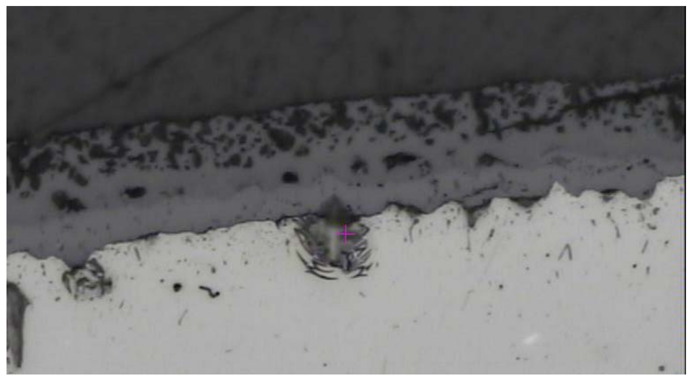
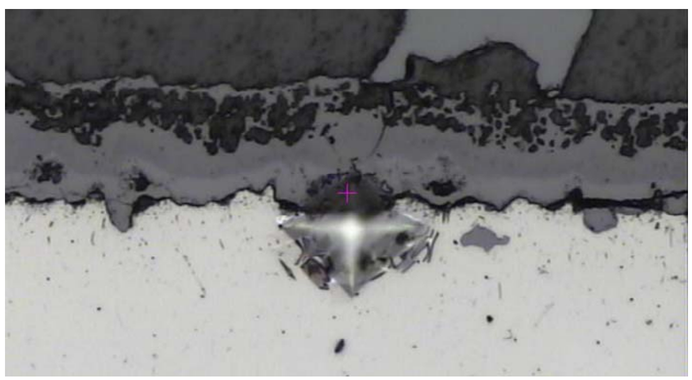
- Developed technical approach using experiments and modeling
  - Uncertainty analysis to capture scatter in the experimental data

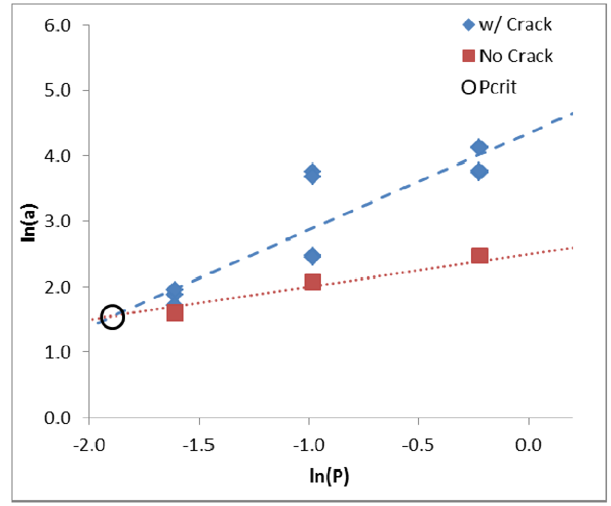


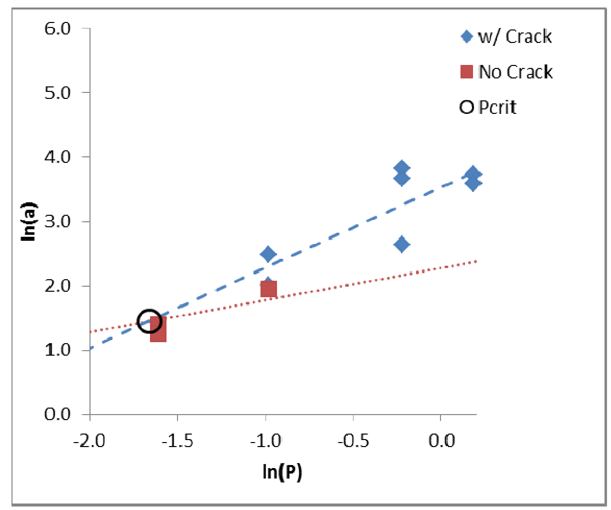
- Specimen Conditions
  - 800-850°C
  - 2k,10k, 14k, 20k, 26k, 30k hours
  - Surface blast (SB) and surface grind (SG)
- Plot crack size versus load











2) Uncertainty in Critical Indentation Load

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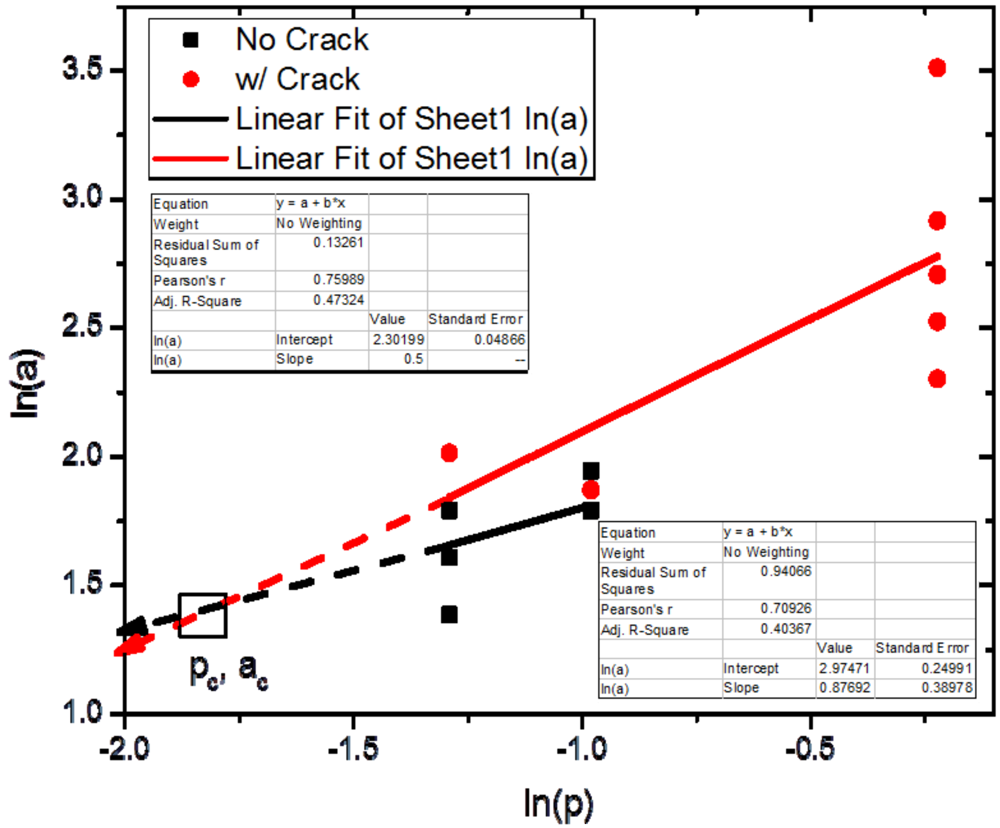
- For the indent where a valid crack propagates along the interface, the log of the average crack size is plotted versus the log of the applied load
- For all indents with or without a crack, the log of the indent diagonal is plotted versus the log of the applied load (i.e., hardness)
- Linear regression is then used for each set of data

$$HV = 1.8544\frac{P}{a^2}$$



	Intercept (mean)	Intercept (std dev)	Slope (mean)	Slope (std dev)
No Crack	2.302	0.049	0.5	0
W/ Crack	2.975	0.250	0.877	0.390

3) Uncertainty in K_I

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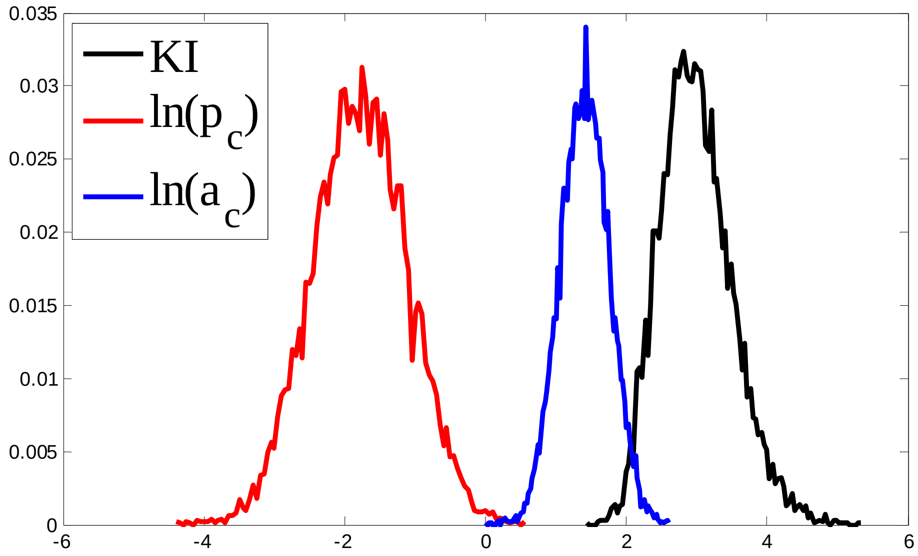
- Using the mean and standard deviation statistics from the linear fits, determine the distribution for the interface toughness
- Perform 1000 Monte Carlo simulations
- Determine the probability distribution function (PDF) of $\ln(P_c)$, $\ln(a_c)$, and K_I

$$K_{in} = 0.015 \frac{P_c}{a_c^{3/2}} \left(\frac{E}{H}\right)_I^{1/2}$$

	$\ln(P_c)$	$\ln(a_c)$	K_I
mean	-1.78	1.41	2.99
std dev	0.68	0.35	0.51



4) Uncertainty in h_c

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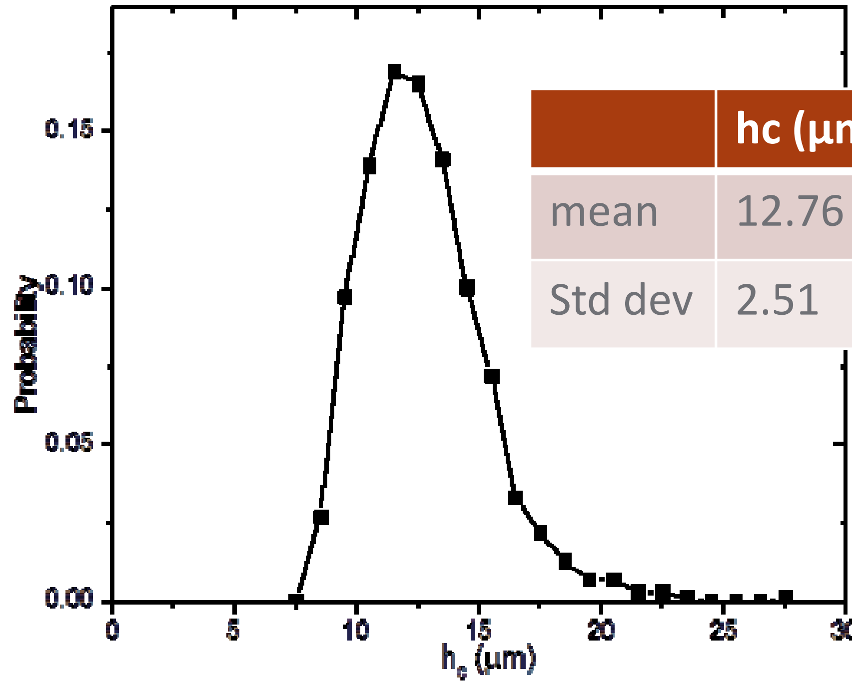
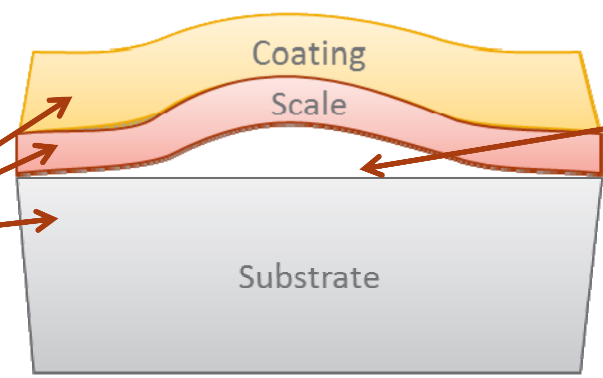
- Assume linear elastic materials with constant thermal expansion for three layers
- Next, evaluate the critical thickness h_c and its uncertainty with derived distribution information on K_I.



	hc (μm)
mean	12.76
Std dev	2.51

Mechanical Properties
E, υ, α

Thicknesses
h



Interface Toughness
K_I

Temperature Change
ΔT

5) IC Life from Oxidation Kinetic Curves

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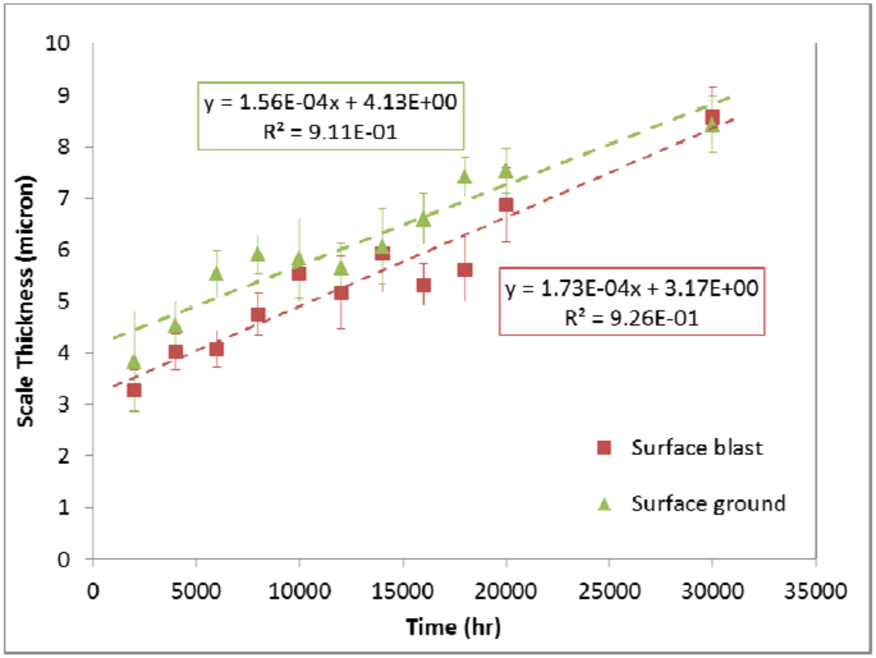
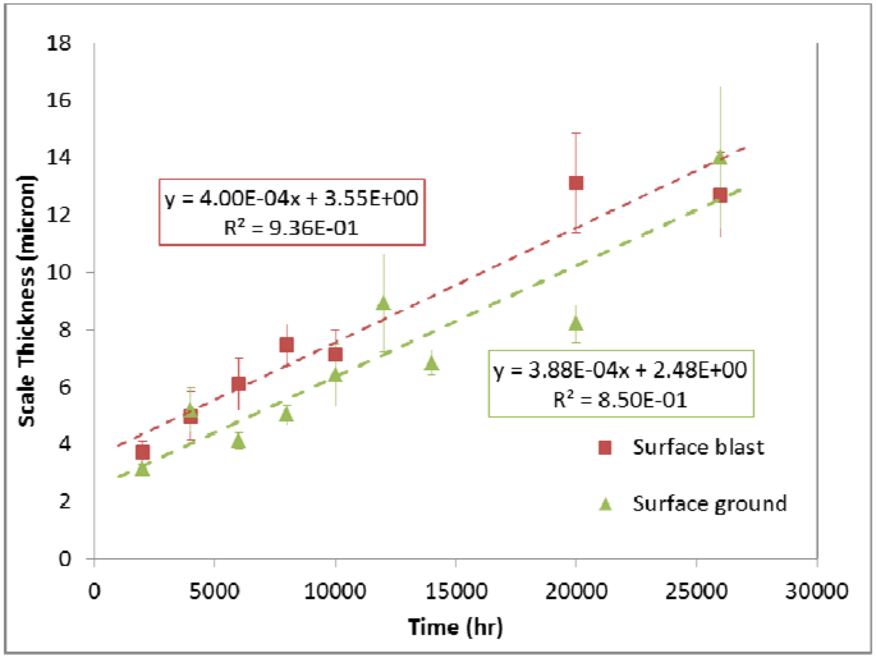
- From h_c, IC life can be identified from the oxidation kinetic curves.
  - Long term oxidation found to better correlate with linear fit than parabolic fit
- By this way, IC life can be quantitatively predicted, and the effect of interface toughness, coating property, etc. can be systematically investigated.

Scale Growth History at 800°C



Scale Growth History at 850°C



Probability Density and Cumulative Distributions of Material Lifetime

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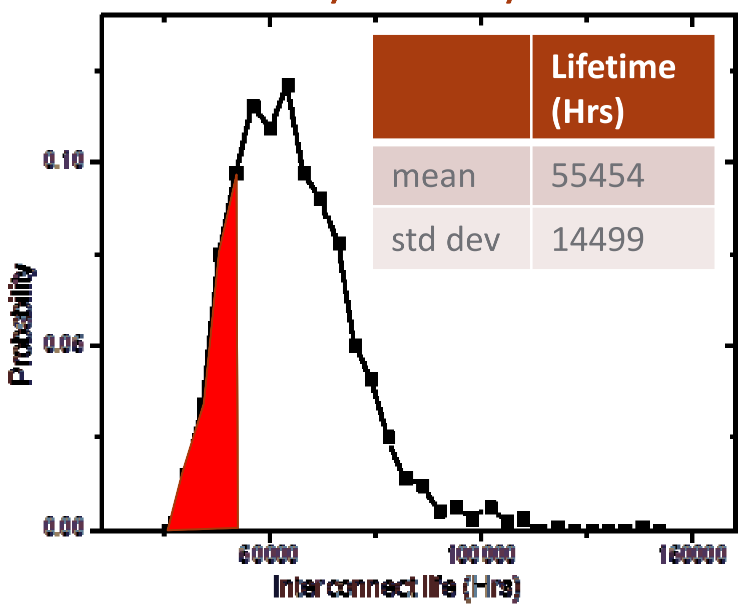
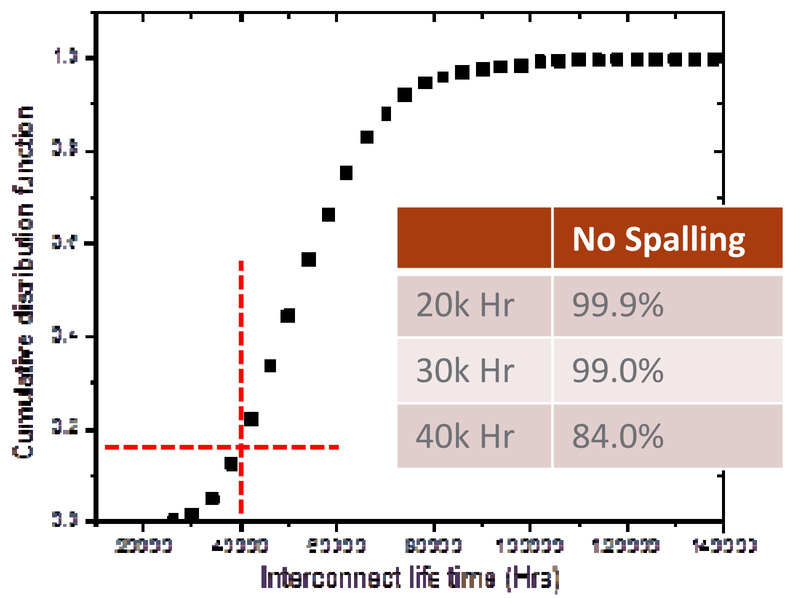
- Using the experimentally determined oxide growth rate and the K_{IC} distribution, the predicted probability density function for the expected lifetime is obtained
- This is integrated to determine the cumulative distribution function

Probability Density Function



	Lifetime (Hrs)
mean	55454
std dev	14499

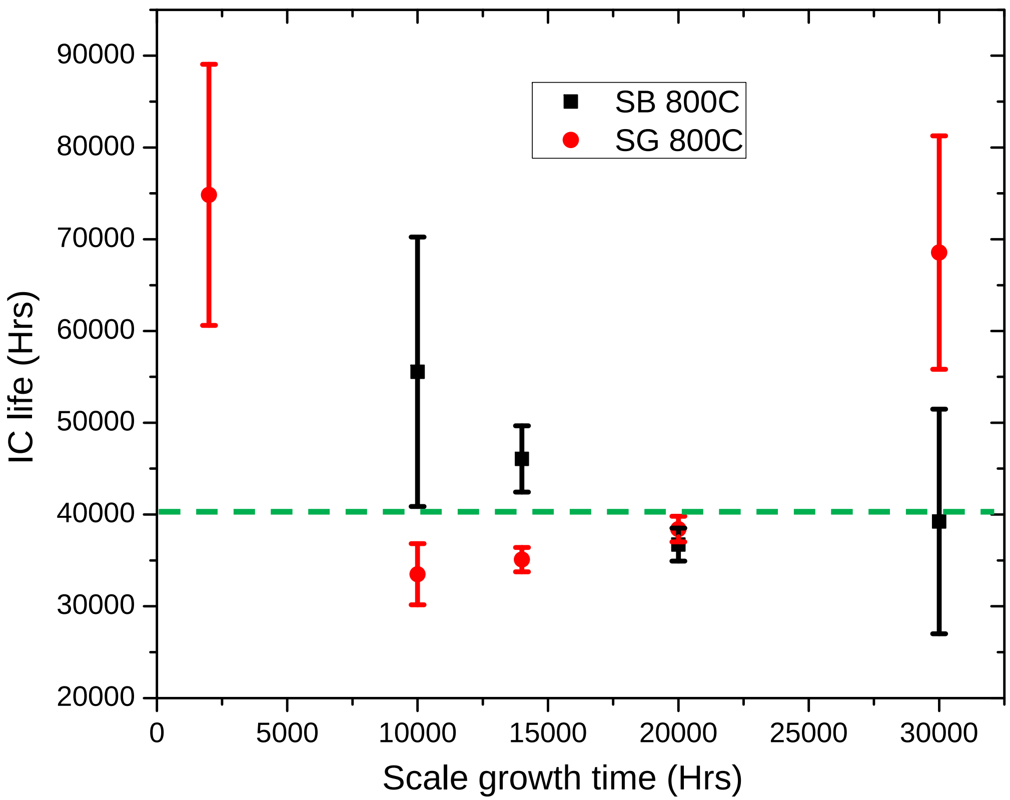
Cumulative Distribution Function



	No Spalling
20k Hr	99.9%
30k Hr	99.0%
40k Hr	84.0%

Initial Lifetime Estimates

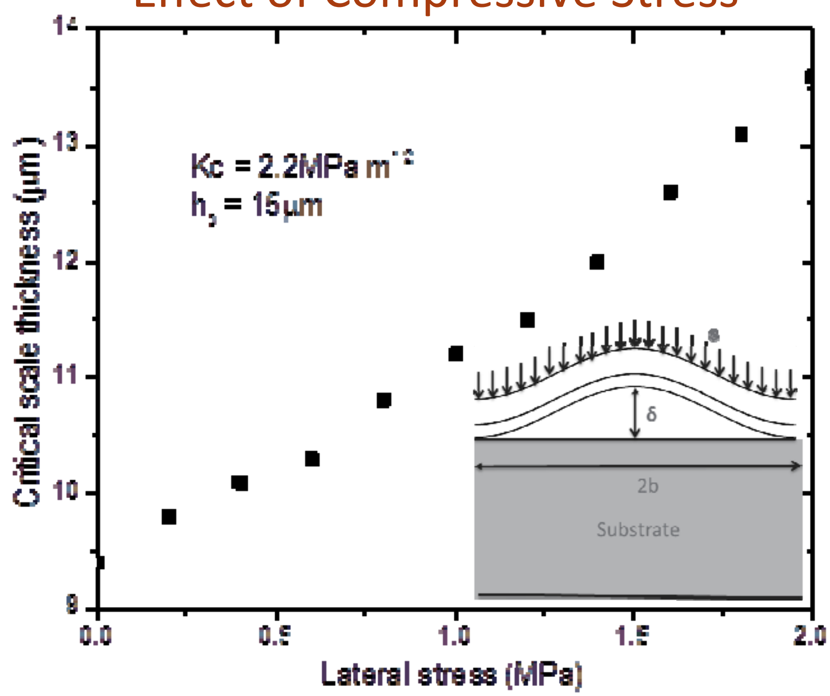
- Data scatter
- Still evaluating whether this is due to the methodology or the intrinsic variability of the specimens themselves
- SB is generally better than SG, but SG has much wider range of estimated strength
  - Observations of cross-sections suggest that the grinding is non-uniform
- Assuming the coupon results are representative of the entire IC, mean lifetimes of 34-57k hr at 800°C for SB specimens



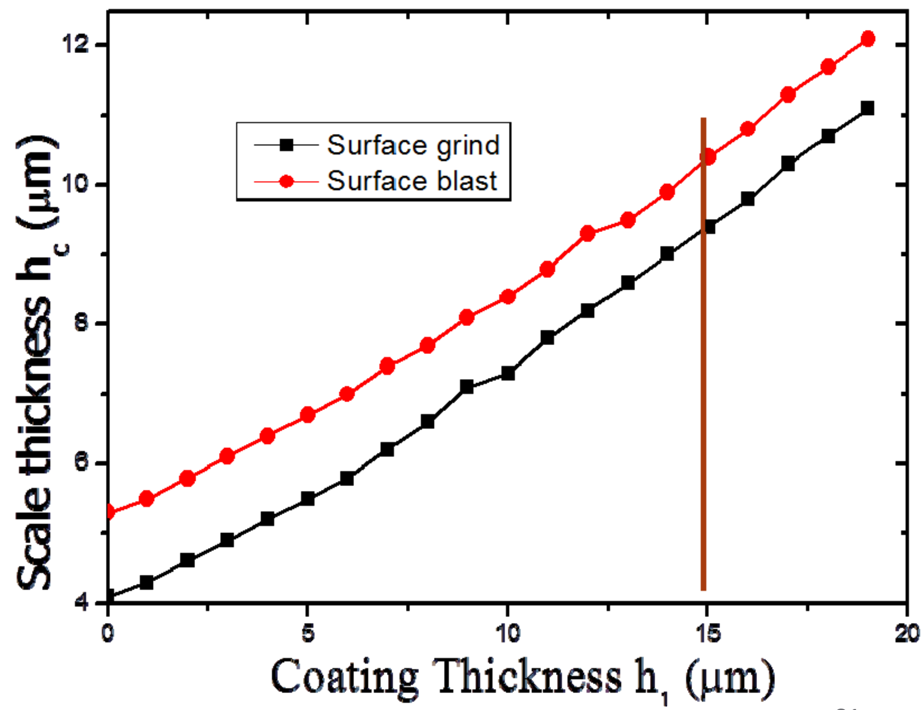
Sensitivity of Critical Scale Thickness

- Evaluated effect of various parameters on critical oxide thickness h_c
- Mild compressive stress on the interconnect increases the h_c
  - Areas under stack preload will be more resistant to delamination
- The coating itself acts a mechanical support for the scale to resist buckling, so thicker coating is beneficial to resist this failure mode

Effect of Compressive Stress



Effect of Coating Thickness



Summary for Interconnect Modeling

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- The uncertainty in the IC life prediction was quantitatively derived from the variability in the indentation experiments
- The sensitivity of the critical oxide thickness to various other design parameters were evaluated
- Trends in the IC lifetime evaluations are still being investigated, but the SB surface modification is more uniform than SG which showed wide variability in strength

Ongoing/Proposed Modeling Activities

- Complete IC analysis
- Continue work with BAH/NETL modelers to implement and test a reduced order model made by the SOFC-ROM tool in IGFC/NGFC Aspen system models
- Perform mechanical reliability study of realistic state-of-art planar stack components and interfaces for different operating conditions and amount of on-cell reforming using SOFC-MP and FEM tools
- Simulate new contact material fabrication methods and engineered surface textures to improve contact layer bulk and interfacial strength
- Evaluate effects of residual stresses from contact layer fabrication on overall stack mechanical reliability

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