AN INTEGRATED APPROACH TO MODELING AND MITIGATING SOFC FAILURE

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Outline

- First Order Failure Criteria for SOFC PEN Structure
- Creep Modeling of YSZ/Ni Cermet
- Fracture Mechanics Analysis Tool
- Thermal Transient Modeling

First Order Failure Criteria for SOFC PEN Structure

- Objectives
- Local Failure Criteria
  - Failure Modes
  - Strength Failure Criteria
  - Fracture Failure Criteria
- Global Failure Criteria
- Analyses for Various Crack Cases
- Conclusion

Objectives

Develop first-order failure criteria to be used for the initial design, material selection and optimization against thermomechanical failure of the PEN structure in high temperature SOFCs.

Failure Modes

Material Characteristics

- Static Strength
- Fracture Toughness
- Fatigue Strength

Does a material contain flaws above certain threshold value?

No -> Failure is strength-controlled

Yes -> Failure is fracture toughness-controlled

Strength-Based Failure Theory

Failure occurs when

$$\bar{\sigma} = f(\sigma_1, \sigma_2, \sigma_3) = \sigma_f$$

where

$\bar{\sigma} = f(\sigma_1, \sigma_2, \sigma_3)$	Effective Stress
$\sigma_1, \sigma_2, \sigma_3$	Principle Stresses
σ_f	Material Strength

Fracture-Based Failure Theory

Fracture occurs when

$$G = \frac{1-\nu^2}{E}\left(K_I^2 + K_{II}^2 + \frac{K_{III}^2}{1-\nu}\right) = G_c$$

where

G	Energy Release Rate
$K_I \quad K_{II} \quad K_{III}$	Stress Intensity Factors
G_c	Fracture Toughness

YSZ Electrolyte

Maximum Normal Stress Criterion ⟺ $\bar{\sigma} = \sigma_f$

$$\bar{\sigma} = f(\sigma_1, \sigma_2, \sigma_3) = \max\left\{|\sigma_1|, |\sigma_2|, |\sigma_3|\right\}$$

$$\sigma_f = 100 \sim 300\ \text{MPa}$$

Fracture Criterion

$$G = \frac{1-\nu^2}{E}\left(K_I^2 + K_{II}^2 + \frac{K_{III}^2}{1-\nu}\right) = G_c$$

$$G_c = 7.8 \sim 13.7\ \text{J/m}^2$$

YSZ/Ni Cermet

Von Mises Criterion (elevated temp) ⟷ $\bar{\sigma} = \sigma_f$

$$\bar{\sigma} = \sqrt{\left(\sigma_x - \sigma_y\right)^2 + \left(\sigma_y - \sigma_z\right)^2 + \left(\sigma_z - \sigma_x\right)^2 + 2\tau_{xy}^2 + 2\tau_{yz}^2 + 2\tau_{zx}^2}$$

Maximum Normal Stress Criterion ⟷ $\bar{\sigma} = \sigma_f$

$$\bar{\sigma} = f(\sigma_1, \sigma_2, \sigma_3) = \max\left\{\left|\sigma_1\right|, \left|\sigma_2\right|, \left|\sigma_3\right|\right\}$$

$$\sigma_f = \sigma_{YSZ}\left[V_{YSZ} + \frac{E_{Ni}}{E_{YSZ}(1-\nu_{Ni})}\left(1 - V_{YSZ} - V_{Void}\right)\right]$$

$\sigma_{YSZ} = 100 \sim 300\text{MPa}$ = YSZ tensile strength

E_{Ni} = Ni Young's modulus

E_{YSZ} = YSZ Young's modulus

ν_{Ni} = Ni Poisson's ratio

V_{YSZ} = YSZ Volume fraction

V_{Void} = Void Volume fraction

Global Failure Criteria

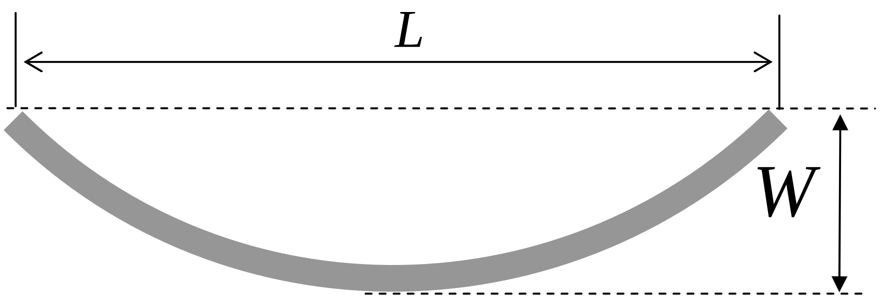
Processing → Warpage → Stack Assembly → Fracture

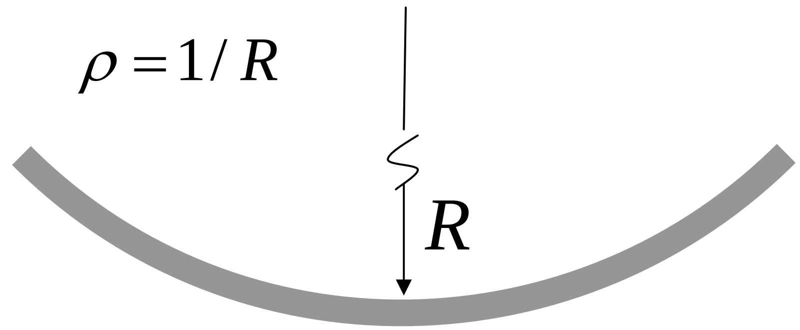
Warpage Criterion

$$W < W_c$$

Curvature Criterion

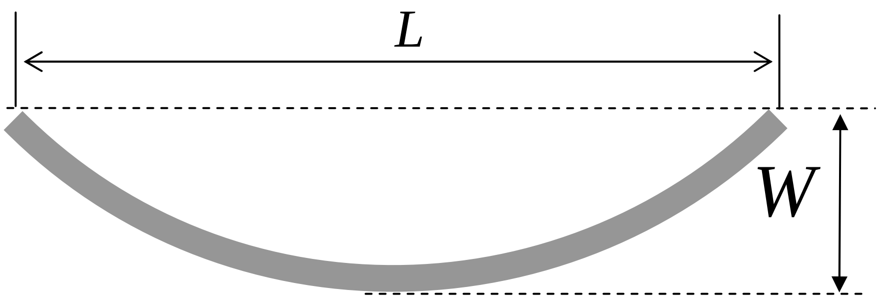
$$\rho < \rho_c$$

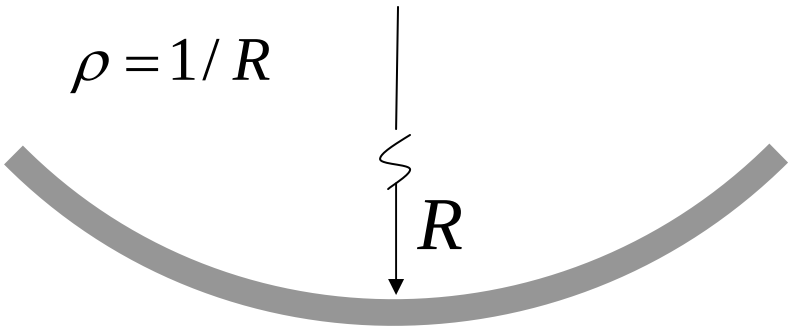




Implementation

- Based on material/geometry parameters to compute W_c and ρ_c
- Measure W or ρ of each cell after sintering
- Compare the measured W with W_c or ρ with ρ_c





Crack Types

A – crack in the cathode

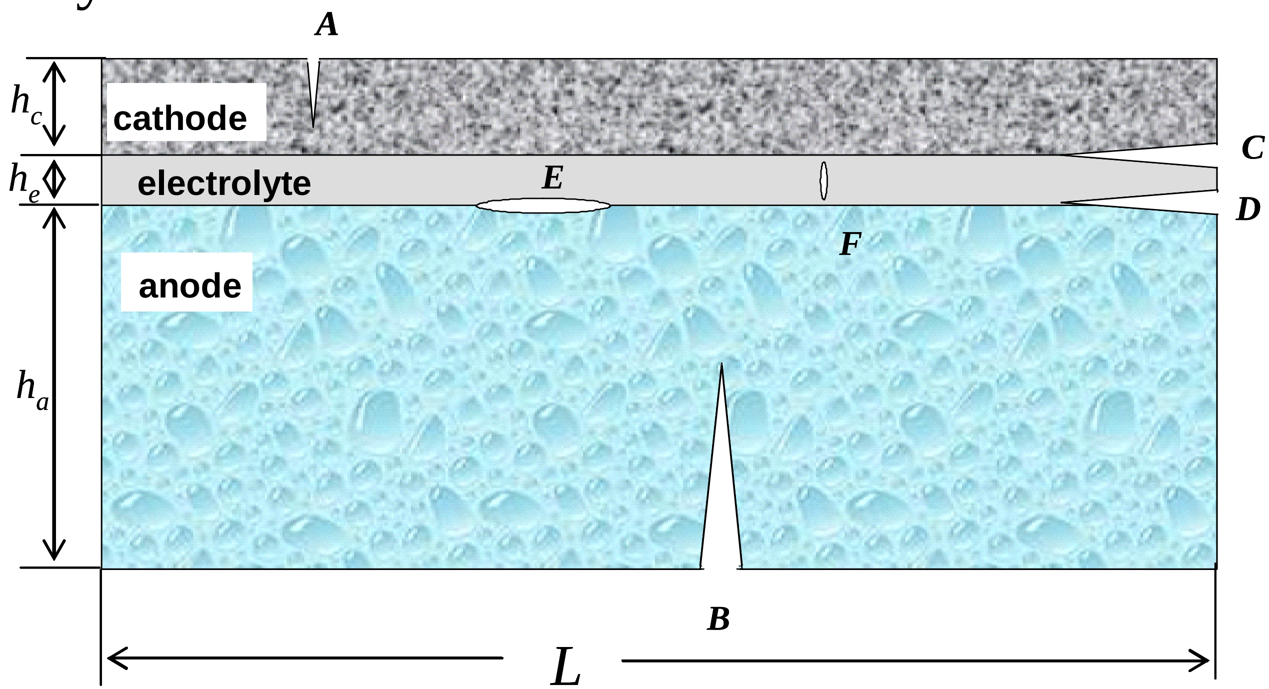
B – crack in the anode

C – delamination crack between the cathode and electrolyte

D – delamination crack between the anode and the electrolyte

E – blister crack on the anode/electrolyte interface

F – crack in the electrolyte



Max. Allowable Warpage

$$\frac{W_c}{L} = Y\sqrt{\frac{G_c}{h_e E_e}\left(\frac{L}{h_e}\right)}$$

G_c = fracture toughness
h_e = electrolyte thickness
E_e = modulus of electrolyte

Crack *A*	$Y = \left(\frac{h_2^3 E_2}{16 H^2 E_3}\right)^{1/2}\left[\left(\frac{\Delta\alpha}{Q(1-\nu_3)}\right)^2 + \left(t_4 - \frac{a}{2}\right)^2\right]^{-1/2}$
Crack *C*	$Y = \left(\frac{h_2^3 E_2}{16}\right)^{1/2}\left(\frac{c_3 F_2}{16 h_3^3} Q_1^{-2} + \frac{4 h_3 (\Delta\alpha_2)^2 F_1}{c_3} Q^{-2}\right)^{-1/2}$
Crack *D*	$Y = \left(\frac{h_2^3}{16\pi a E_2}\right)^{1/2}\left[\left(\frac{\Delta\alpha}{Q(1-\nu_2)}\right)^2 + \left(\frac{h_1 - h_3}{2}\right)^2\right]^{-1/2}$
Crack *E*	$Y = \left(\frac{h_2^3 E_2}{16 h_2}\right)^{1/2}\left(\frac{c_{ce} F_2}{16 h_{ce}^3} Q_1^{-2}\rho^2 + \frac{4 h_{ce} (\Delta\alpha)^2 F_1}{c_{ce}} Q^{-2}\right)^{-1/2}$
Crack *F*	$Y = \frac{Q h_2 \sqrt{h_2 E_2}}{4\Delta\alpha}\sqrt{\left(\frac{1}{P_1} + \frac{P_1 P_2^2}{G_c}\right)}$

Implementation

Definition of Variables:

- ✓ See our monthly report or e-mail jianmin.qu@me.gatech.edu

Basic Assumptions:

- ✓ Linear elastic fracture mechanics

Implementation:

- ✓ A FORTRAN code

Material Properties Needed:

- ✓ Elastic moduli
- ✓ Coefficient of thermal expansion
- ✓ Fracture toughness

Other Parameters needed:

- ✓ Layer thickness
- ✓ Warpage (curvature)

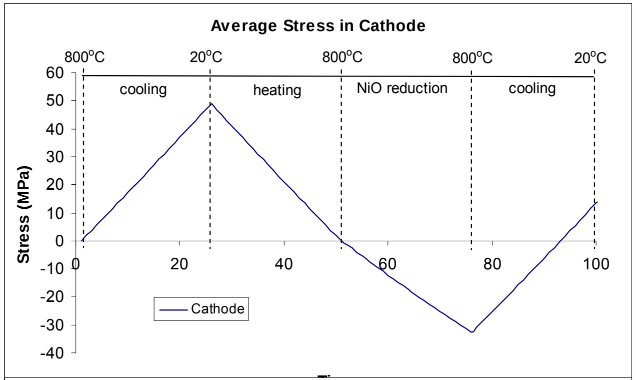
Materials Properties

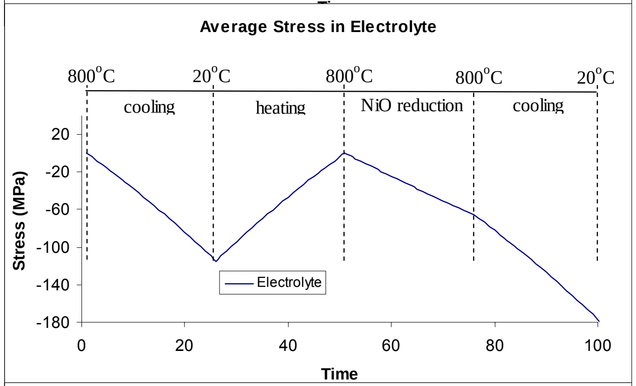
	Young's Modulus (GPa)	Poisson's Ratio	CTE($10^{-6}/^oC$)	Thickness (μm)
Cathode	90	0.3	11.7	75
Electrolyte	200	0.3	10.8	15
Anode	96	0.3	11.2	500

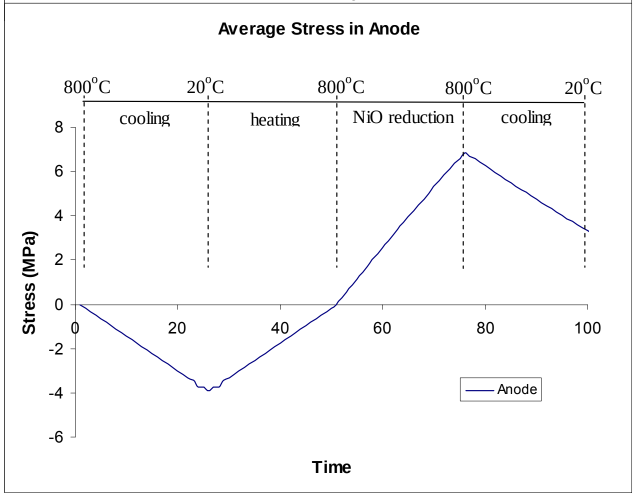
Considering sintering process, the set of materials in table will result in

- tensile stress in cathode;
- compressive stress in electrolyte;
- compressive stress in anode;

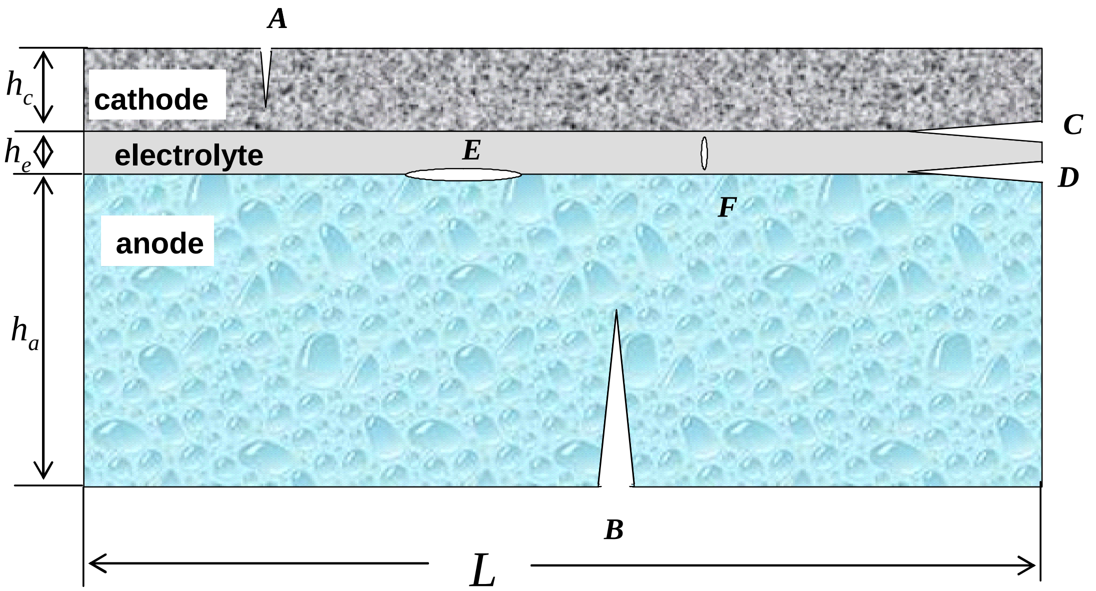
Average in-Plane Stress in the PEN Layers





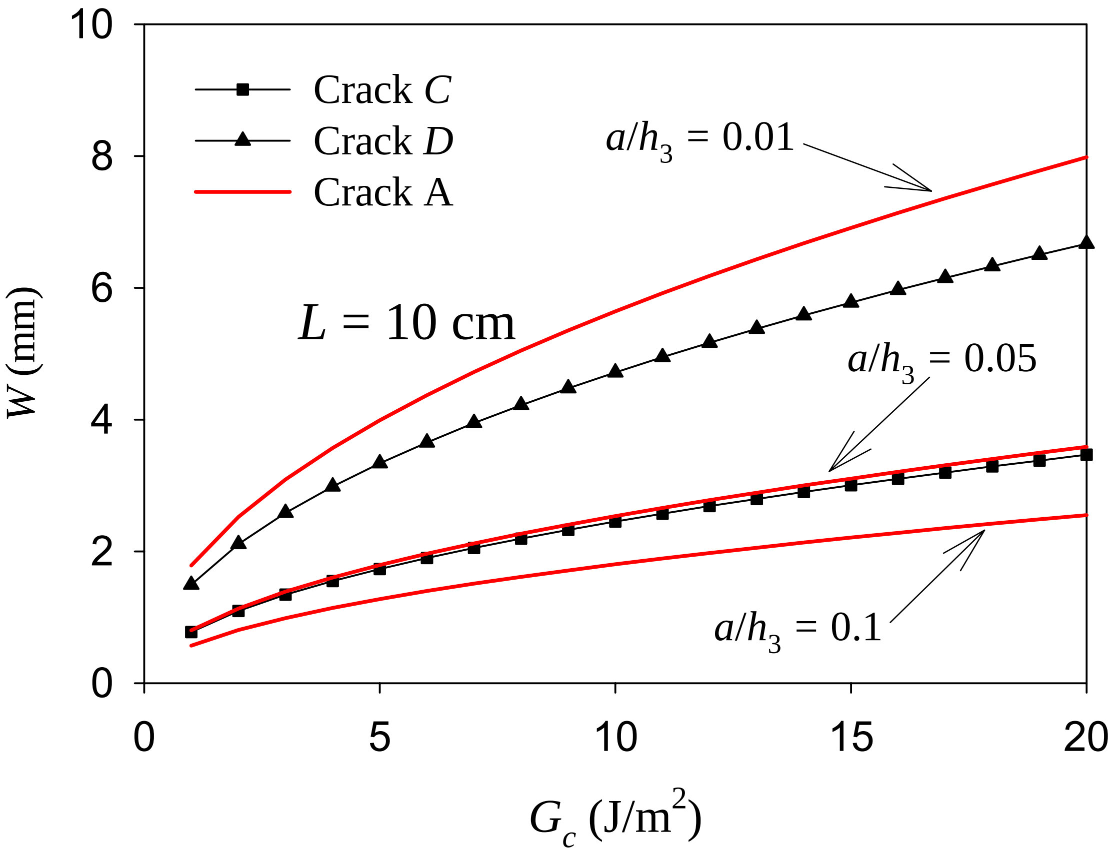


- Stress free at 800°C
- No-creep
- NiO reduction results in 0.1% vol. shrinkage

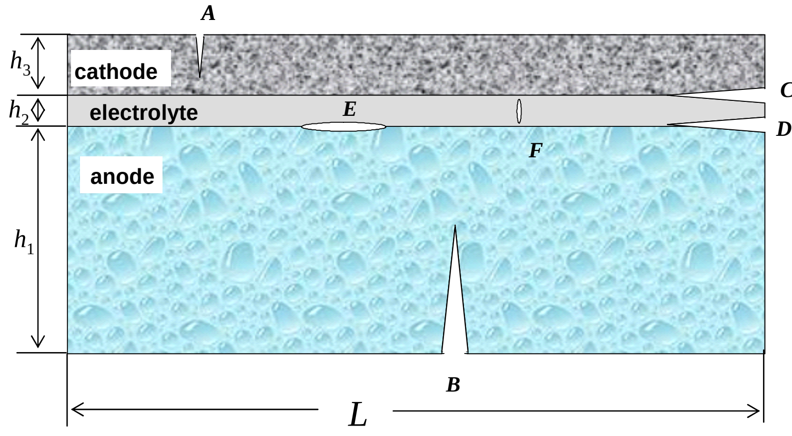


Numerical Examples of Max. Allowable Warpage

	Crack A				Crack C	Crack D
	$a = 0.01\,h_3$	$a = 0.05\,h_3$	$a = 0.1\,h_3$	$a = 0.2\,h_3$		
Y	4.63e-3	2.08e-3	1.48e-3	1.06e-3	3.87e-3	2.01e-3



Crack C is the limiting factor, unless crack A is larger than 5% of the cathode thickness.



Statistical Consideration

Failure theories have the following form:

Failure occurs when $\Sigma > \Sigma_f$

where Σ is the "stress"(e.g., max. normal stress, Mises stress, or SIFs, max. warpage, etc.) and Σ_f is the "strength"(e.g., yield strength, fracture toughness, etc.)

Both Σ and Σ_f can be random variables with certain distributions, such normal distribution, Weibull distribution, etc.

Assume:

$g(\sigma)$ = distribution of stress; $g_f(\sigma)$ = distribution of strength

The probability of failure at a given stress σ is

$$\int_{-\infty}^{\sigma} g_f(x)dx$$

The probability of failure for a given stress distribution $g(\sigma)$ is

$$p_f = \int_{-\infty}^{\infty} g(\sigma)\left[\int_{-\infty}^{\sigma} g_f(x)dx\right]d\sigma$$

Example (Normal Distributions)

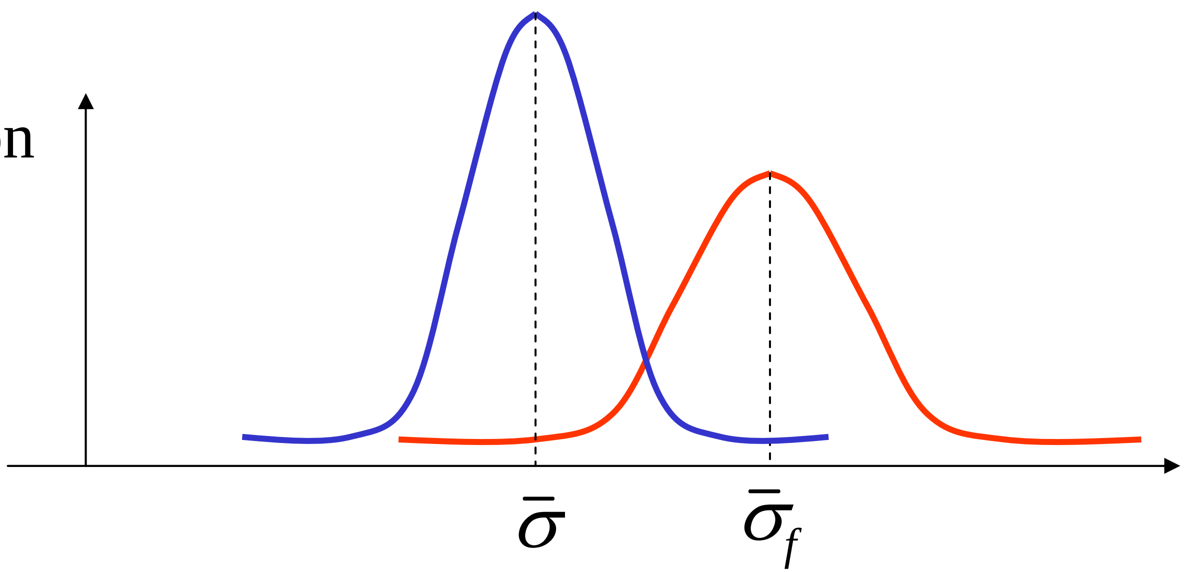
Strength distribution $$g_f(\sigma) = \frac{1}{s_f\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\sigma - \bar{\sigma}_f}{s_f}\right)^2\right]$$

Stress distribution $$g(\sigma) = \frac{1}{s\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\sigma - \bar{\sigma}}{s}\right)^2\right]$$

s = Standard deviation

$\bar{\sigma}$ = Mean value

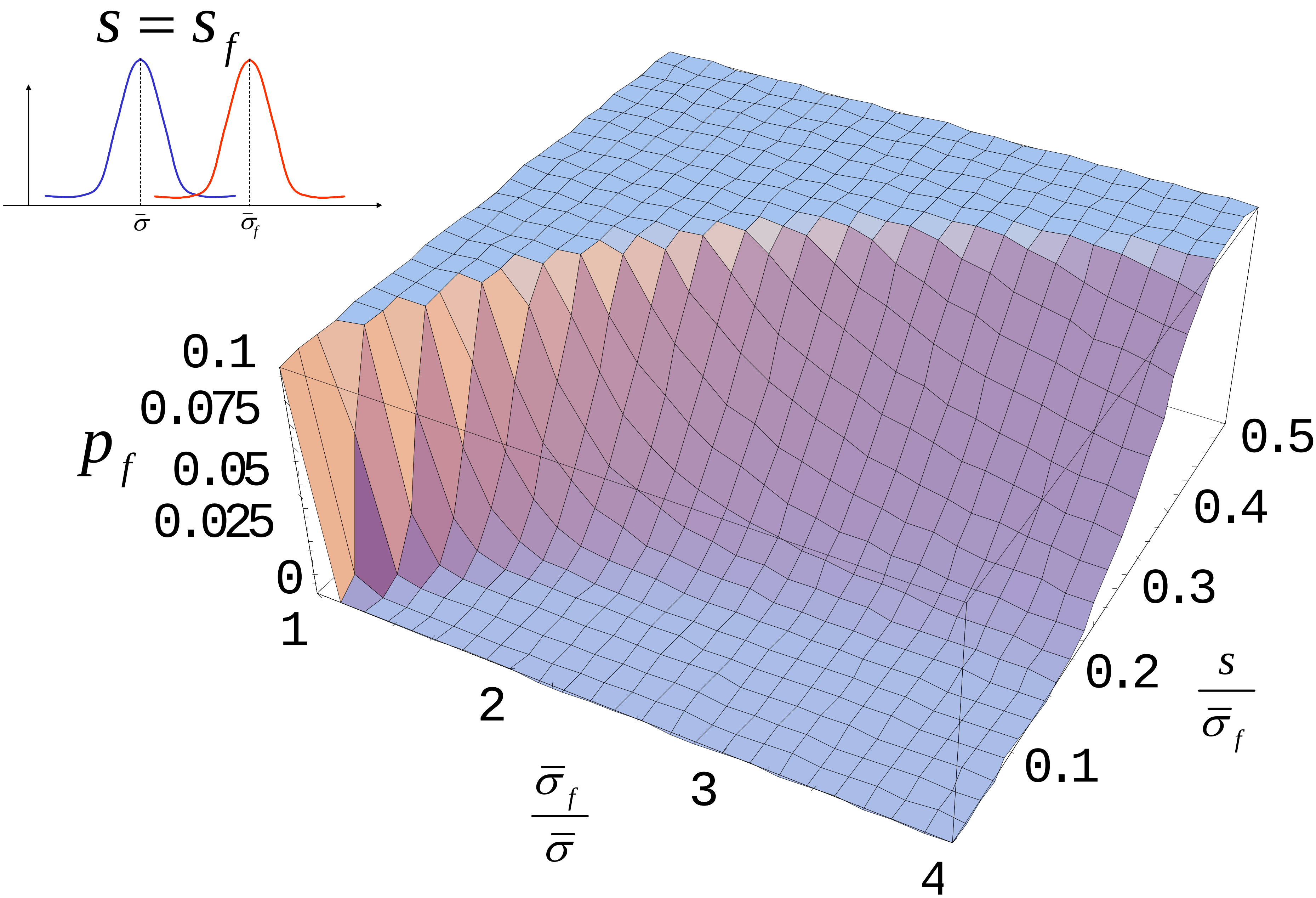
$$\int_{-\infty}^{\infty} g(\sigma) d\sigma = 1$$



$$p_f = \frac{1}{2s\sqrt{2\pi}} \int_{-\infty}^{\infty} \mathrm{Exp}\left[-\left(\frac{\sigma - \bar{\sigma}}{s\sqrt{2}}\right)^2\right] \mathrm{Erfc}\left[\frac{\bar{\sigma}_f - \sigma}{s_f\sqrt{2}}\right] d\sigma$$

Failure Probability

$\bar{\sigma}_f / \bar{\sigma}$ **Factor of Safety**	$s/\bar{\sigma}_f = s_f / \bar{\sigma}_f$ **Deviation**	p_f **Failure Probability**
1.0	any value	0.5
2.0	0.2	3.8×10^{-2}
5.0	0.2	2.3×10^{-4}
10.0	0.2	7.3×10^{-5}
1.5	0.1	9.2×10^{-3}
2.0	0.1	2.0×10^{-4}
3.0	0.1	1.2×10^{-6}
4.0	0.1	5.7×10^{-8}
1.5	0.05	1.2×10^{-6}
2	0.05	7.7×10^{-13}
1.5	0.02	2.3×10^{-32}



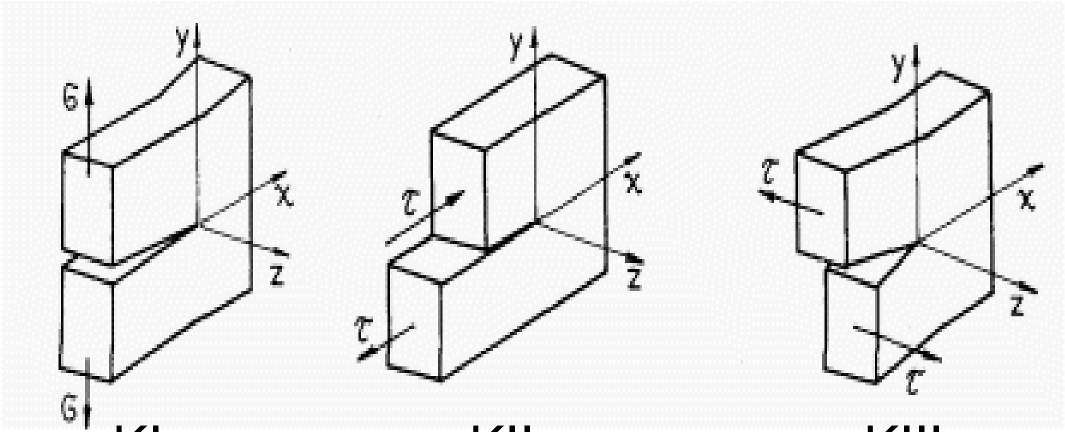
$s = s_f$
$\bar{\sigma}$
$\bar{\sigma}_f$
p_f
0.1
0.075
0.05
0.025
0
1
2
3
4
$\frac{\bar{\sigma}_f}{\bar{\sigma}}$
0.1
0.2
0.3
0.4
0.5
$\frac{s}{\bar{\sigma}_f}$

Summary
- First Order Failure Criteria

- Local and global failure criterion were established. These criterion may be easily used to aid the initial design, material selection and optimization of SOFCs.
- Using the local failure criteria, the user can predict (estimate) the potential material failure
- Using the global failure criteria, the user can predict whether a cell can survive the stacking assembly process

A Numerical Simulation Tool for Fracture Analysis in Solid Oxide Fuel Cells

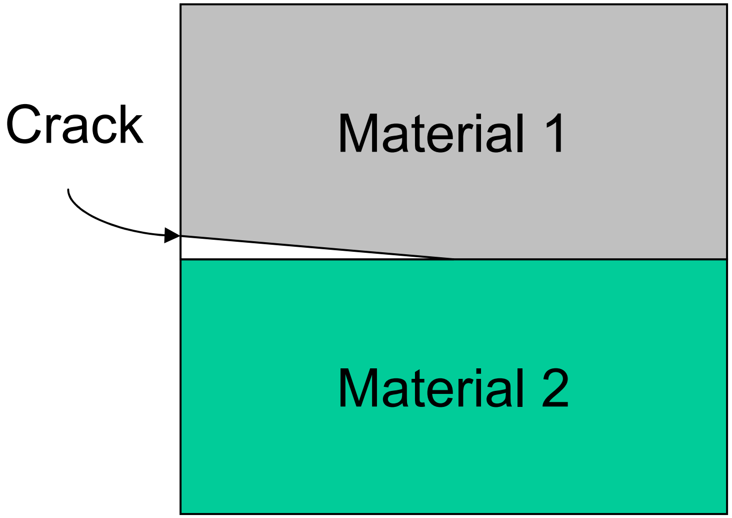
$$\mathbf{K} = K_I + iK_{II} \equiv (\text{applied stress}) \times FL^{1/2 - i\varepsilon}$$



KI
opening

KII
shear

KIII
out of plane



Significance of SIFs

1. Will the crack grow?

$$\frac{1}{E^*}\frac{\mathbf{K}\overline{\mathbf{K}}}{\cosh^2(\pi\varepsilon)}+\frac{K_{III}^2}{2\mu^*}=G_{ic}$$

2. In what direction? (What is mode mixity?)

$$\psi=\tan^{-1}\left[\frac{\mathrm{Im}[\mathbf{K}L^{i\varepsilon}]}{\mathrm{Re}[\mathbf{K}L^{i\varepsilon}]}\right]$$

Computing Fracture Parameters Using Volume Integrals

$$\bar{I} = \bar{G}_{\text{int}} = -\int_V \left(P_{jk}^{\text{int}} \frac{\partial q_j}{\partial x_k} + \frac{\partial P_{kj}^{\text{int}}}{\partial x_k} q_j \right) dV$$

Virtual crack growth (Q)

$$P_{jk}^{\text{int}} = \sigma_{mn}\varepsilon_{mn}^{aux}\delta_{jk} - \sigma_{ik}\frac{\partial u_i^{aux}}{\partial x_j} - \sigma_{ik}^{aux}\frac{\partial u_i}{\partial x_j}$$

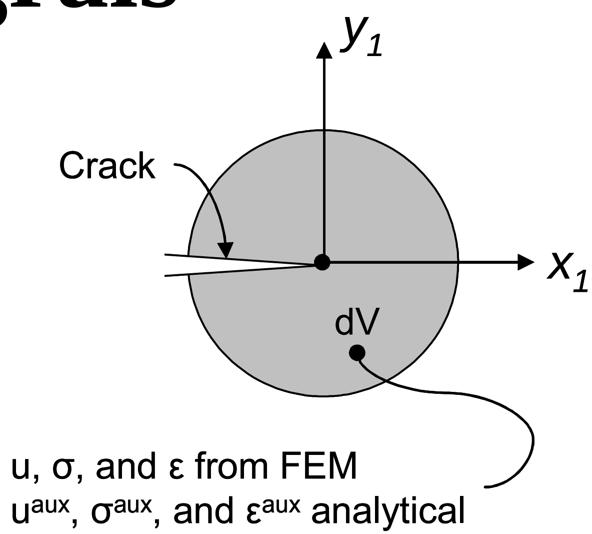
Strain Energy

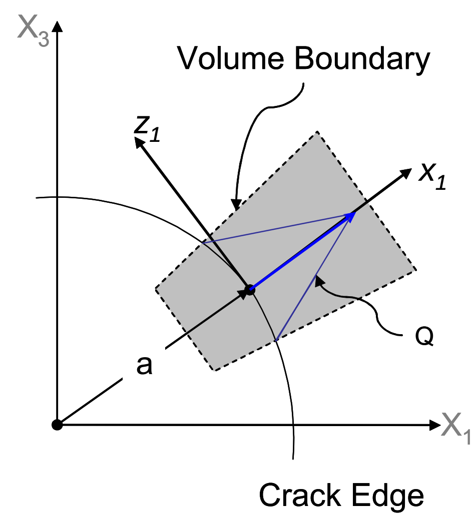
Stress and spatial derivatives of displacements

$$\frac{P_{kj}^{\text{int}}}{\partial x_j} = \sigma_{ij}\varepsilon_{ij,k}^{aux} - \sigma_{ij}u_{j,ik}^{aux} - \sigma_{ij,i}^{aux}u_{j,k} - \alpha\sigma_{ii}^{aux}\theta_{,k}$$

Curvilinear

Temperature





- Pointwise Value

$$I(s) = \frac{\bar{I}}{\int_{L_c} \Delta a(s) ds}$$

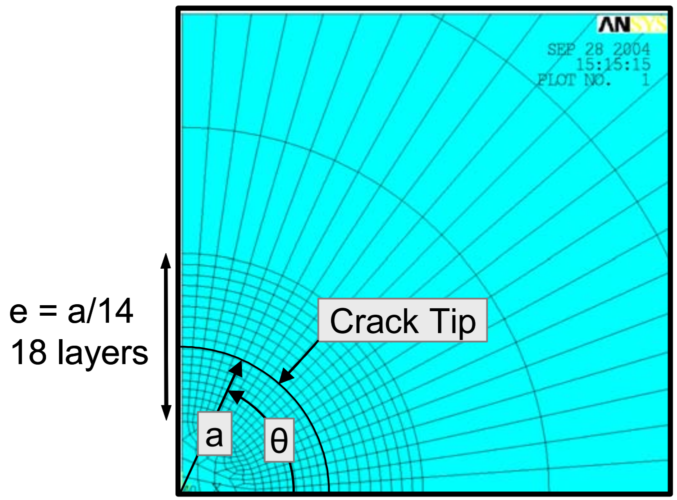
$$I(s) = \frac{2}{E^* \cosh^2(\pi\varepsilon)} \left[K_I K_I^{aux} + K_{II} K_{II}^{aux} \right] + \frac{1}{\mu^*} K_{III} K_{III}^{aux}$$

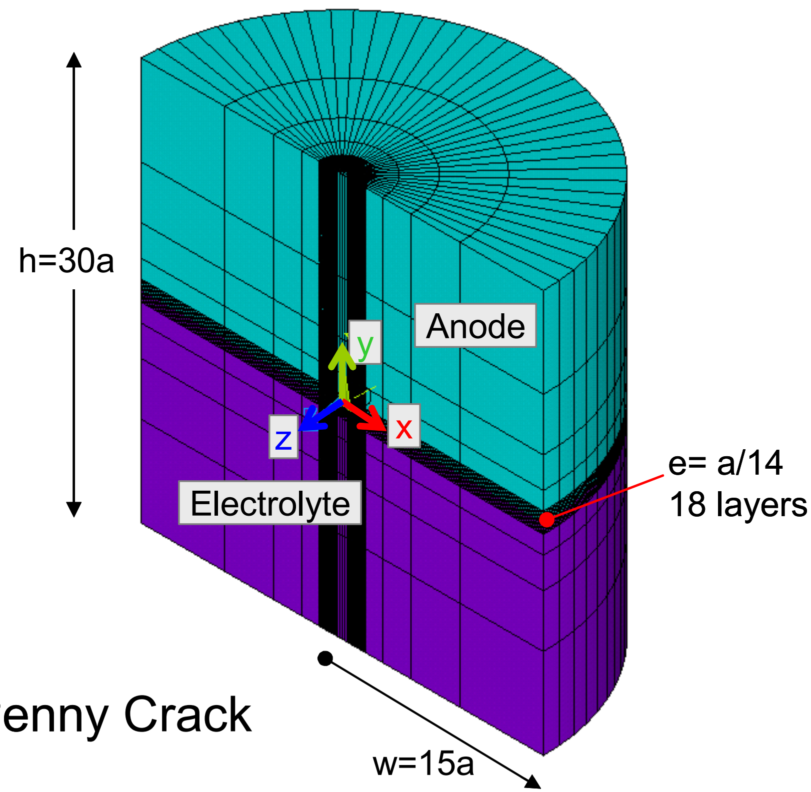
- To find K_I by setting
  - $K_I^{aux}=1$
  - $K_{II}^{aux}=K_{III}^{aux}=0$

$$\Rightarrow \quad K_I = \frac{I(s)}{2} E^* \cosh^2(\pi\varepsilon)$$

A Penny-Shaped Crack on Electrolyte/Anode Interface

Mesh footprint near crack



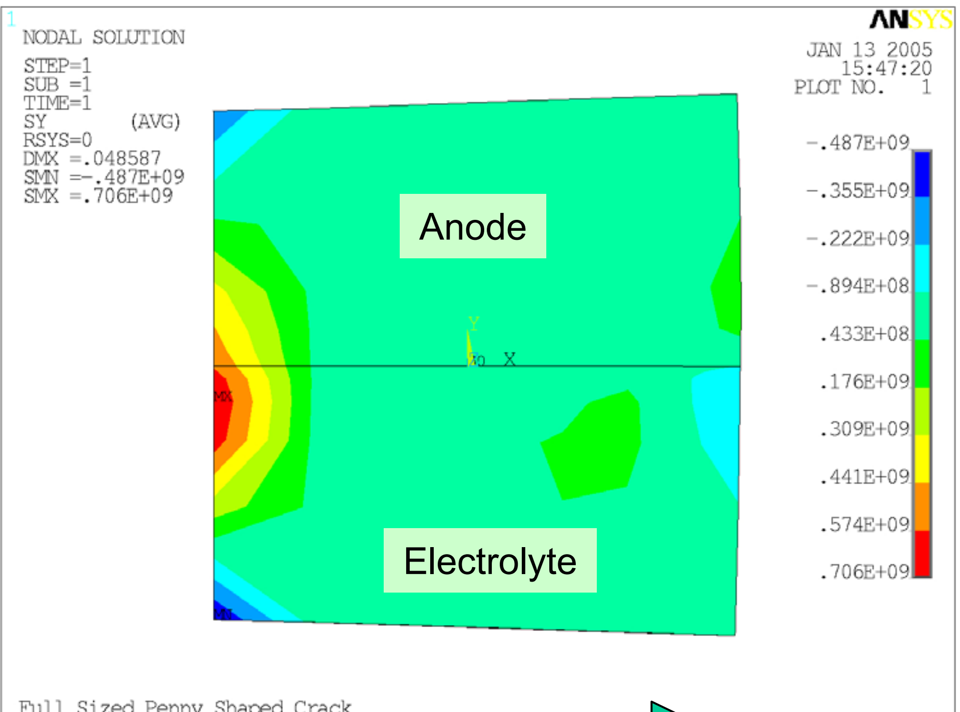


Half Model - Interface Penny Crack

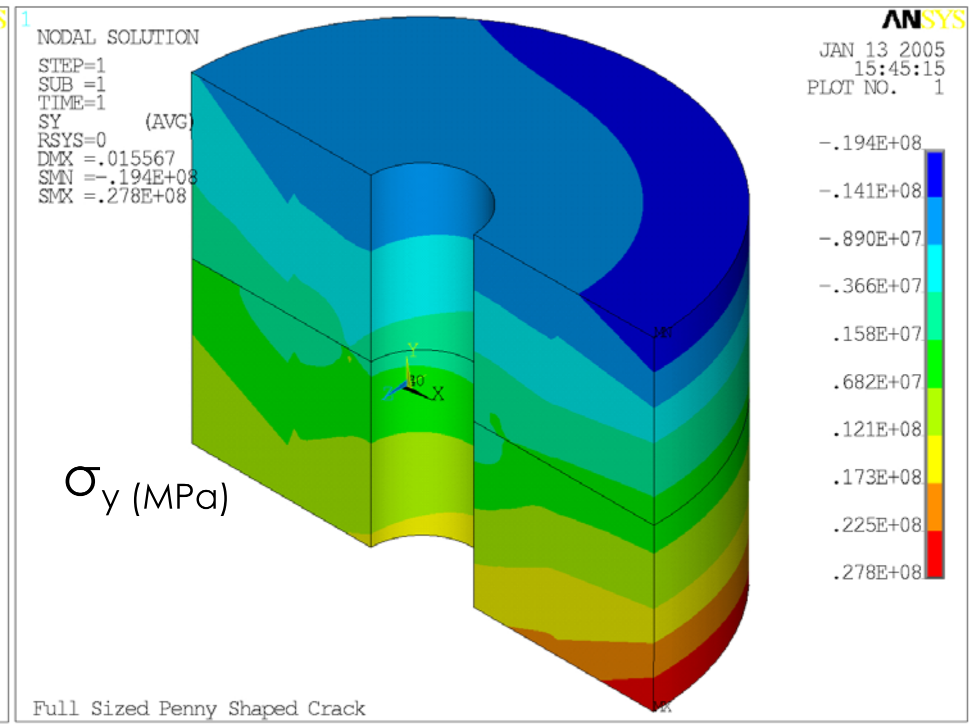
Temperature gradient parallel to crack plane

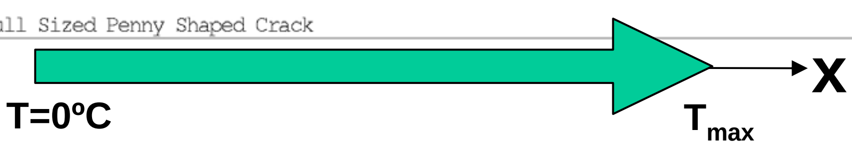
Temperature Gradient Parallel to the Crack Plane

ANSYS Model

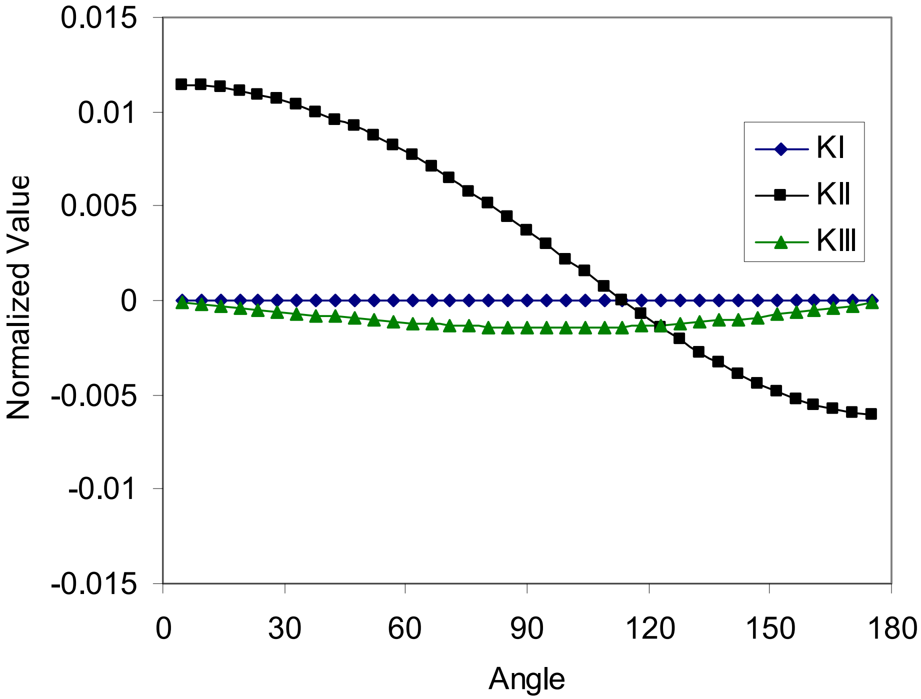


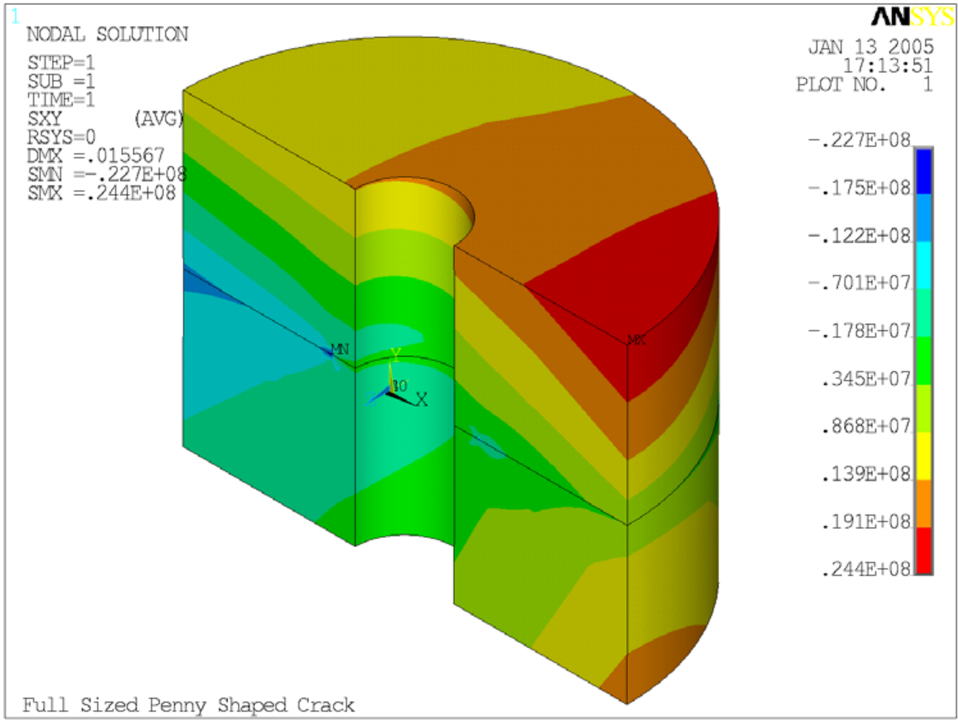
FMA Volume





K_I, K_{II}, K_{III} Variation Along Crack Front





Summary

-- Fracture Mechanics Analysis Tool

Fracture Mechanics Analysis Tool:

- Based on volume integral (requires less mesh density)
- Written in MatLab language (run on both Window and Unix)
- Add-on to any commercial FEM codes (requires less processing time)

Capabilities:

- Calculate energy release rate and individual stress intensity factors
- 2D and 3D planar cracks of arbitrary shapes
- Homogeneous and interfacial cracks
- Arbitrary mechanical and thermal loading

Transient Heat Transfer Analysis: Convective-Conductive Heating of SOFC

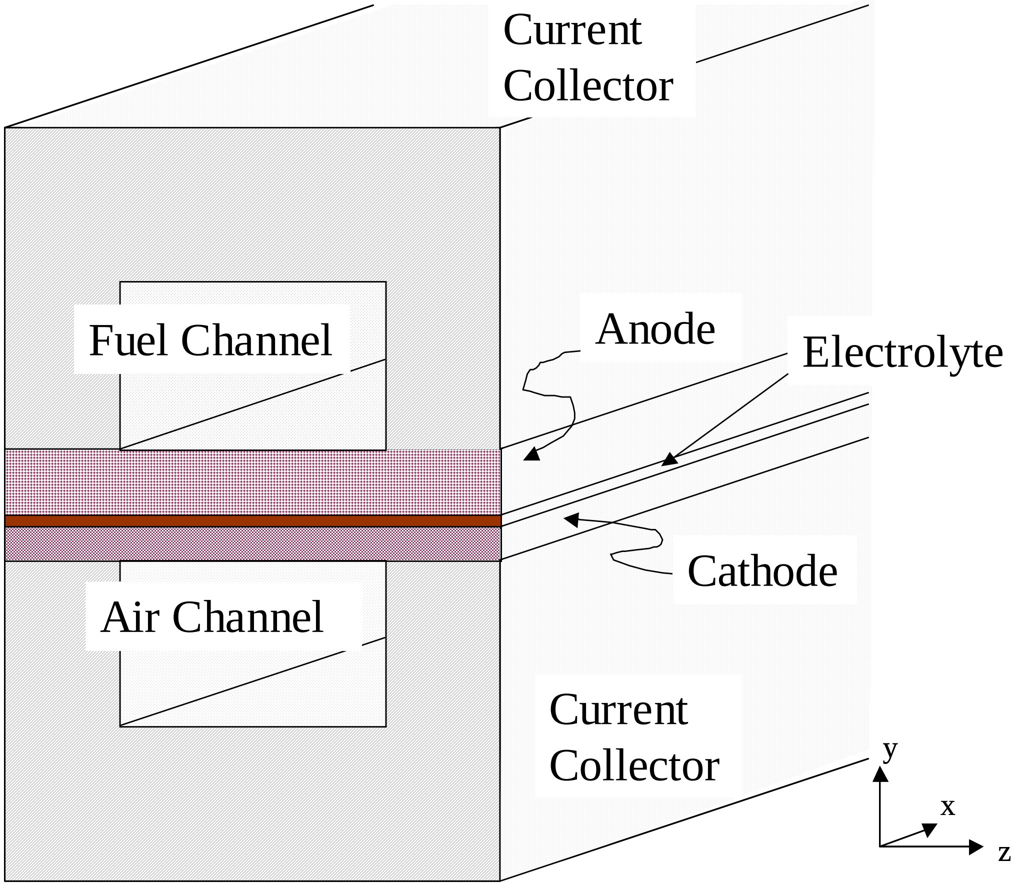
SOFC unit cell Transient Thermal Modeling

Key Question/Focus: Provide model-based design tool(s) to assess how quickly a cell/stack cane be heated without excessive (damaging) thermomechanical gradients?

Design/Model Outputs:
- total time required for heating
- max temperature spatial gradient
- max temperature time-derivative

Model Inputs:
- size of components; thickness of layers
- thermal properties of components
- boundary conditions; heating strategies

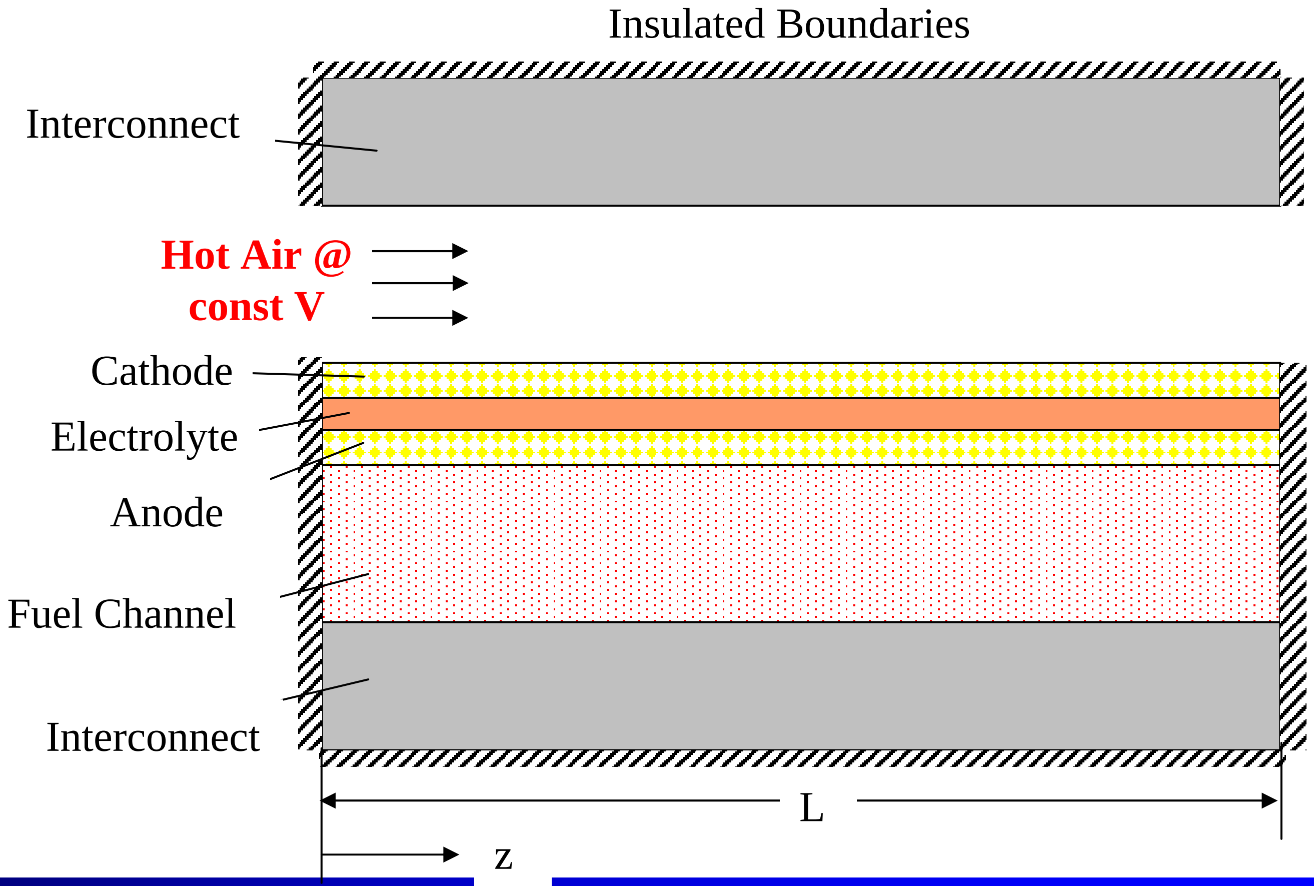
Multi-Level Methodology:
- 3-D CFD modeling (e.g. FLUENT)
- Reduced order numerical modeling
- Simplified order analytical modeling



Simplified Analytical Model/Design Tool: Key Ideas & Assumptions

Heating by hot air supplied at prescribed time-dependent inlet temperature

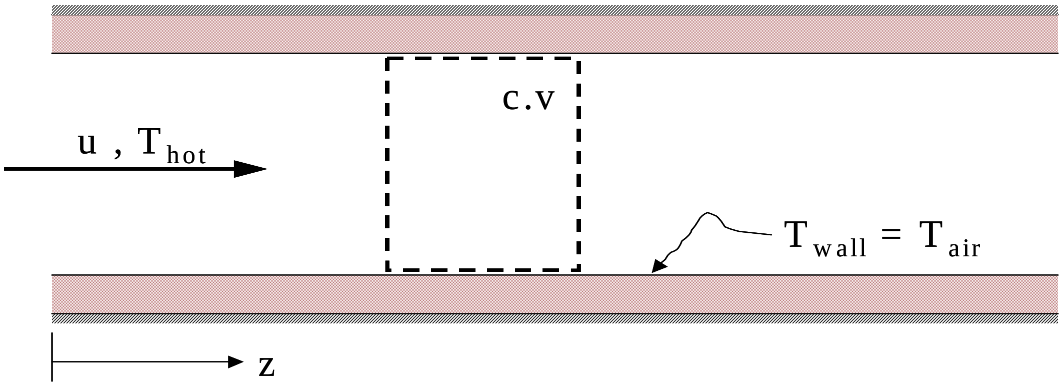
- 1-D temperature profile in each component → $T_{layer} = f\,(z,t)$ *only*
- Constant velocity plug flow in channel
- Constant properties
- Radiation neglected *(to be included later)*
- Adiabatic boundaries *(no heat losses)*



1^{st} order 1-D Purely Convective Heating Model

Key Assumptions:

- thermally thin cell materials (i.e. no energy storage)
- thermal equilibrium between air and channel walls



Governing Equation → $$\frac{\delta T}{\delta t} + u\frac{\delta T}{\delta z} = 0$$

B.C. & I.C.

$$T\left(z=0,t\right) = f\left(t\right)$$

$$T(z,t=0) = T_o$$

Closed-form analytical solution:

$$T\left(z,t\right) = \begin{cases} T_o & \text{for} \quad z > ut \\ f\left(t - \dfrac{z}{u}\right) & \text{for} \quad z \le ut \end{cases}$$

2^{nd} Order Convective-Conductive Heating Model

InterConnect$_1$: $$(\rho c_p A)_{IC_1}\frac{\partial T_{IC_1}}{\partial t}=(kA)_{IC_1}\frac{\partial^2 T_{IC_1}}{\partial z^2}+hP_{g-IC_1}(T_g-T_{IC_1})-\frac{P_{IC_1-C}}{R_{IC_1-C}}(T_{IC_1}-T_C)$$

Air Channel (gas): $$(\rho c_p A)_g\left[\frac{\partial T_g}{\partial t}+u\frac{\partial T_g}{\partial z}\right]=(kA)_g\frac{\partial^2 T_g}{\partial z^2}-hP_{g-C}(T_g-T_C)-hP_{g-IC_1}(T_g-T_{IC_1})$$

Cathode: $$(\rho c_p A)_C\frac{\partial T_C}{\partial t}=(kA)_C\frac{\partial^2 T_C}{\partial z^2}+hP_{g-C}(T_g-T_C)+\frac{P_{IC_1-C}}{R_{IC_1-C}}(T_{IC_1}-T_C)-\frac{P_{C-E}}{R_{C-E}}(T_C-T_E)$$

Electrolyte: $$(\rho c_p A)_E\frac{\partial T_E}{\partial t}=(kA)_E\frac{\partial^2 T_E}{\partial z^2}+\frac{P_{C-E}}{R_{C-E}}(T_C-T_E)-\frac{P_{E-A}}{R_{E-A}}(T_E-T_A)$$

Anode: $$(\rho c_p A)_A\frac{\partial T_A}{\partial t}=(kA)_A\frac{\partial^2 T_A}{\partial z^2}+\frac{P_{E-A}}{R_{E-A}}(T_E-T_A)-hP_{f-A}(T_f-T_A)-\frac{P_{A-IC_2}}{R_{A-IC_2}}(T_A-T_{IC_2})$$

FuelChannel: $$(\rho c_p A)_{fuel}\frac{\partial T_{fuel}}{\partial t}=(kA)_{fuel}\frac{\partial^2 T_{fuel}}{\partial z^2}+hP_{f-A}(T_f-T_A)+hP_{f-IC_2}(T_f-T_{IC_2})$$

Applying thermal equilibrium between flow channels and components; model reduces to a single equation dependent only on effective Peclet number and inlet temperature function!

$$\boxed{\frac{\partial T}{\partial t}+\frac{\partial T}{\partial z}=\frac{1}{Pe}\frac{\partial^2 T}{\partial z^2}}$$

$$Pe=\frac{u_{eff}L}{\alpha_{eff}}\equiv\frac{\text{advection}}{\text{conduction}}\text{ of thermal energy}$$

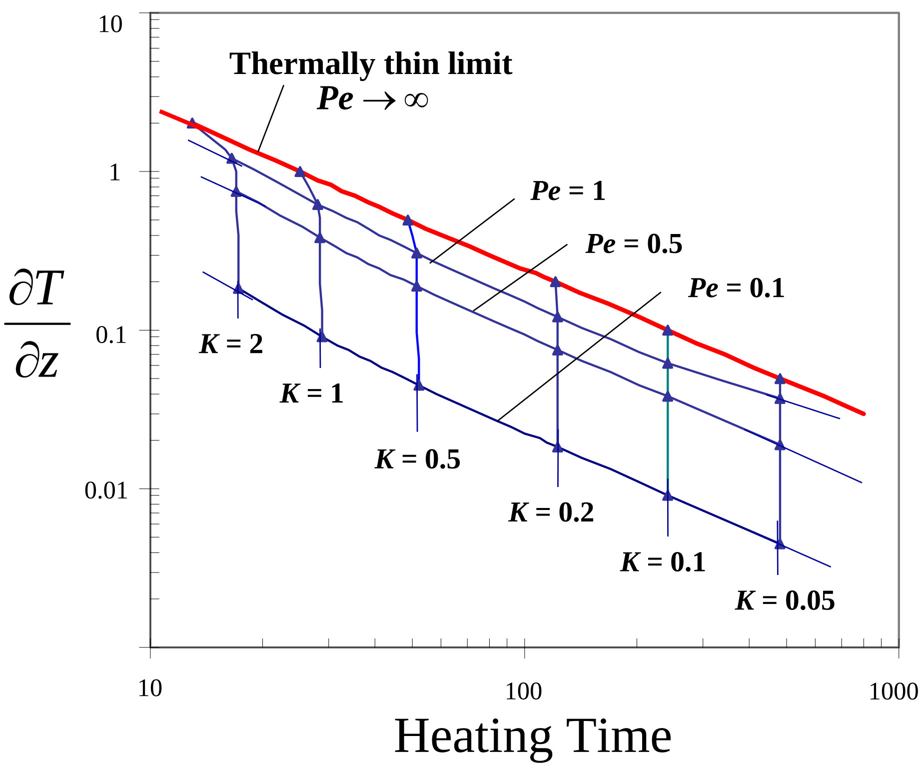
B.C.& I.C.: $T(0,t)-Pe\frac{\partial T}{\partial z}(0,t)=F(t);\quad \frac{\partial T}{\partial z}(1,t)=0;\quad T(z,0)=1$

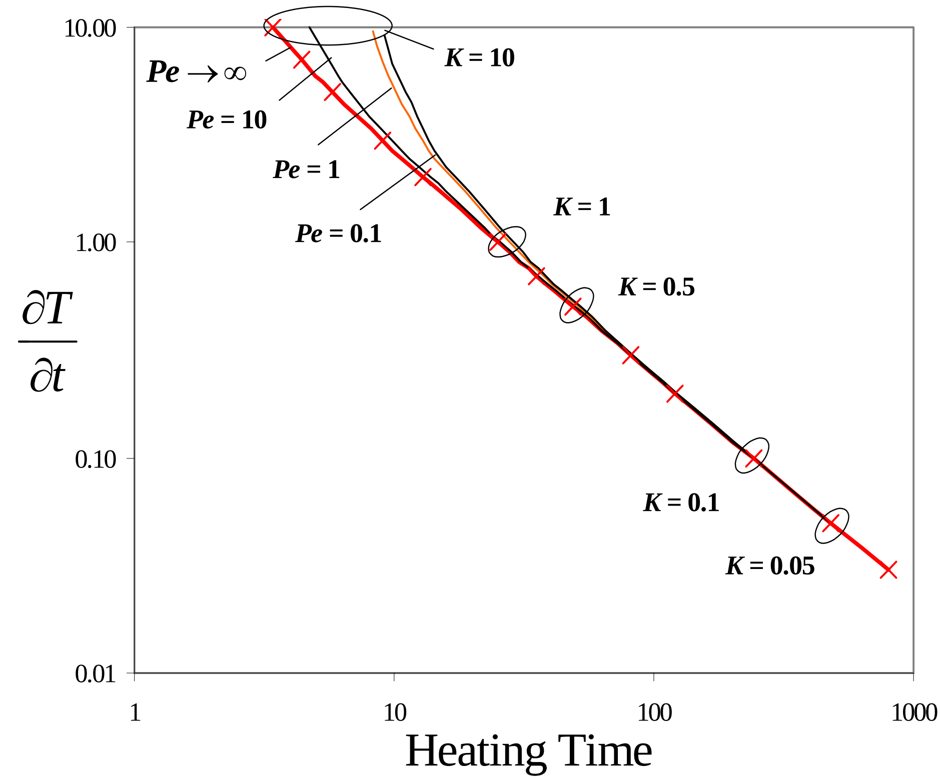
Closed-form analytical solution has been obtained!!!

Results: Comparison of 1st and 2nd order models

Key advantages demonstrated:

- Computationally efficient, analytical models capture key physics of heating process!
- 1st order model is the limiting case of 2nd order model (Pe → large) provided it is properly re-scaled. The guidelines for re-scaling have been developed!





Design Maps: Dimensionless plots of temperature gradient and time-derivative vs. total heating time for various rates of inlet temperature rise (K) and Peclet numbers (Pe).

Summary
-- Convective-Conductive Heating of SOFC

- Developed reduced order solutions for transient thermal analysis.
- Obtained closed-form analytical solutions that provide a relationship between heating rate and the spatial temperature gradient
- Obtained closed-form analytical solutions that provide a relationship between rate and the temporal temperature gradient

Future Work

- Refine and validate the first order failure criteria
- Develop and implement the global-local computational algorithm in MARC
- Validate and implement a suitable FEA tool for analysis of fracture failure in the context of various pre-existing flaws within SOFC cells under various operating conditions.
- Validate and implement a computationally-efficient transient thermal model.