SECA Core Technology Program (CTP) Overview

Prabhakar Singh and Jeff Stevenson

Pacific Northwest National Laboratory

Presented at

Fourth Annual SECA Meeting, Seattle

April 15-16, 2003

Outline

- Program Goals and Objectives
- CTP Organization and Structure
- Major Accomplishments
- Technology Development Programs
- Status and Results
- Conclusions

SECA program

- DOE Office of Fossil Energy Initiative.
- Program coordination through National Energy Technology Laboratory (NETL) and the Pacific Northwest National Laboratory (PNNL)

The SECA Alliance is coordinated so that cost-effective solid oxide fuel cell prototypes for diverse applications are produced commercially and environmental concerns associated with current methods of generating electricity from fossil fuels are mitigated.

SECA - Structure

Two Major Program Elements:

- Industrial Development Teams pursuing commercial SOFC development (60%).
  - General Electric Power Systems
  - Siemens Westinghouse Power Corp.
  - Delphi Automotive Systems / Battelle Memorial Institute
  - Cummins Power Generation / SOFCo

- Core Technology Program provides problem-solving research aimed at overcoming problems identified by industry teams (40%).
  - Universities, national laboratories, and other research-oriented organizations

SECA CTP Goals and Objectives

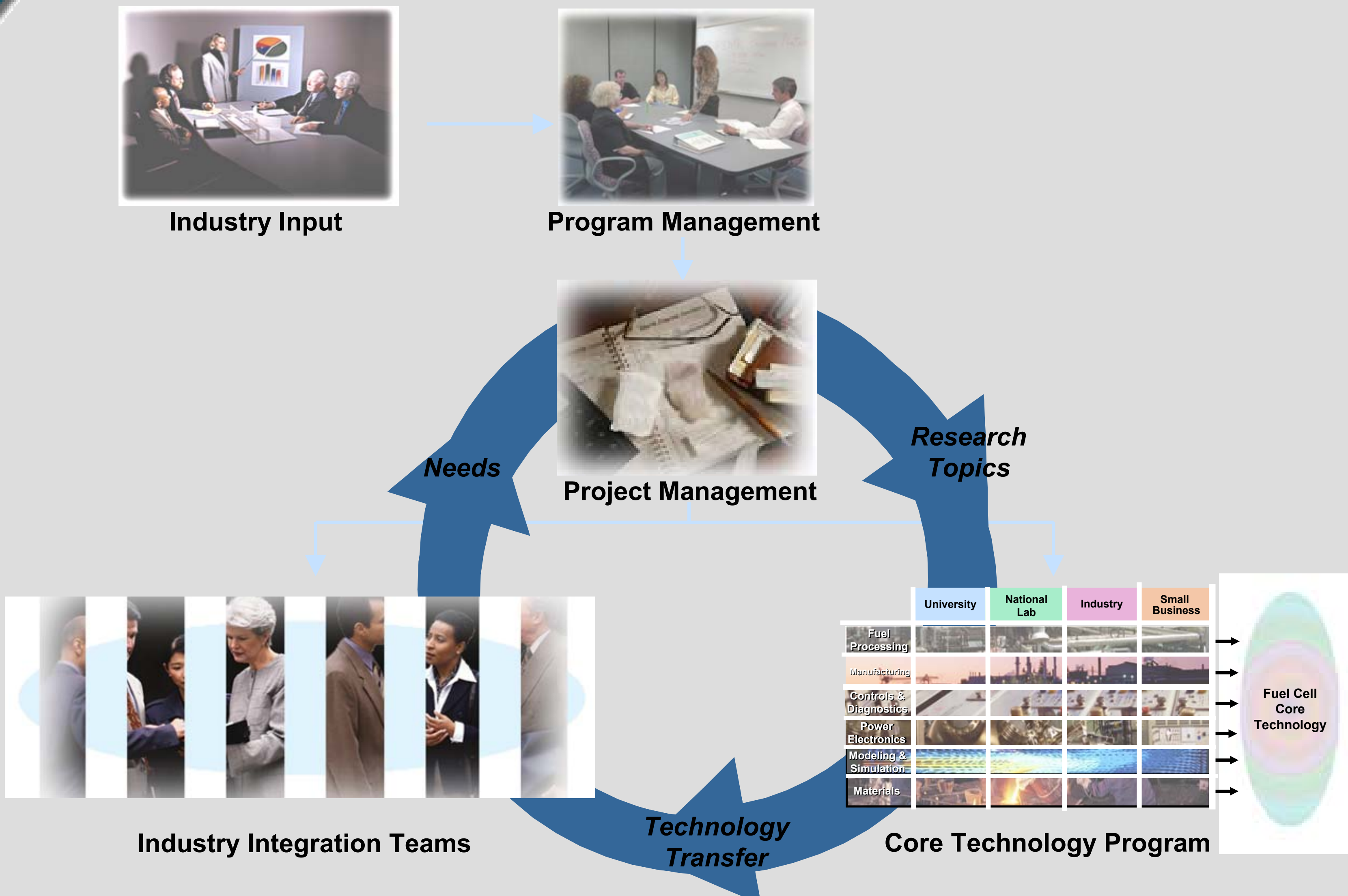
SECA Core Technology Program, in consultation and agreement with SECA Industry teams, will develop cost effective technologies, manufacturing processes and advanced knowledge base to support the cost and performance targets and development schedule of the modular SOFC power generation systems.

CTP Objectives

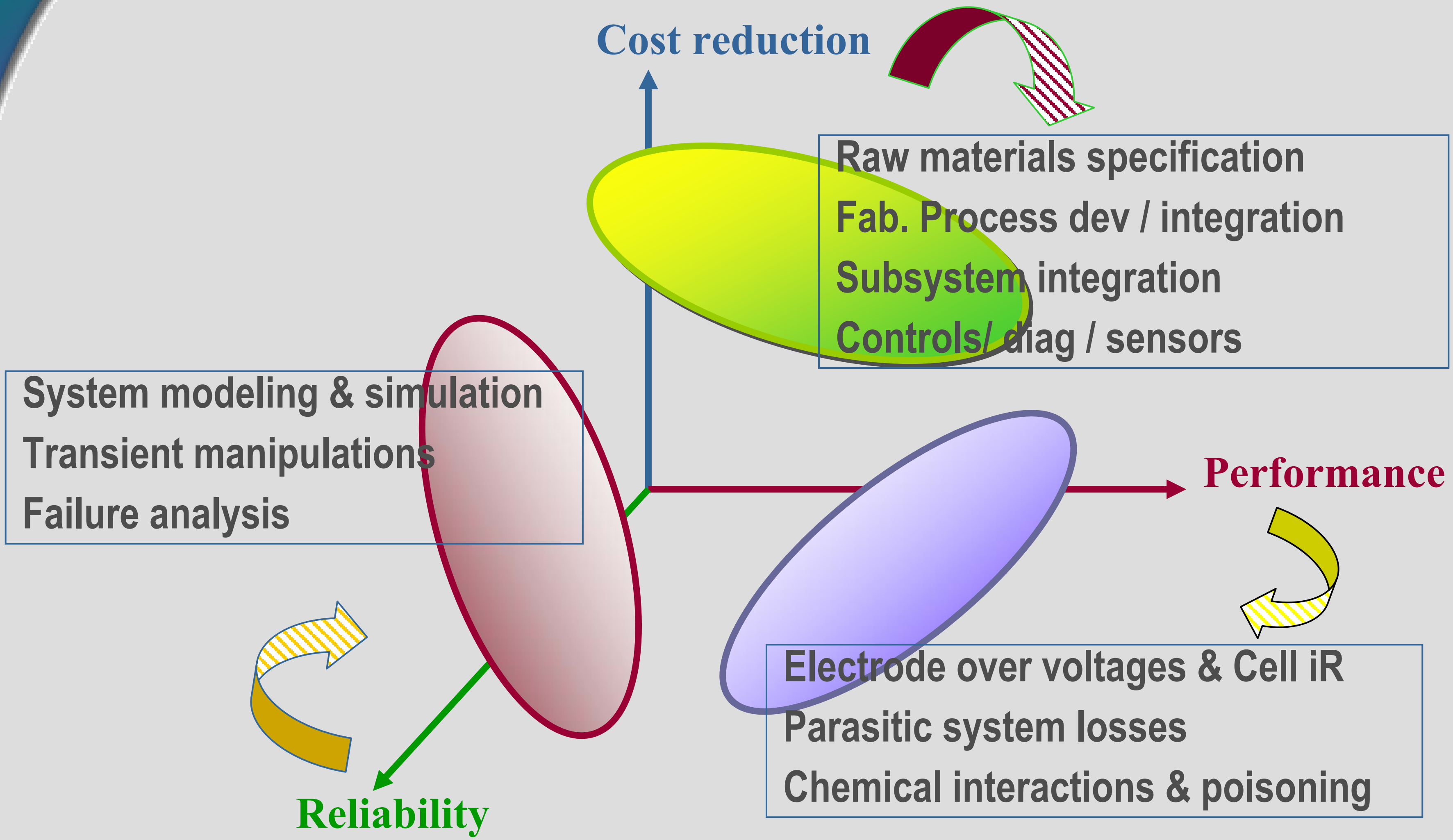
- Identify technology gaps and development needs
- Prioritize development needs
- Develop and execute technology programs
- Disseminate results through meetings/ publications

- Meet SECA Modular SOFC Cost Targets
- Meet SECA Performance Targets
- Meet SECA Development Schedule

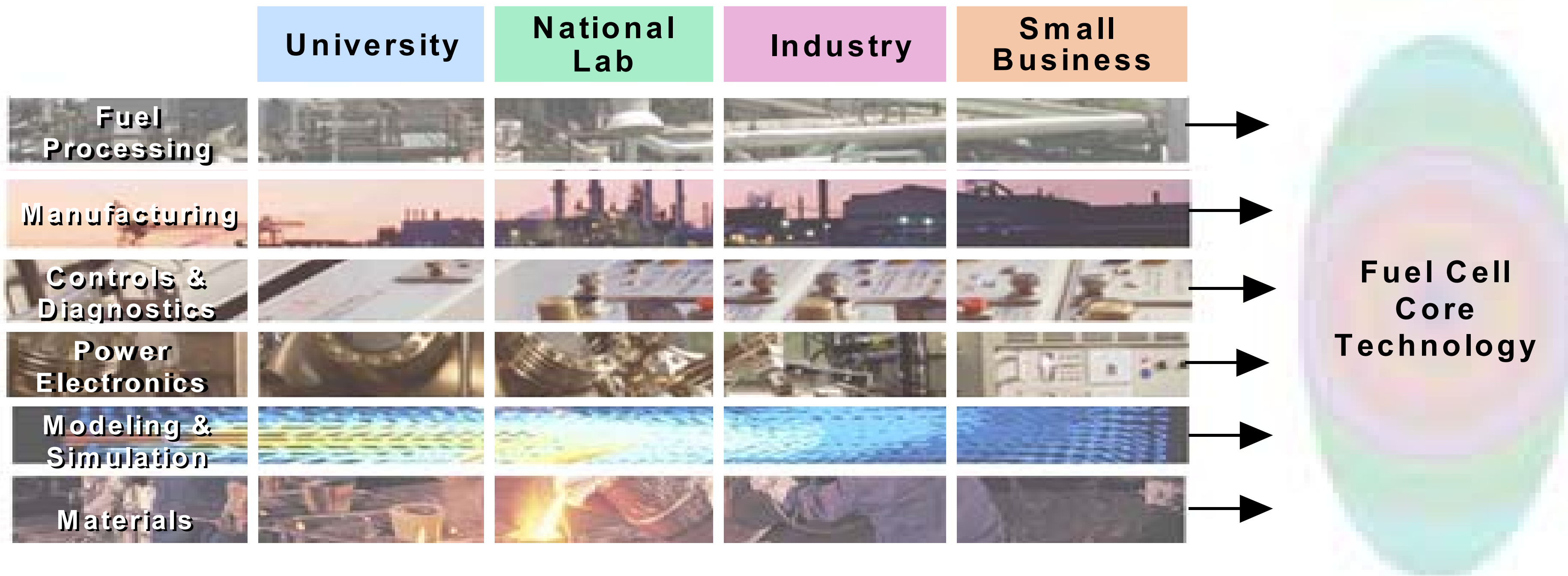
SECA Program Structure



CTP Focus Areas



Core Technology Program
The Technology Base



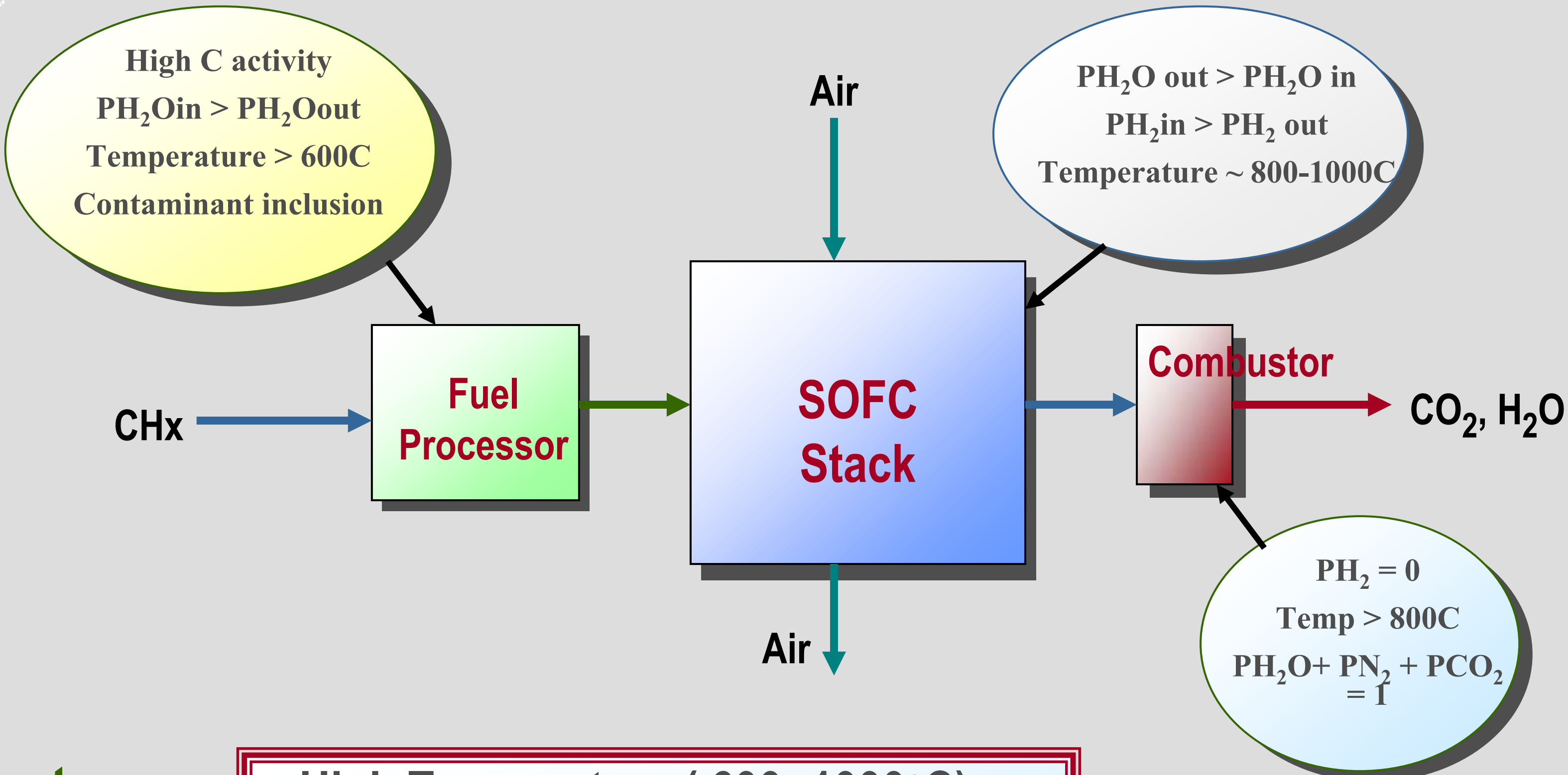
Core Technology Thrust Area Leaders

SECA Coordinator: Mr. Wayne Surdoval

SECA CTP Coordinator : Dr. Prabhakar Singh

Thrust Area	***NETL***	***PNNL/ORNL***
• Materials	Dr. Lane W. Wilson	Dr. Jeff Stevenson
• Manufacturing	Mr. Wayne Surdoval	Dr. Prabhakar Singh
• Fuel Processing	Dr. Dave Berry	Dr. Dave King
• Modeling & Simulation	Dr. William Rogers	Dr. Moe Khaleel
• Power Electronics	Dr. Don Collins	Dr. Don Adams

Fuel Cell Power Generation Systems



Features:

- High Temperature (600- 1000°C)
- Complex fuel gas environment
- Changing PH_2O
- H_2O Recirculation

Workshop Findings

(Workshop held in Atlanta, 2001)

Topical Area	Top 3 Development Needs
Cell/Stack Materials & Manufacturing	1. Stable Interconnect 2. Fuel/ Oxidant Seals 3. Internal Reforming/ Direct oxidation
Fuel processing	1. Sulfur Tolerant Anode 2. Catalyst Kinetics, Parameters & Deactivation 3. On anode Fuel Utilization
Stack/ Systems Performance & Modeling	1. Fast start up and Thermal Cycles 2. Cell & Stack Performance Model 3. Low Cost HX, Insulation, Blowers & sensors
Power electronics	1. Fuel cell / PE Interface 2. Materials & Fabrication Processes 3. Modeling: Electrical Interfaces

SECA Core Technology Broad Based Projects

Cathodes
Georgia Tech Research Corporation
University of Washington
Functional Coating Technologies, LLC
University of Utah
University of Missouri-Rolla
Interconnects
University of Pittsburgh
Ceramatec
Southwest Research Institute
Fuel Cell Failure Analysis
University of Florida
Georgia Tech Research Corporation

Contaminant Resistant Anodes and Reforming Catalysts for Intermediate Temperature Solid Oxide Fuel Cell Power Systems
Northwestern University
Gas Technology Institute
Interaction Between Fuel Cell, Power Conditioning Systems & Application Loads
University of Illinois
Low Cost Production of Precursor Materials
NexTech Materials, Ltd.
University of Utah
Manufacturing Models
TIAX (previously Arthur D. Little)
DC-to-DC Converters for Solid-Oxide Fuel Cells
Texas A&M University
Virginia Polytechnic Institute and State University
Sensors
NexTech Materials, Ltd.

National Laboratory Participation

- Pacific Northwest National Laboratory
  - National Program Management
  - Cell and stack component materials
  - Computational modeling, tools and training
- Argonne National Laboratory
  - Cell cathode materials
  - Current collector materials and concepts
- Oak Ridge National Laboratory
  - Materials properties and engineering model verification
  - Power electronics development coordination
- Lawrence Berkeley National Laboratory
  - Advanced cell components development and testing
  - Advanced cell designs and concepts
- Los Alamos National Laboratory
  - Fuel processing
- National Energy Technology Laboratory
  - Fuel processing
  - Computational modeling
  - Advanced current collector materials and coatings

SECA CTP Accomplishments

Technical

- Low cost anode-supported cell fabrication process (based on tape casting/ screen printing) developed; Technology transferred to SECA Industry team
- > 1W/cm^2 power density operation demonstrated on experimental anode supported cells in H_2-H_2O fuel.
- Low cost, high performance intermediate-temperature cathode compositions developed (doped lanthanum ferrite)
- S, C, O tolerant anode electrodes formulated and tested
- Cell and stack multi physics models developed; Studies included fuel and oxidant utilizations, flow and temperature distribution, current generation and stress profiles in single cell and stacks

SECA CTP Accomplishments

Technical

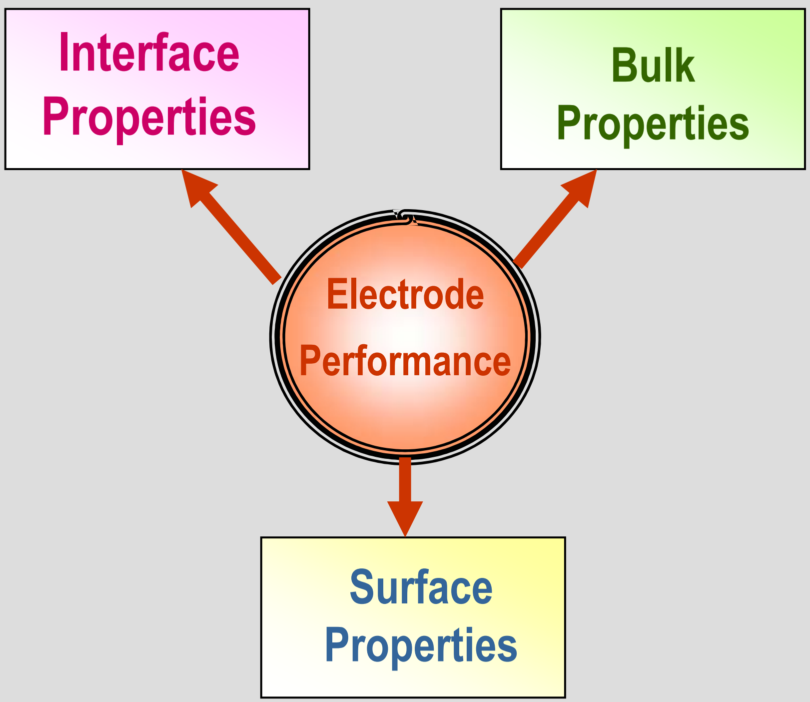
- Modeling tools made available for SECA industry teams
- Training program for SECA teams formulated
- Advanced metallic current collector materials tested; corrosion processes and mechanisms established
- Catalytic processes and reaction kinetics studies initiated
- Liquid fuel processing and carbon deposition studies initiated
- High efficiency power electronics sub systems development initiated

SECA CTP Accomplishments

Programmatic

- 2 workshops/ review meetings held in Yr 2002
- Status of R&D activities peer reviewed
- Topical reports issued to industry teams
- Technical papers published in refereed journals
- Inter-agency collaborations initiated (NASA, DOD)
- Accelerated communications through electronic web posting

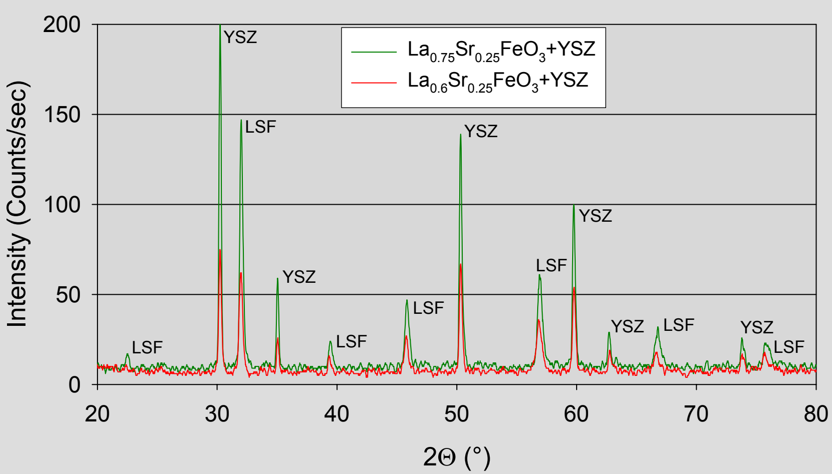
Electrode Performance Matrix

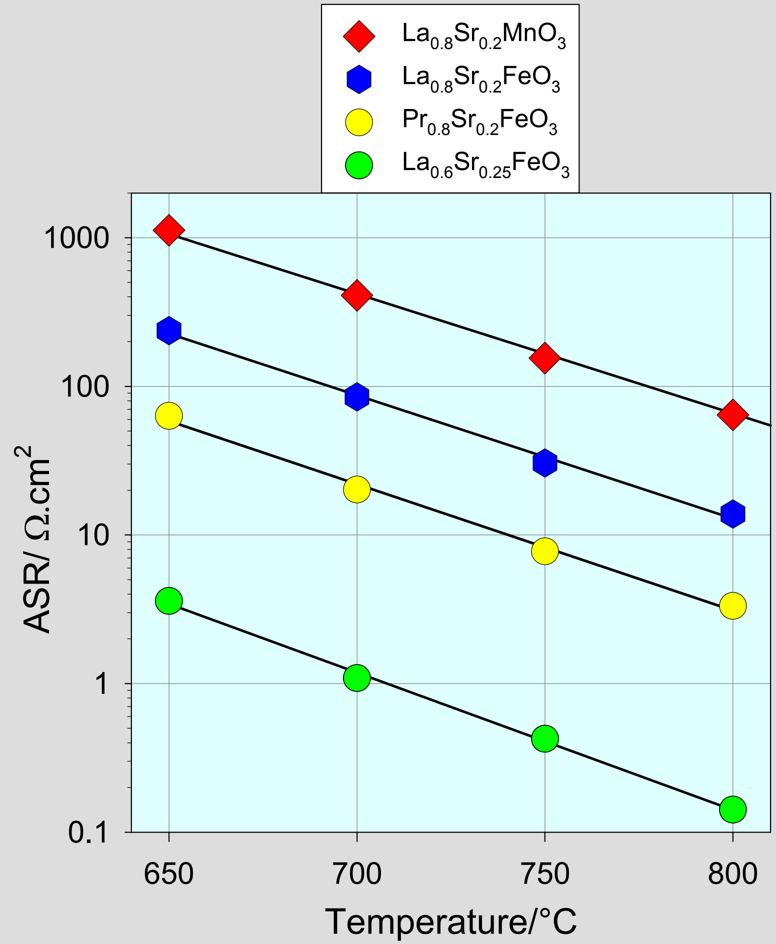


- **Bulk Properties** → Strength, Thermal & Elect. Conductivity, Diffusivity, creep etc.
- **Interface Properties** → Activation polarization, electron transport, Defect stoichiometry, chemical stability etc.
- **Surface Properties** → Adsorption, Electrochemical poisoning, surface condensation etc.

Electrode Development

Developed mixed conducting cathode materials improve intermediate temperature cell performance.





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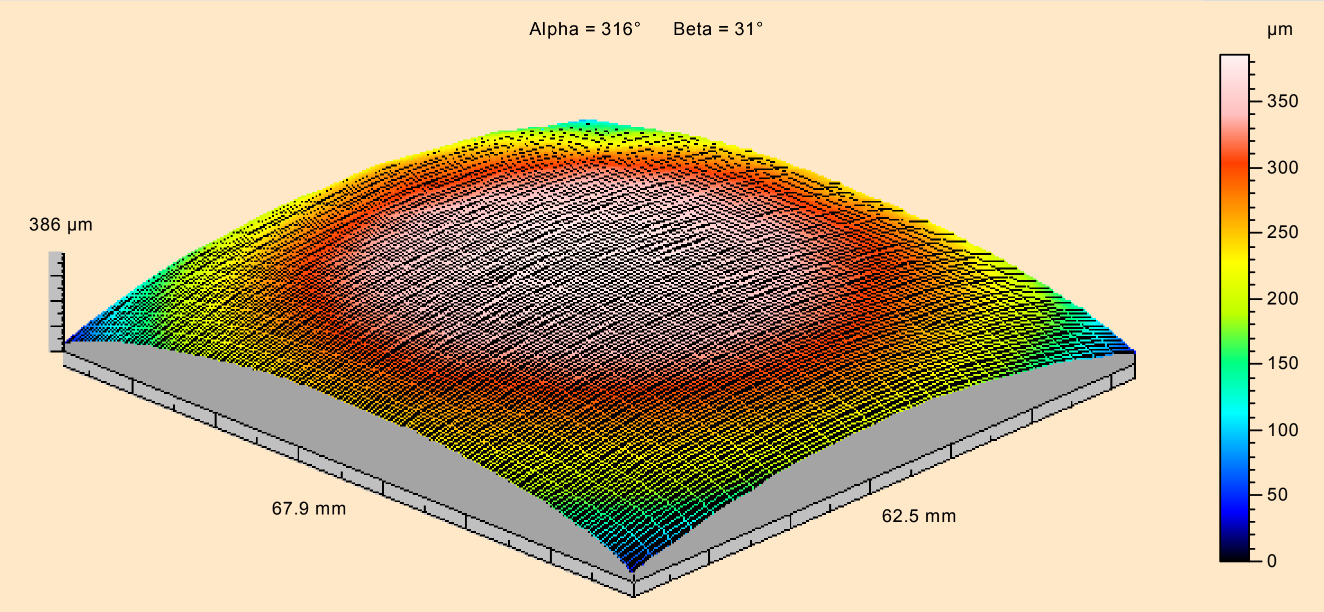
Minimizing camber of sintered bilayers

To minimize cost, mass, volume, and gas diffusion distances, targeted cell thickness is <600 um; thinner cells offer improved flexibility, but also exhibit post-sintering camber due to TEC mismatch

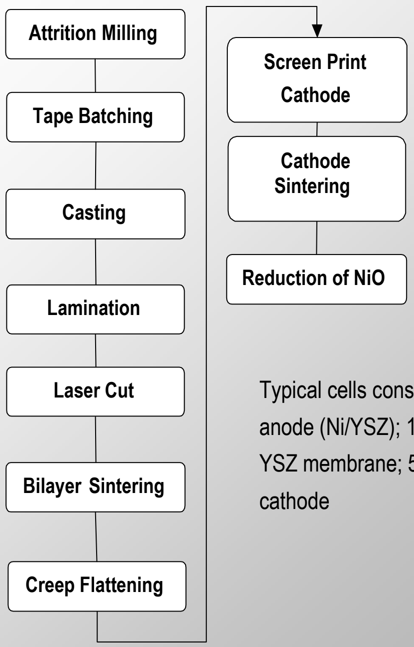


Optimization of bilayer structure and fabrication techniques yield substantial improvement in flatness (reduce camber by 30-40%)

PNNL

Component manufacturing and technology transfer

Cost effective tape casting, screen printing and sintering processes,developed for anode supported cell fabrication, has been transferred to SECA Industry teams for implementations in manufacturing.

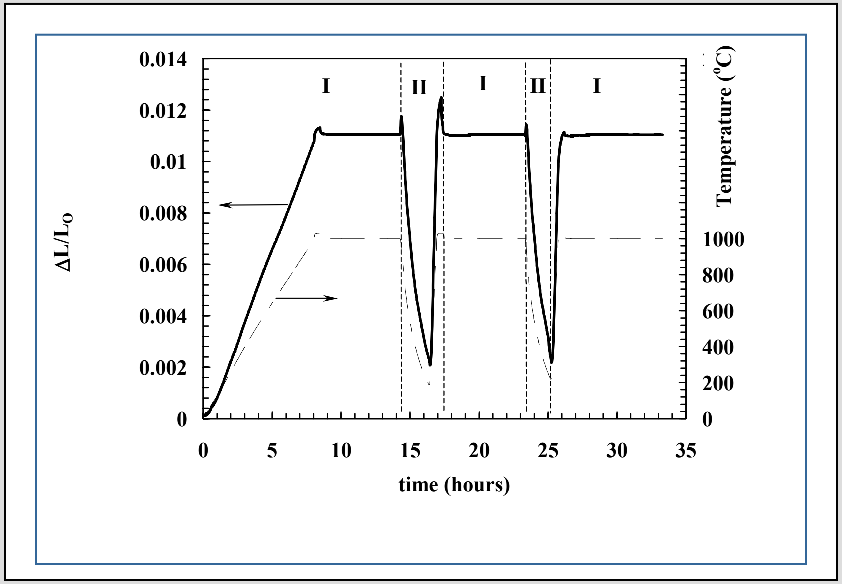


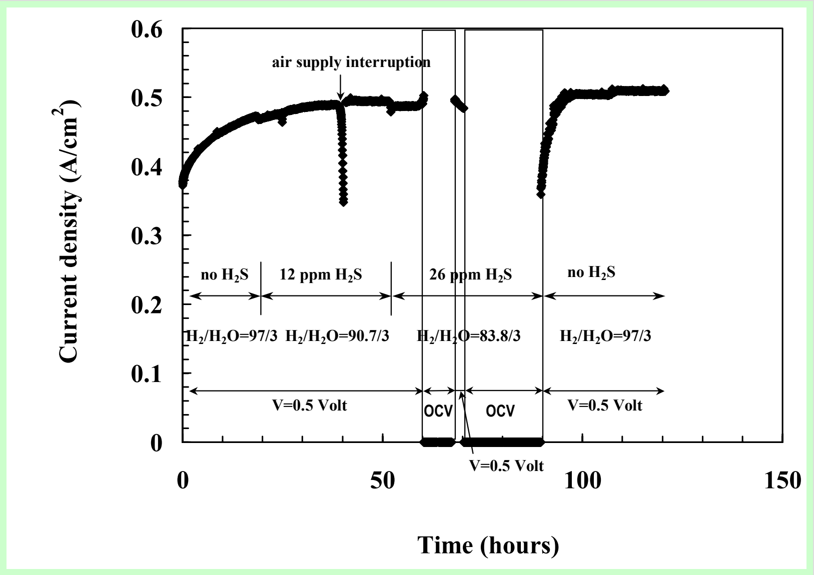
Typical cells consist of ~500 µm thick bulk anode (Ni/YSZ); 10 µm active anode; 8 µm YSZ membrane; 5 µm ceria layer; 25 µm cathode

PNNL

Redox and Sulfur Tolerant Anode Development

Developed SrTiO3 based ceramic anodes offer improved sulfur and redox cycle tolerance compared to conventional Ni/YSZ anodes





PNNL

Interconnection Functional Requirements

An integrated approach for IC development has been designed and implemented

SOFC Interconnection Materials

Electrical
Metal & Oxide electrical conductivity

Chemical
- Resistance to corrosion in multi-component gases
- Resistance to interfacial interactions
- Resistance to embrittlement

Mechanical
- Thermal expansion match
- High temperature yield strength
- Creep resistance

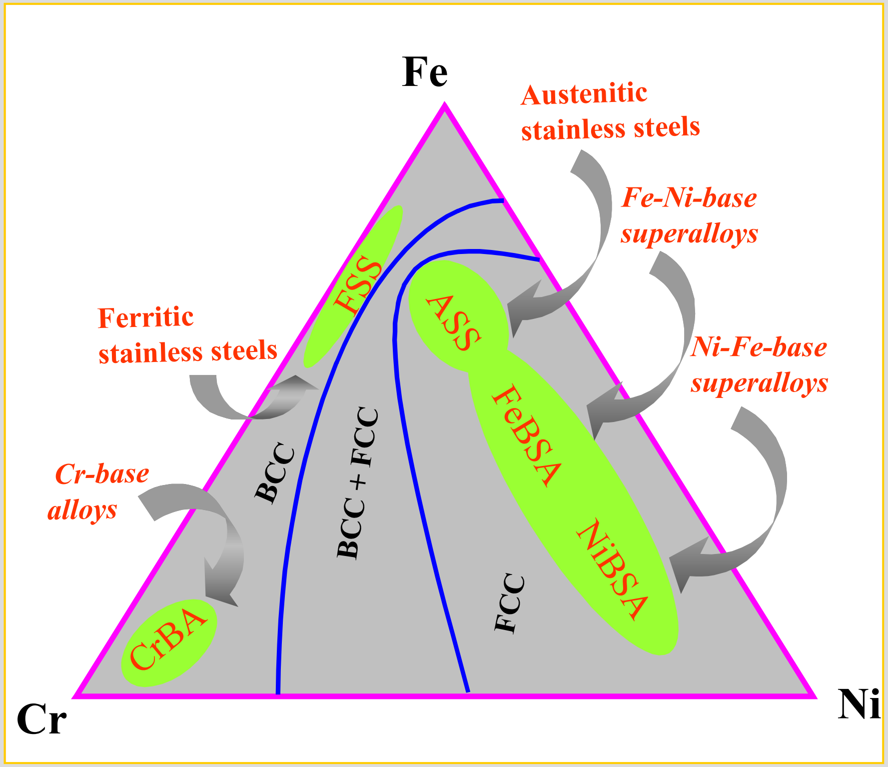
Fabrication
- Fabricability
- Joinability

Cost
- Lower raw materials cost
- Lower processing cost
- Materials of non-strategic importance

A Collaborative Program with Universities, National Laboratories, Manufacturers and Users

Potential Candidates

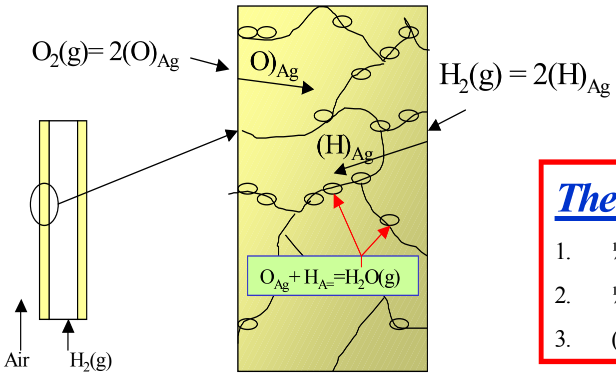
Low cost alloys are being developed for cell to cell interconnection and BOP applications. Materials evaluation studies include oxidation studies, oxide conductivity, seal interactions and metal-oxide interface stability.



- Ferritic stainless steels
- Austenitic stainless steels
- Fe-Ni-base superalloys
- Ni-Fe-base superalloys
- Cr-base alloys
- Co-base superalloys

Accelerated Corrosion in Dual Environment

Mechanistic understanding of corrosion processes are being developed for addressing long term performance of materials



Reaction Steps

1. $(H_2)_g = 2(H)_{Ag}$ Ag/Fuel interface
2. $(O_2)_g = 2(O)_{Ag}$ Ag/ Air Interface
3. $2(H)_{Ag} + (O)_{Ag} = (H_2O)_g$ Bulk Ag

Thermochemical Data

1. $\frac{1}{2}(H_2)_g = (H)_{Ag}$, –G
2. $\frac{1}{2}(O_2)_g = (O)_{Ag}$, –G
3. $(H_2)_g + \frac{1}{2}(O_2)_g = (H_2O)_g$, –G

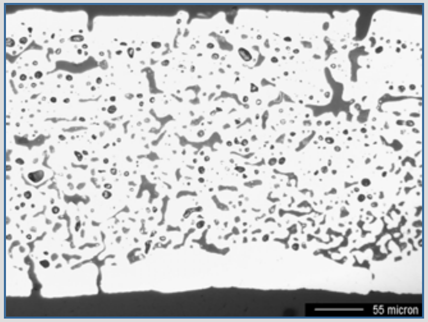
Bulk Degradation Related to :

1.Dissociation and dissolution of H and O in the bulk metal

2. Interaction between dissolved H & O to form High pressure H_2O gas

3. Nucleation and growth of steam bubbles/voids at GB/defects

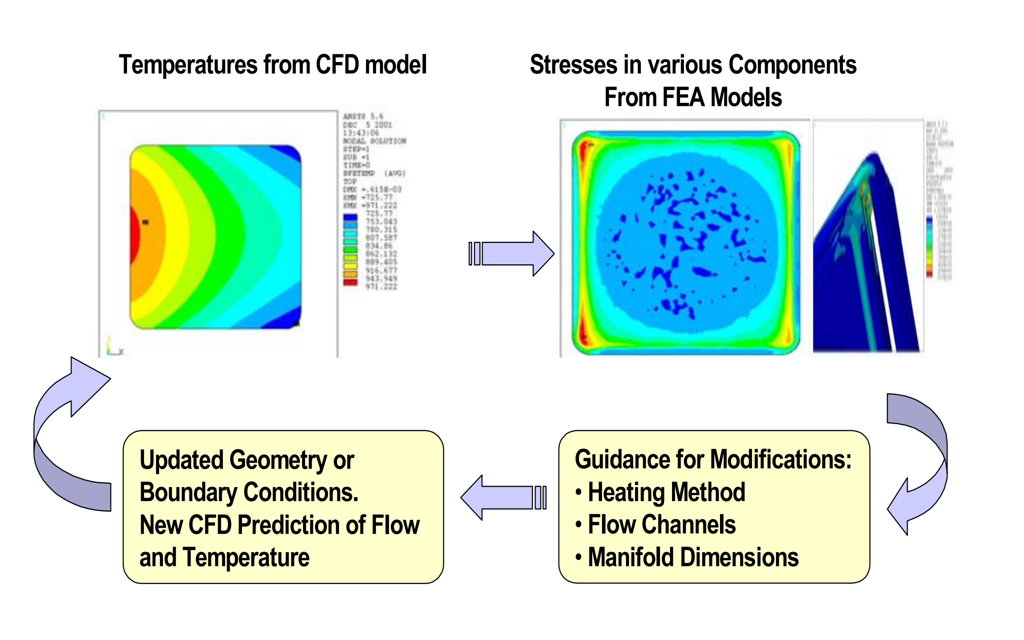
Air/ Air



Air/ Fuel

Modeling – Tools for Stack/system Optimization

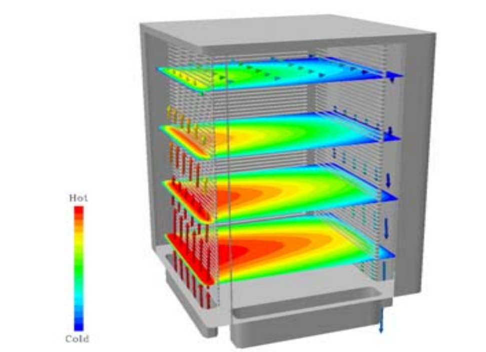
Electrochemical modeling tools allow for the selection and optimization of cell and stack design parameters and geometries



Performance Modeling

Steady state & transient computational modeling and simulations

- *Thermal*
- *Fluid flow*
- *Stress*
- *Electrochemistry*
- *Cell, stack and system modeling*



Current CTP Technology Development Programs

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University of Utah
University of Missouri-Rolla
Interconnects
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CTP Thrust Areas

- Materials Development
  - Cathode
  - Anode
  - Interconnect
  - Seals
- Materials Manufacturing
- Simulation and Modeling
- Fuel Processing
- Power Electronics, Sensors, Controls & Diagnostics

Cathode Development - Challenges

- High electrocatalytic activity towards oxygen reduction
  - High ionic conductivity, high surface exchange kinetics
- Stability (chemical, phase, microstructural, dimensional) at high temperature in oxidizing atmosphere
- Thermal expansion compatible with other SOFC materials
- Minimal chemical interaction with the electrolyte and interconnect materials during fabrication and operation
- High electronic conductivity
- Optimized microstructure to maximize oxygen reduction kinetics:
- Adhesion to electrolyte surface
- Ease of fabrication
- Low cost

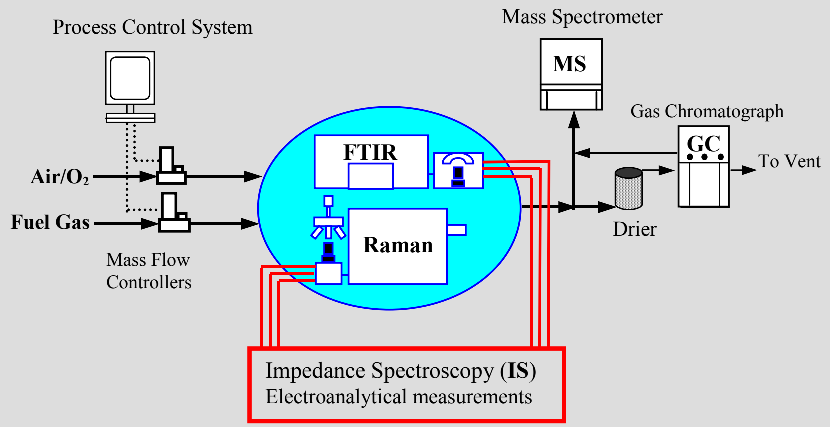
CTP Cathode Development Activities

- **Goals:**
  - **Improved understanding of processes occurring at cathode/electrolyte interfaces**
  - **Improved intermediate temperature cathode materials**
- **Participants:**
  - **Univ. of Washington**
  - **Functional Coating Technologies, LLC**
  - **Univ. of Utah**
  - **Univ. of Missouri-Rolla**
  - **Georgia Tech Research Corporation**
  - **Argonne National Lab**
  - **Lawrence Berkeley National Lab**
  - **Pacific Northwest National Lab**

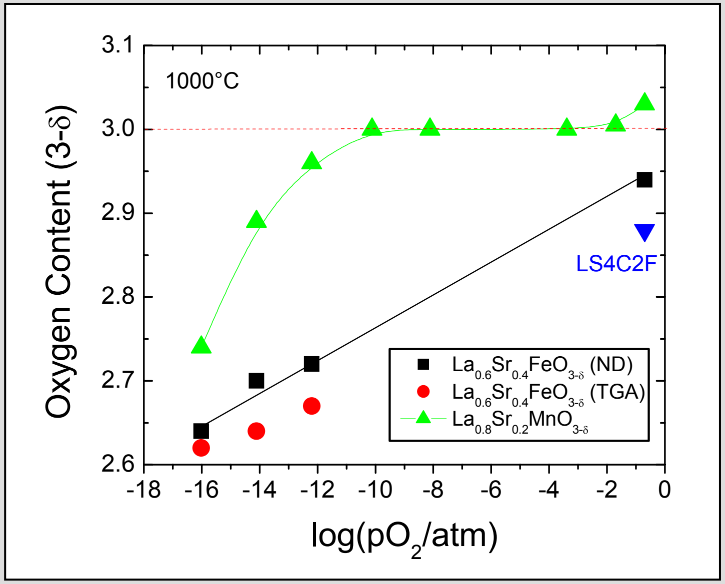
Improved understanding of cathode materials and processes

SECA

- Investigations of cathodic reactions using *in-situ* FTIR/Raman, IS, and MS/GC



Georgia Tech Research Corporation



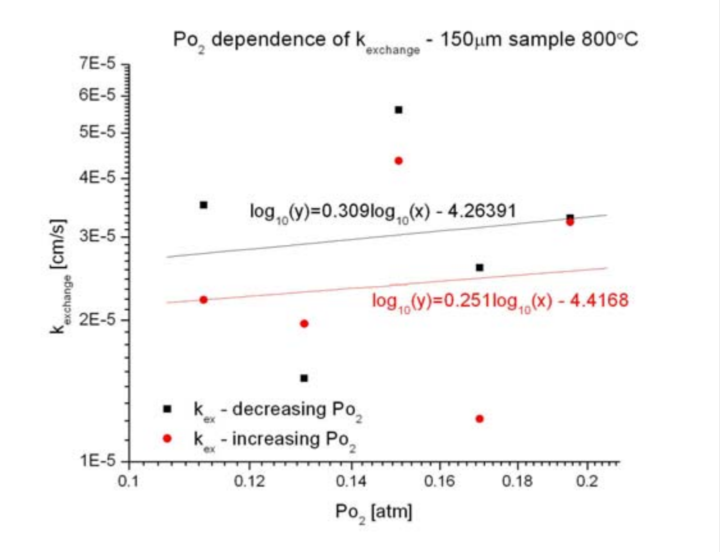
- **Characterization of Cathode Material Defect Structures**

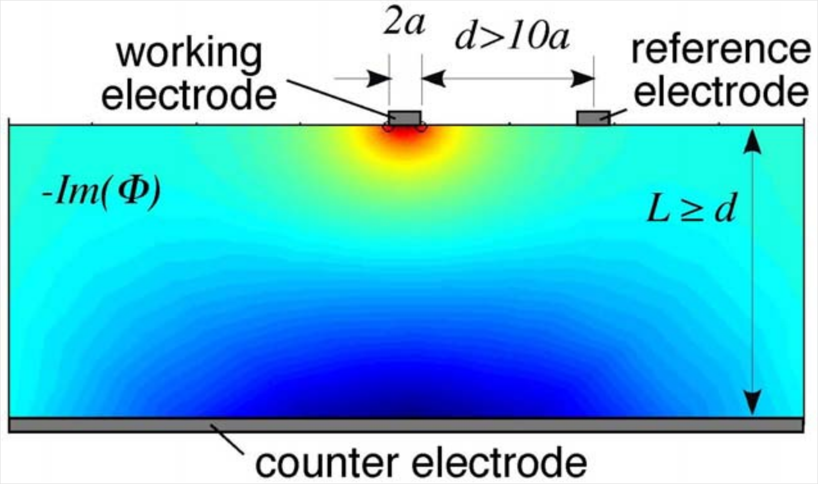
University of Missouri-Rolla

Improved understanding of cathode materials and processes

- Characterization of oxygen surface exchange and diffusion coefficients of cathode materials

University of Utah



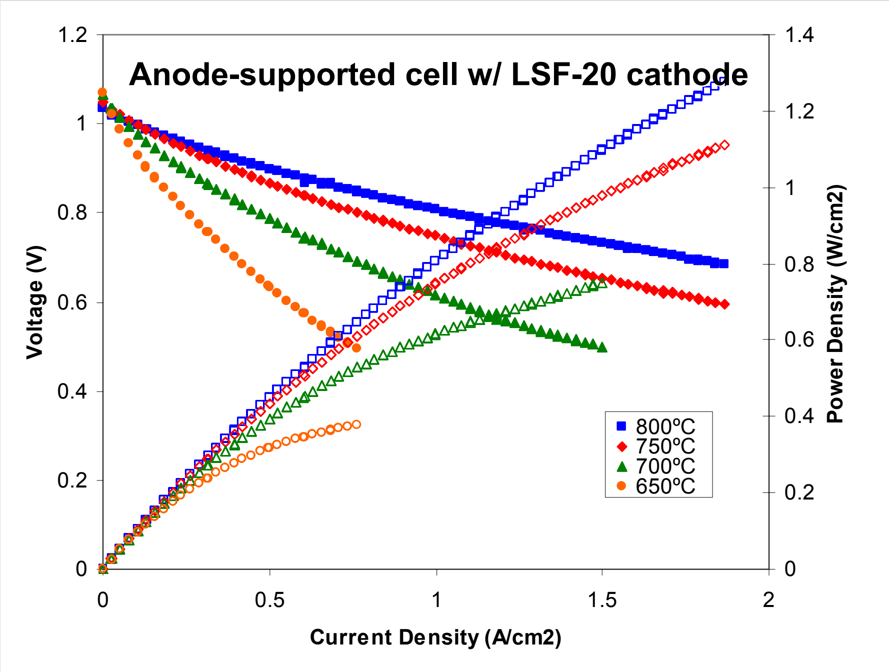


University of Washington

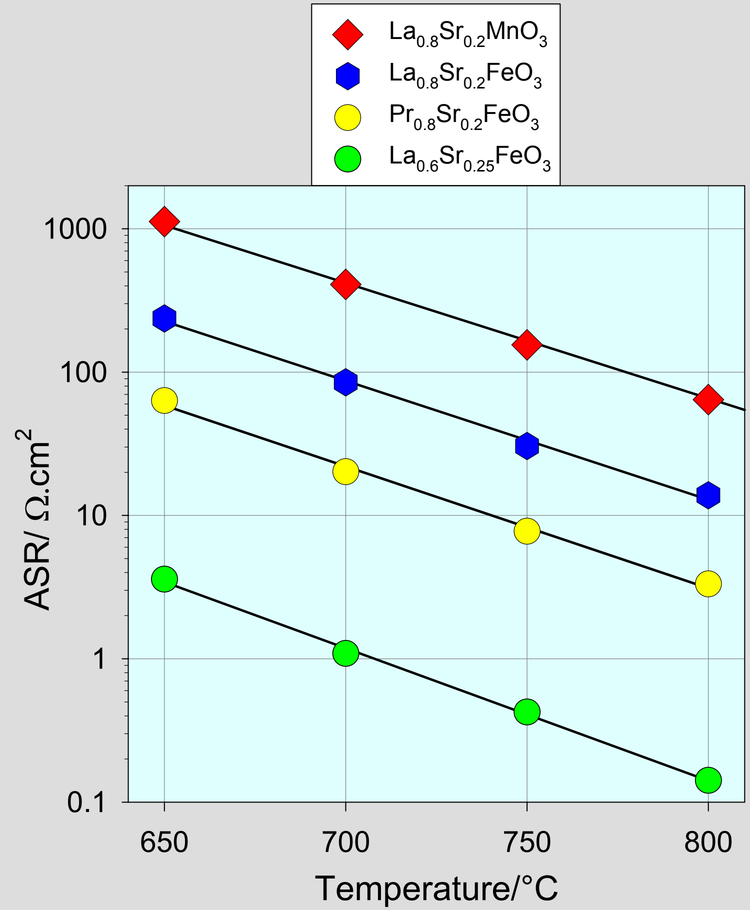
- Microelectrode development for better and faster measurements of cathode response
- Development of nonlinear impedance techniques to resolve overlapping physical mechanisms

Improved intermediate temperature cathode materials

- Mixed Conducting Cathodes
  - Doped Lanthanum Ferrite Cathodes



PNNL

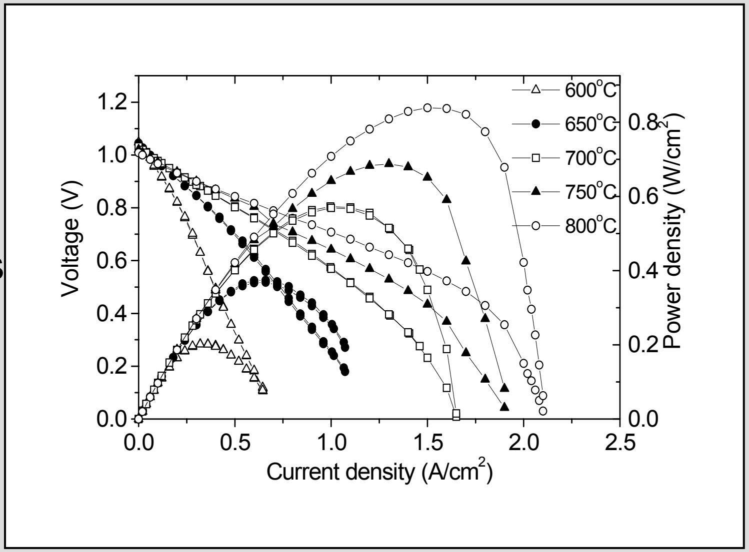


ANL

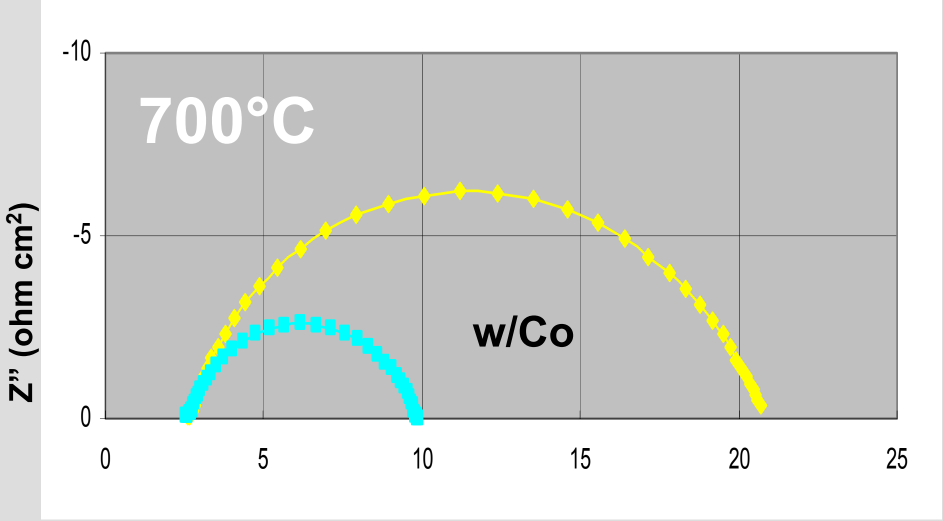
Improved intermediate temperature cathode materials

- **LSCF-GDC composite cathodes**

Functional Coating Technologies



LSM Cathode Impedance



- **Co-doped LSM cathodes**

LBNL

Anode Development - Challenges

- **Requirements:**
  - **High electronic conductivity**
  - **Excellent electrocatalytic activity for fuel oxidation**
  - **Adequate porosity for gas transport;**
  - **Chemical and physical compatibility with YSZ:**
  - **Long-term dimensional and microstructural stability**
- **Conventional Ni/YSZ cermet has proven adequate for operation on clean H_2 or fully reformed fuels, but has other limitations, including:**
  - ***Tolerance to sulfur in fuel***
  - ***Stability under reducing and oxidizing conditions***
  - ***Direct utilization of hydrocarbon fuels***

CTP Anode Development Activities

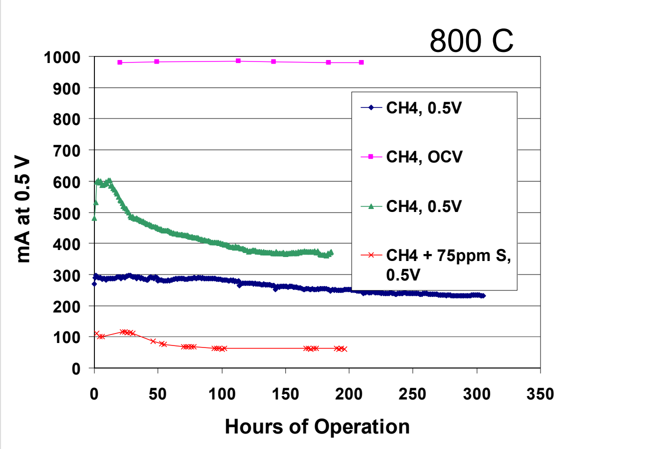
- Goal: Advanced anodes offering redox, sulfur, and hydrocarbon tolerance

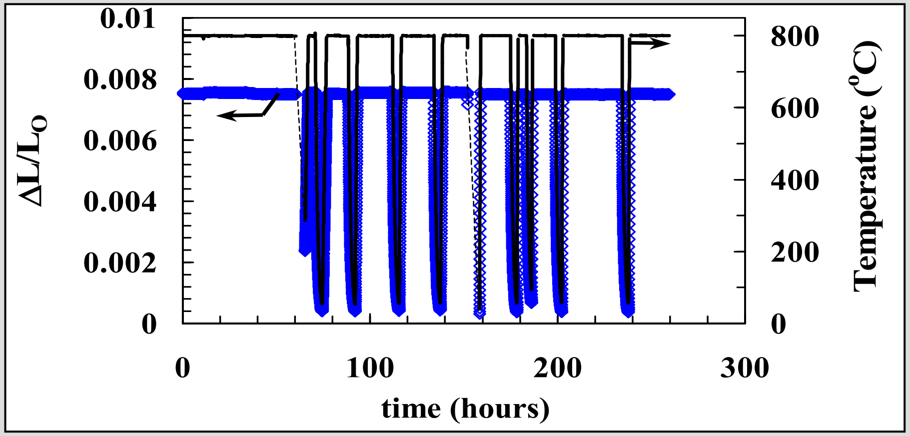
- Participants:
  - Gas Technology Institute
  - Northwestern University
  - Pacific Northwest National Laboratory

CTP Anode Development Activities

- Cu-based Anodes (GTI)
  - Operation on dry methane w/ sulfur
- Ni-based Anodes (Northwestern U.)
  - Improved understanding of operation on methane
- Ceramic anodes - doped titatate/ceria mixtures (PNNL)
  - Sulfur and redox tolerant

GTI





PNNL

SOFC Interconnects: Challenges

Mechanical and chemical stability:

- High temperature oxidation/corrosion resistance
- Multi component gas streams (H_2O, CO_2, O_2 etc.)
- Changing fuel composition (as result of fuel utilization)
- Simultaneous fuel and oxidant gas exposures
- Isothermal (high temperature) and thermal cyclic exposures

Low resistance path for electric current

Low materials and fabrication cost

Preferred high temperature interconnect material: Doped lanthanum chromite

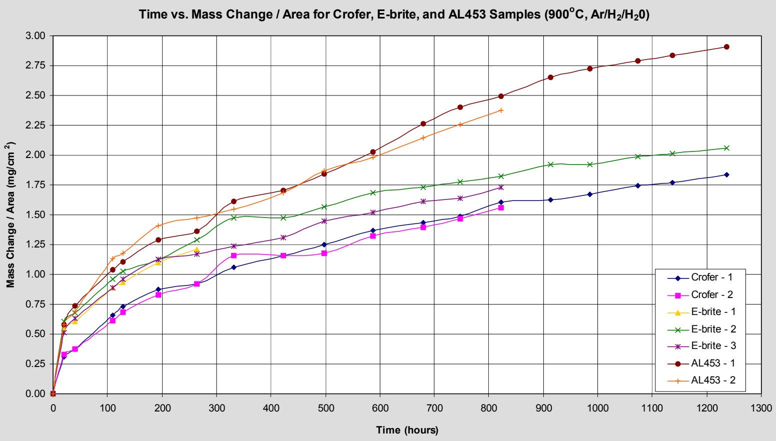
High temperature alloys may satisfy these requirements for lower temperature (<800ºC) SOFC stacks

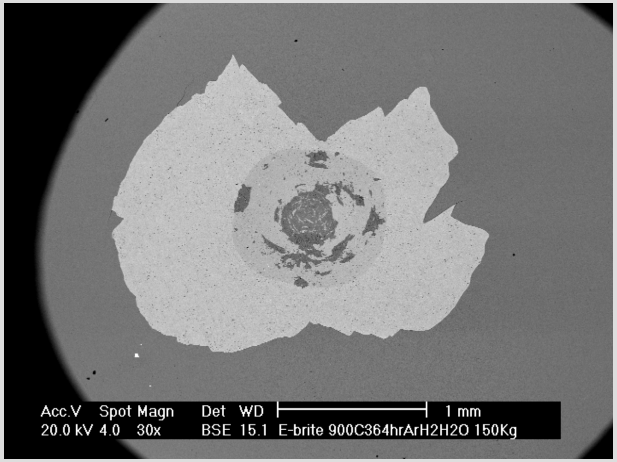
CTP Interconnect Activities

- Goals:
  - Improved understanding of chemical and thermomechanical stability of alloys
  - Optimization of interconnect alloys (bulk and/or surface Modification)
- Participants:
  - Ceramatec, Inc
  - Southwest Research Institute
  - University of Pittsburgh
  - Lawrence Berkeley National Lab
  - Argonne National Lab
  - National Energy Technology Lab
  - Pacific Northwest National Lab

Improved Understanding of Chemical and Thermomechanical Stability

- Evaluation of Candidate Alloys
  - Emphasis on Ferritic Stainless Steels
  - Oxidation Behavior
    - Scale growth rate
    - Scale Chemistry
    - Scale Adhesion
    - Evaporation of Cr

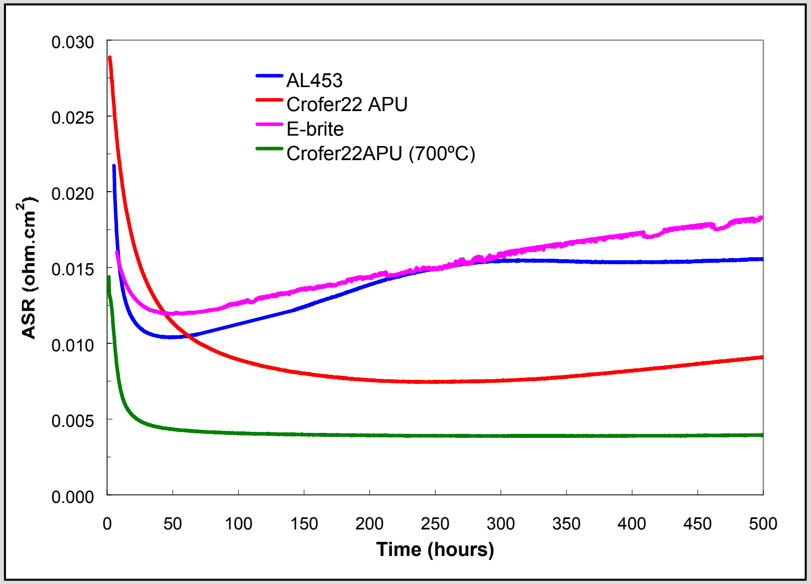


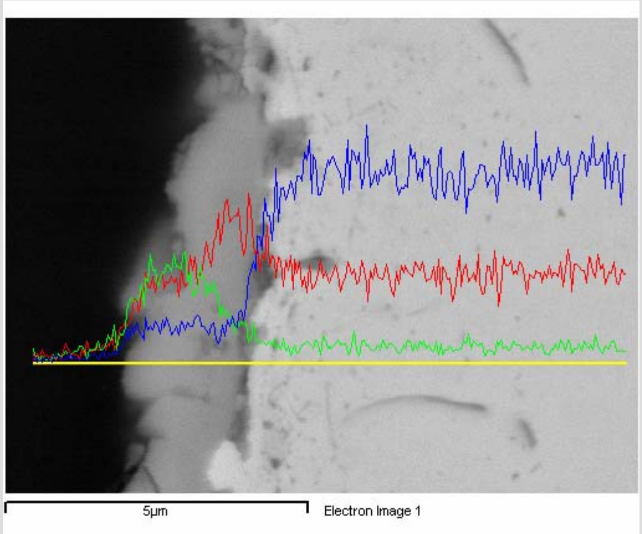


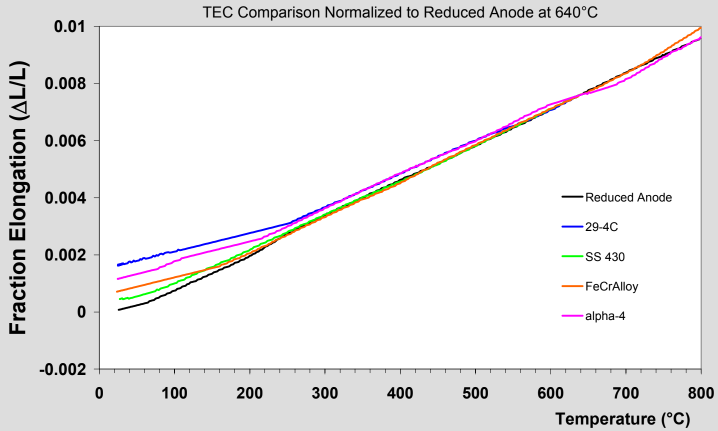
Univ. of Pittsburgh

Improved Understanding of Chemical and Thermomechanical Stability

- Evaluation of Candidate Alloys
  - Scale Resistance Measurements
  - CTE Measurements
  - Dual Atmosphere Tests







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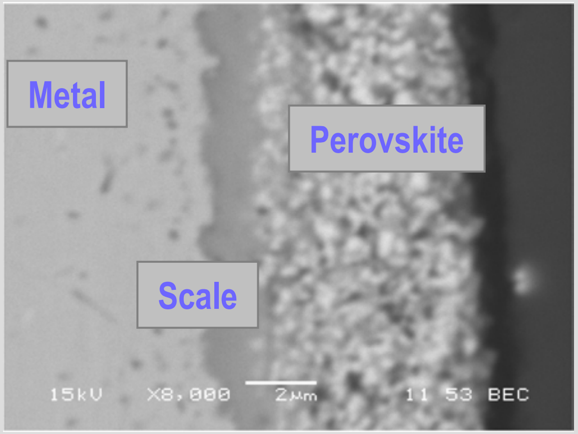
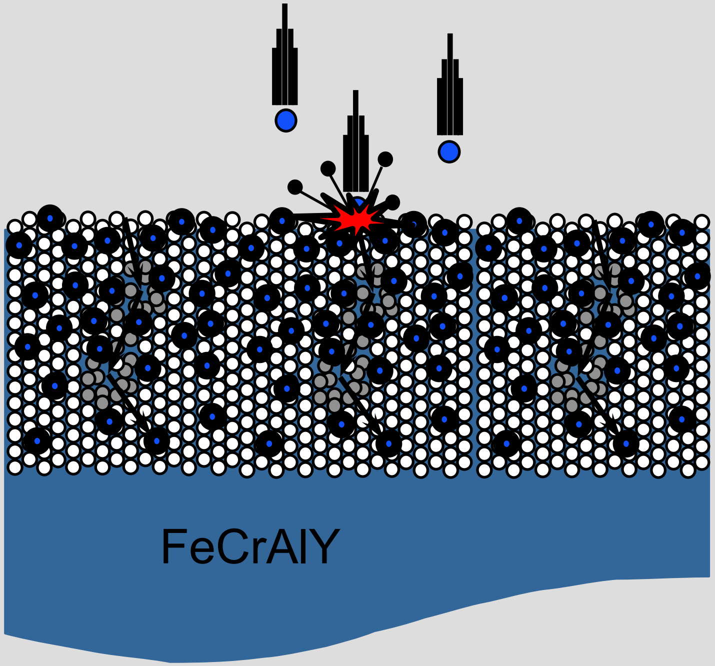
Optimization of Interconnect Alloys

- **Perovskite Coatings to Reduce Interfacial Resistances**

Ceramatec



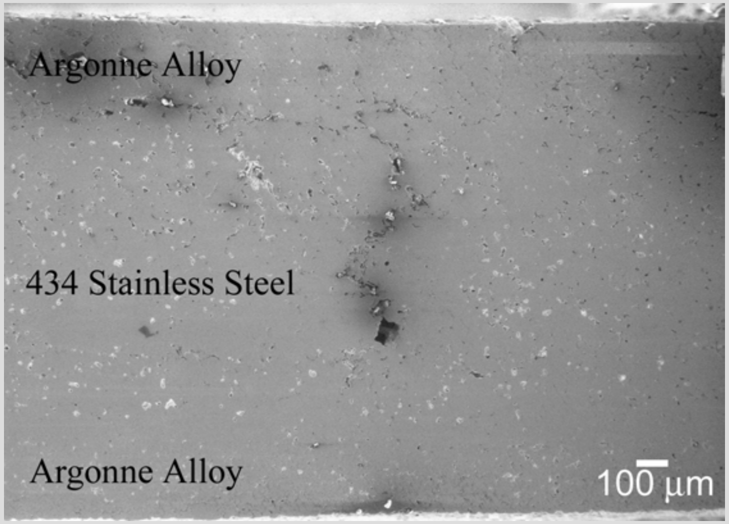


Southwest Research Institute

- **Ion implantation to create a thermodynamically stable and conductive alumina-based scale**

Optimization of Interconnect Alloys

- Fe-Cr Alloys w/ Rare Earth Additions
- Ferritic Alloys Without Chromium
- Compositionally Graded Interconnects



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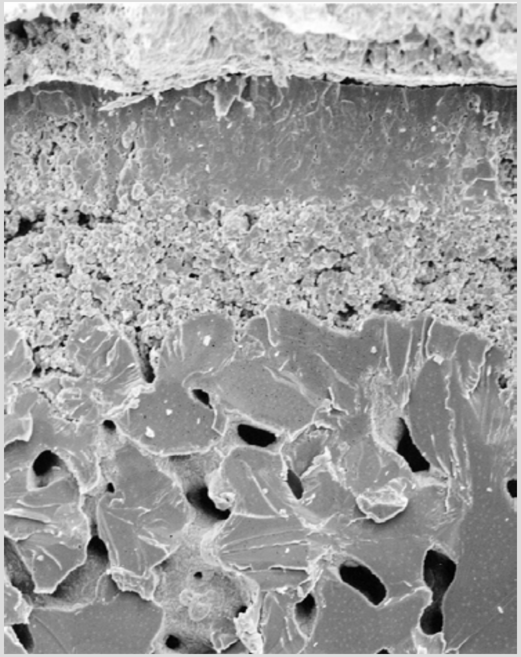


- Alloy-supported SOFCs

LBNL

Seal Development - Challenges

Requirements for seals in planar SOFC stacks

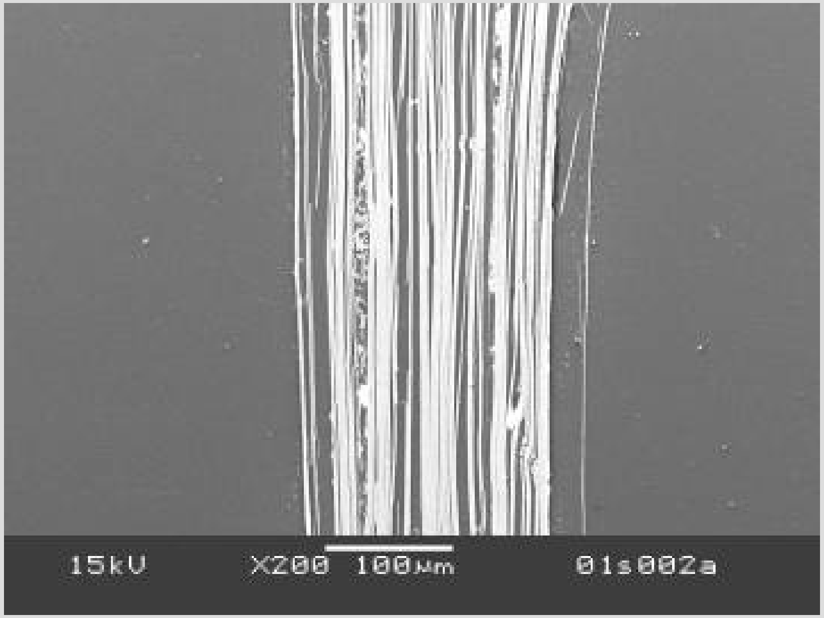
- High degree of sealing (hermetic or allowable leak rate) under minimal compressive load
- Matching CTE (especially for rigid seals)
- Electrically insulating
- Long-term stability at high T in oxidizing/reducing and humid environments
- Inexpensive
- Thermal cycle stability
- Chemically and physically stable
- Thermal shock resistance

Rigid seals (i.e., glass-ceramic) require very close CTE matching of all stack components to minimize stresses

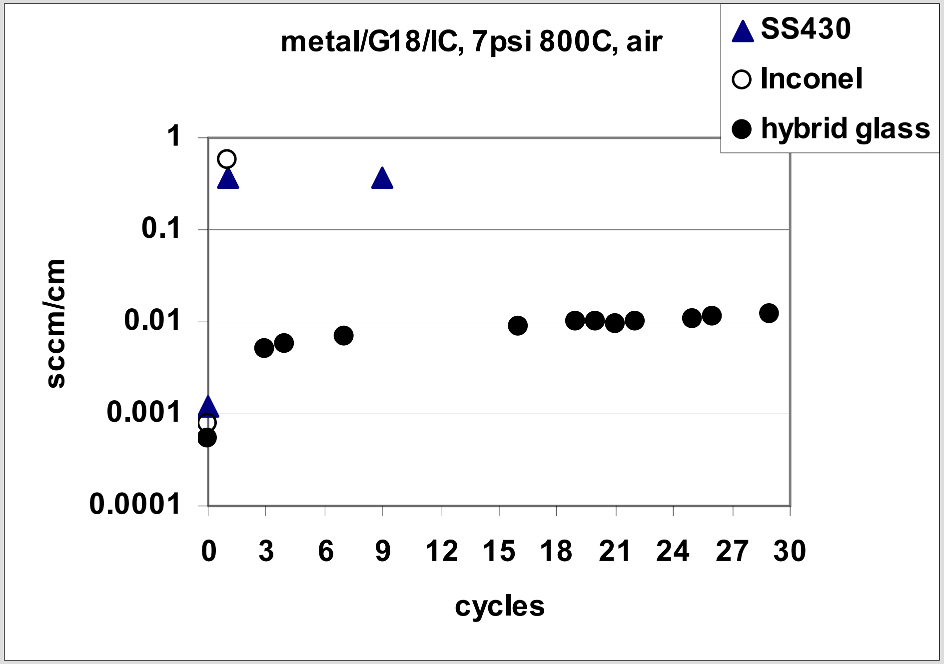
Compressive seals may relax CTE matching requirements by providing compliance in "x-y" plane.

CTP Seal Development Activities

- Modified mica-based compressive seals



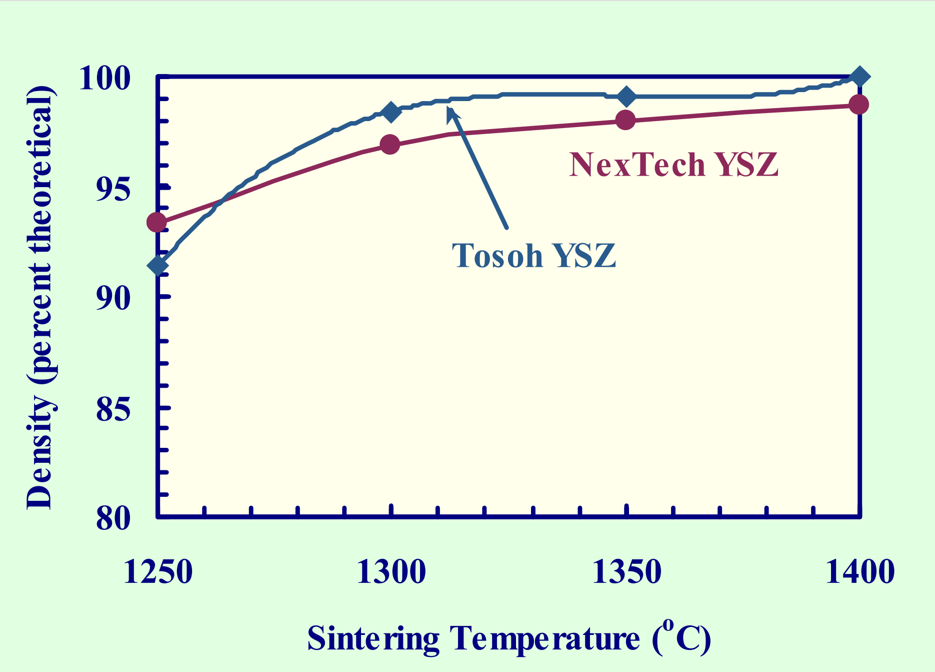
Pacific Northwest National Lab



Improved tolerance to thermal cycling (relative to glass-ceramic seals)

Materials Manufacturing Activities

- Manufacturing processes for low-cost, high-quality yttria-stabilized zirconia electrolyte powders
  - **NexTech Materials, Ltd.**
  - **University of Utah**



NexTech Materials

Fuel Processing--SECA Core Technology

- 3 Primary Topics Identified by Industry
- Topic #1 - Fuel Reforming Fundamentals
  - Major reforming approaches (SMR, ATR, CPOX)
    - Catalyst activity, cost
    - **Kinetics, mechanisms**
    - Effects of fuel type on performance
    - **Deactivation by sulfur, carbon**
  - **Reaction/reactor models**

Fuel Processing--SECA Core Technology

- Topic #2 - Catalyst Status and Development
  - Improved catalysts
    - Dual function catalysts for CPOX, ATR
    - Dry reforming catalysts to reduce H_2O
    - **Sulfur tolerance of reformer, anode**
    - Multi-fuel catalysts
    - Assistance with catalyst characterization, especially post-mortem
    - Faster light-off times for CPOX
  - **Identify state-of the-art in fuel reformation, especially diesel reformation**

Fuel Processing--SECA Core Technology

- Topic #3 - Sulfur Cleanup
  - Subtopics:
    - Desulfurization of liquid fuels
    - High temperature H_2S removal (between sulfur tolerant reformer, anode)
    - Higher capacity, more effective natural gas desulfurization
    - **Sulfur tolerant anodes**

Current Core Technology Efforts to Address Industry Needs in Fuel Processing

- Two contracts awarded and recently initiated (Core Program Solicitation) for sulfur-tolerant / direct oxidation anodes
  - Northwestern University
  - Gas Technology Institute/Technologix Corporation
- Ongoing efforts at NETL in diesel reforming
  - Systems analysis for integration and operational requirements
  - CFD Modeling for heat and mass transfer and reactor performance
  - Kinetic rate measurements for predictive modeling and design
- Ongoing efforts at LANL
  - Investigation of carbon formation in diesel reforming
  - Fuel/steam/air mixing and injection to reformer
  - Catalyst regeneration - de-activated by surface carbon
- Beginning efforts at PNNL
  - Sulfur tolerant reforming catalysts
  - Carbon formation-resistant catalysts

CTP Simulation and Modeling

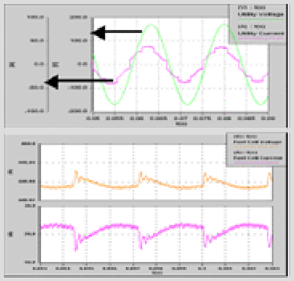
- Goal: Develop tools for optimizing design, predicting performance, minimizing cost, and assessing reliability and lifetime of SOFC cells, stacks, and systems.
- Participants:
  - Univ. of Florida
  - Univ. of Illinois
  - Virginia Tech
  - Georgia Tech
  - TIAX
  - Oak Ridge National Laboratory
  - National Energy Technology Laboratory
  - Pacific Northwest National Laboratory

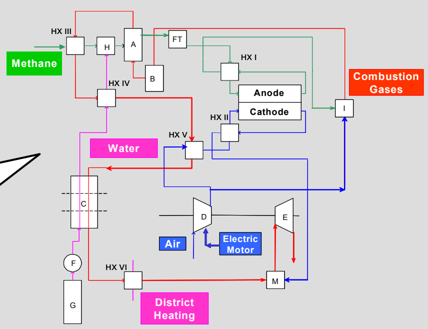
Integrated SECA Core Modeling & Simulation Activities

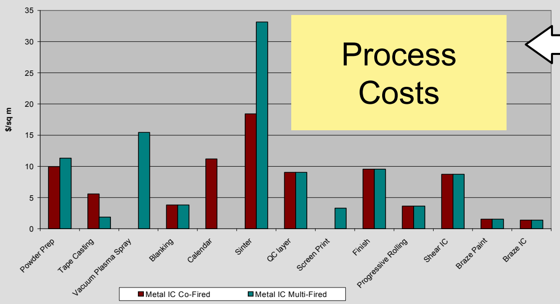
SECA

Consolidated Computational Modeling for SECA

Training







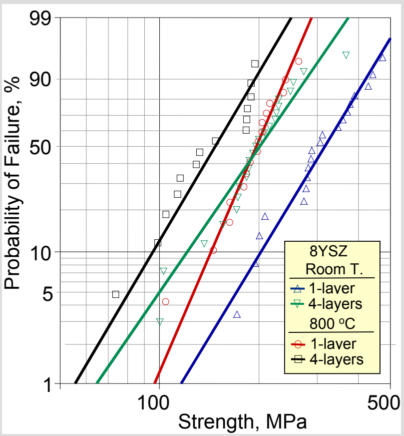
Cost Analysis

Electrical Power System

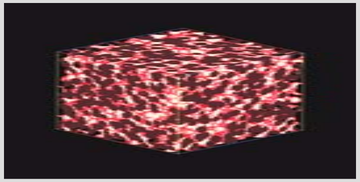
Thermal System

Stack

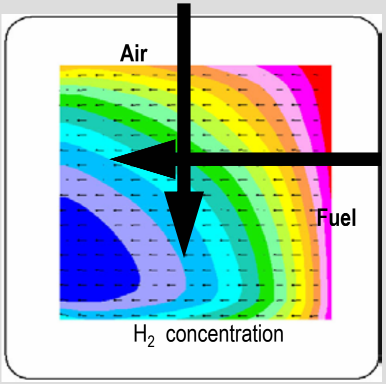
Cell



Experimental Verification



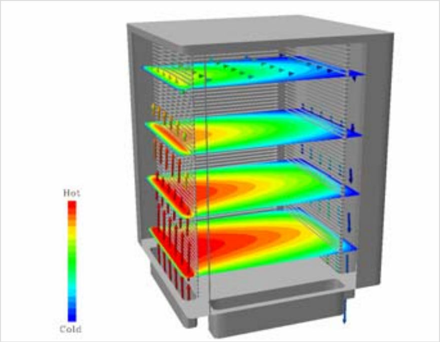
Microstructural electrochemistry



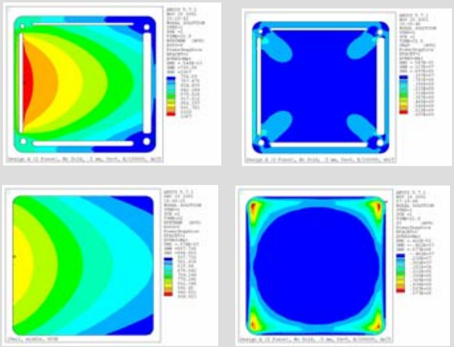
CFD and FEA Models for Transient and Steady State Operation

Flow-Thermal-Stress Models

PNNL is developing modeling tools for rapid start-up and thermal transients; working with GT on integration of failure models in stack models.

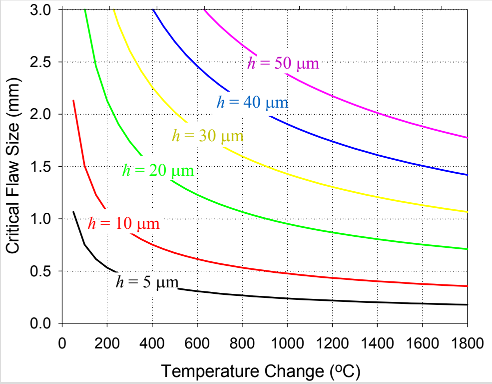


Flow Uniformity and Controls



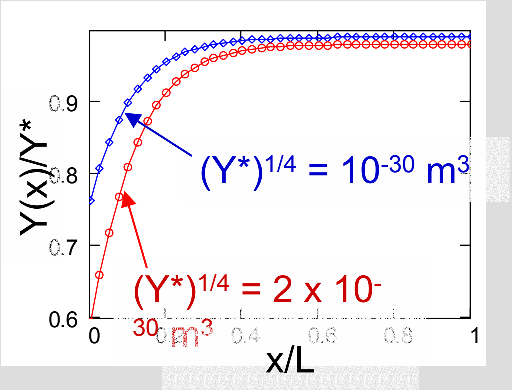
Rapid Start-up Stress Analysis

GT is developing modeling tools for thermomechanical failure of fuel cell materials



Buckling Induced Delamination.

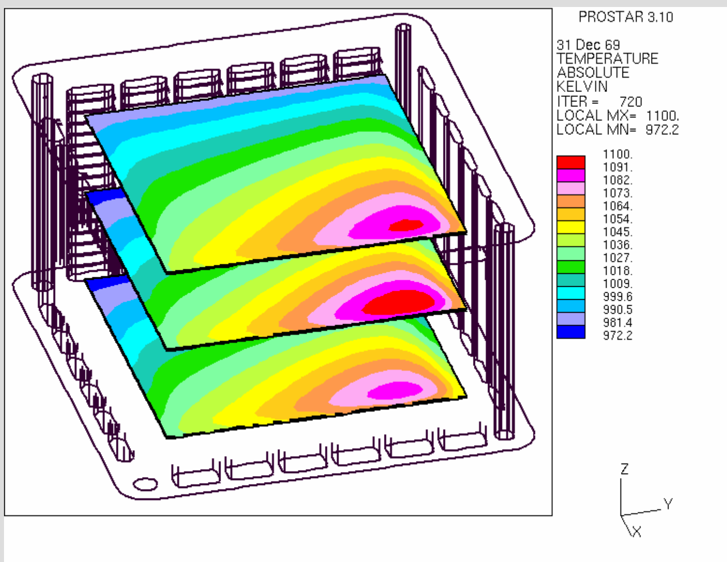
UoF is developing modeling tools for predicting effects of defects on mechanical properties



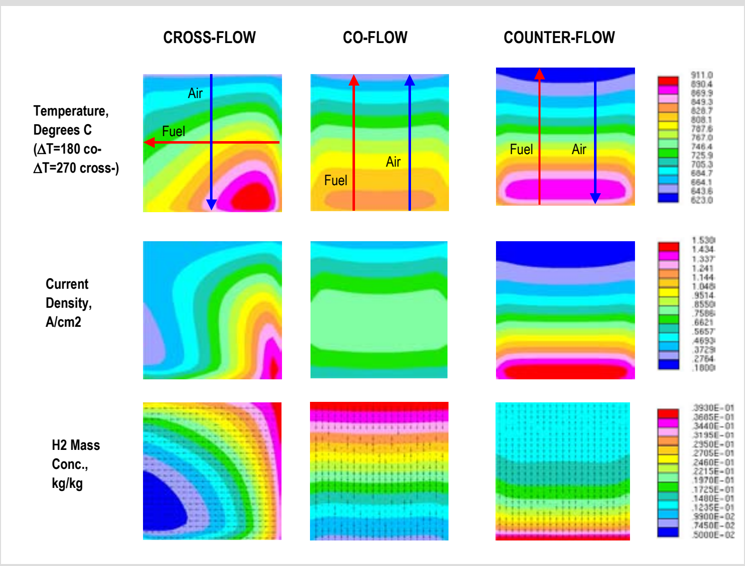
Buckling Induced Delamination.

Continuum Electrochemistry Models

PNNL and NETL are developing stack level electrochemistry models based on FEA and CFD frameworks to optimize design during steady state operation.



Temperature profile for 16-cell stack during steady state

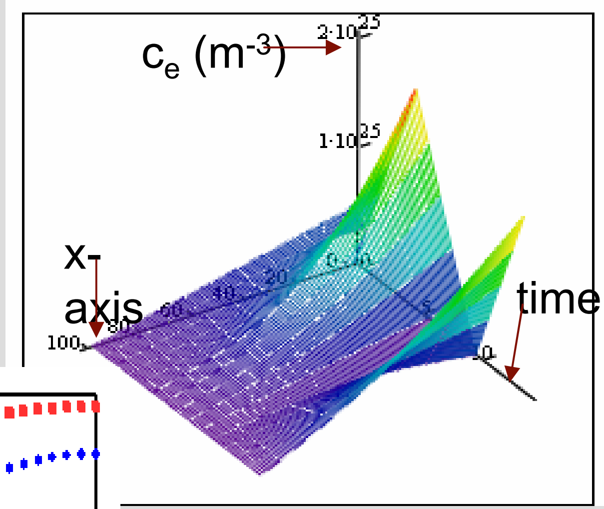


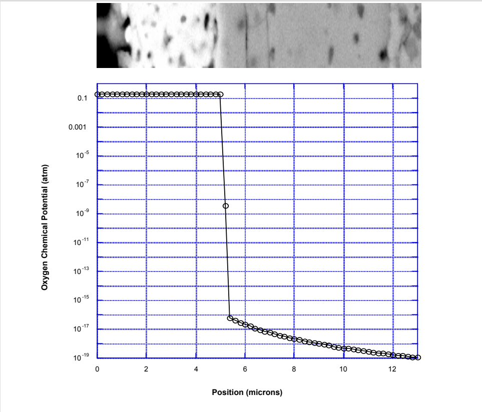
Temperature, current density and H2 concentration for cross-flow, co-flow and counter-flow designs

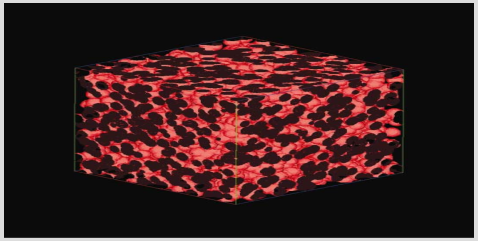
Microstructural Electrochemistry

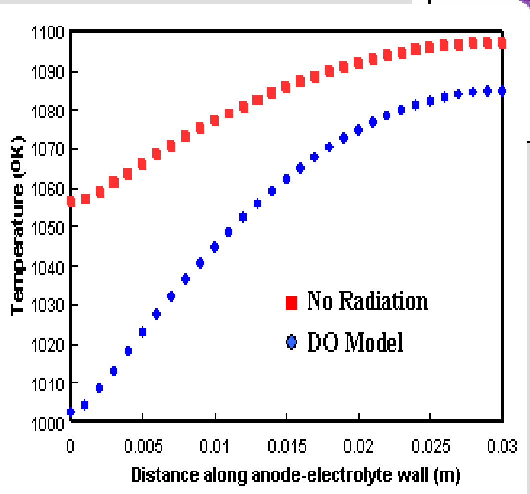
PNNL is developing coupled CFD and chemical models for design and optimization of cell material microstructure and understanding detailed electrochemical mechanisms at the microstructure level

UoF is developing electrochemical performance models using defect density and will be utilized in PNNL's models





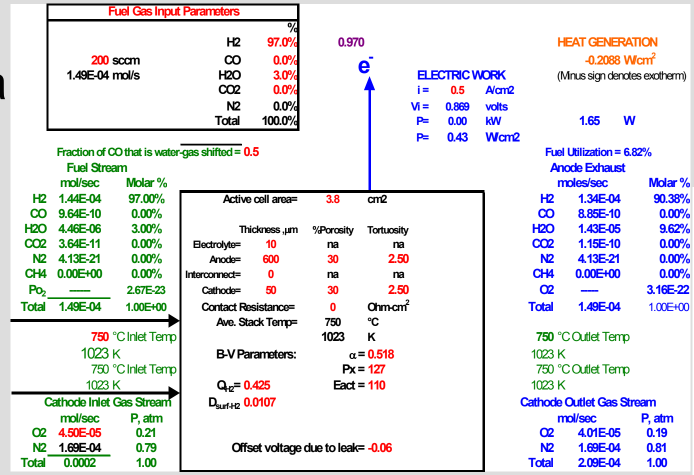




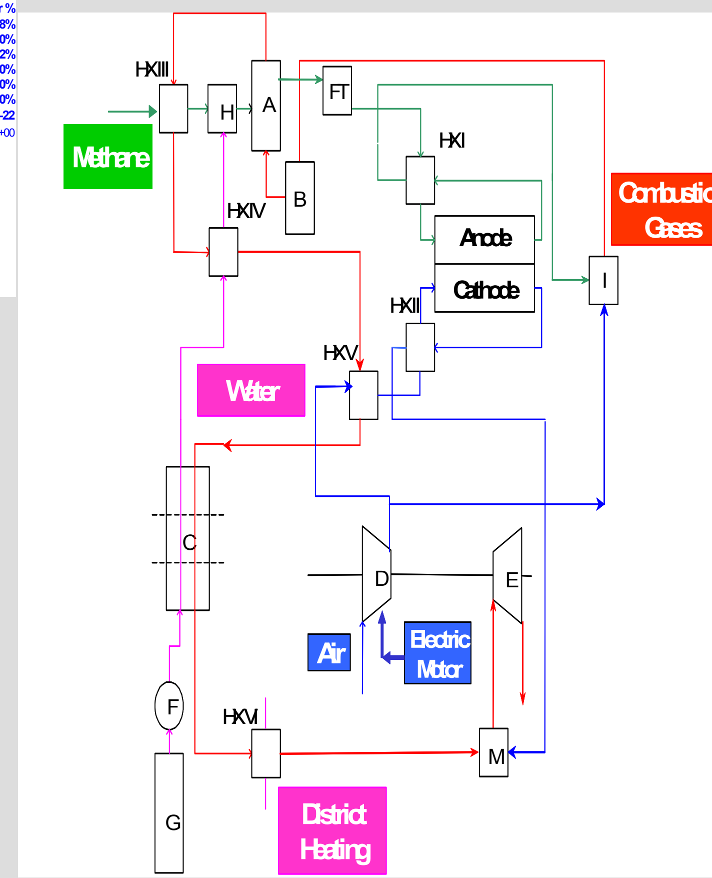
GT is developing thermal models for internal thermal radiation in porous media to be incorporated in PNNL's models

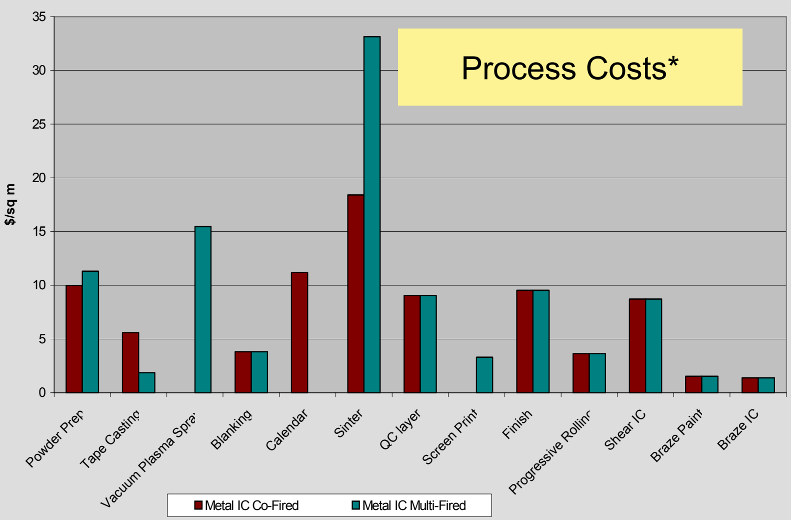
System and Cost Models

PNNL is developing a stack model which has been incorporated by vertical teams in system and control software



GT & VT are developing subsystem models

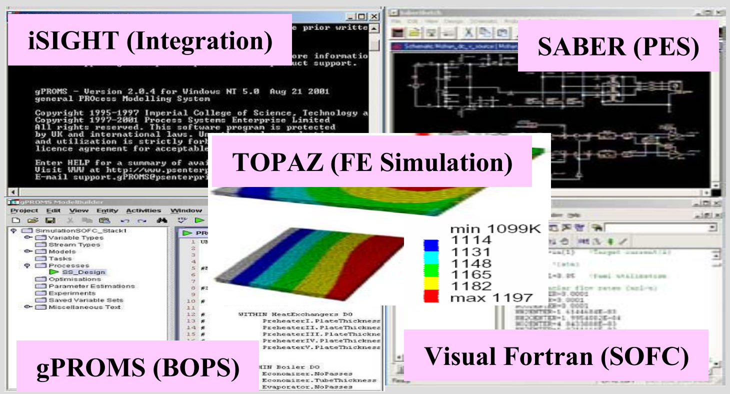




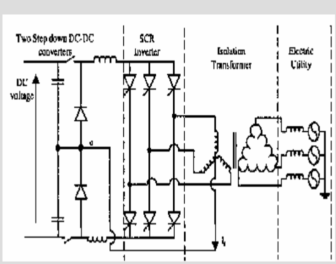
TIAX is developing tools to estimate the cost impact of manufacturing and design decisions

Modeling Electrical Interactions

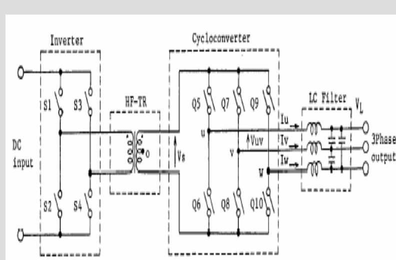
UoI is developing integrated SOFC system models for optimizing interactions between SOFC, power conditioning system, and application load



SOFTWARE SYSTEM INTEGRATION



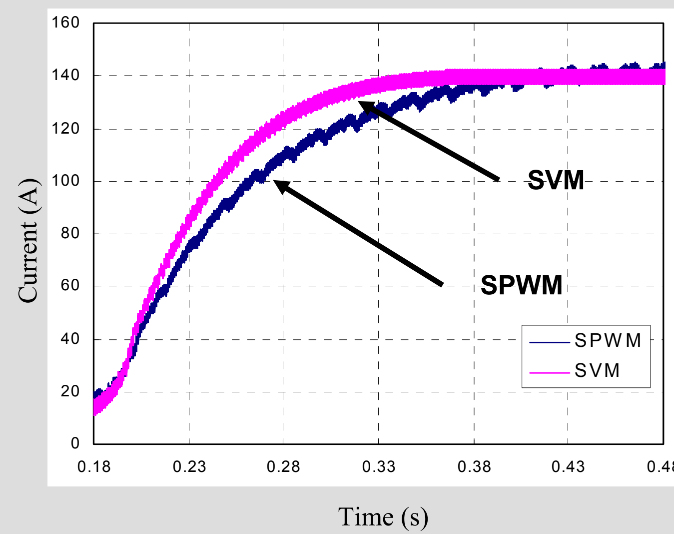
Line Commutation



Transformer Assisted

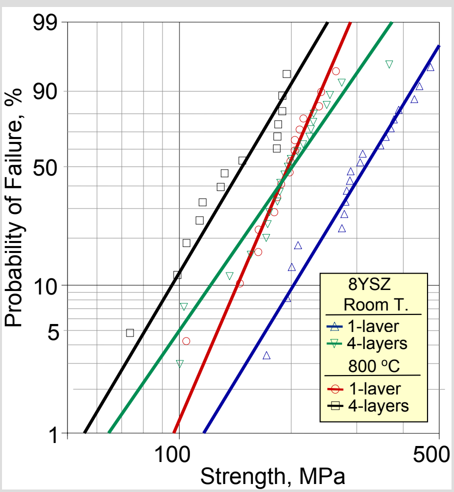
VT is developing advanced control and modulation strategies

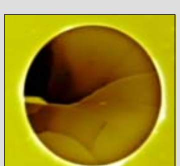
Load transient
(no load to full load in 0.15s)

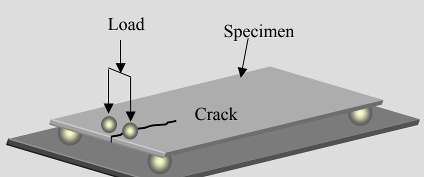


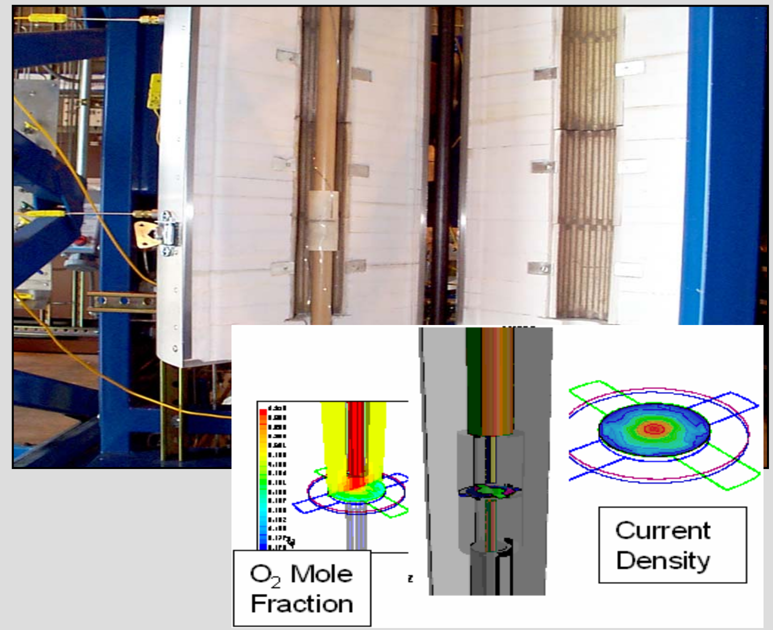
Experimental Data and Model Validation

ORNL is evaluating the behavior and properties of PEN materials and identifying mechanical failure mechanisms









NETL and PNNL are using cell tests to validate electrochemistry models

SECA

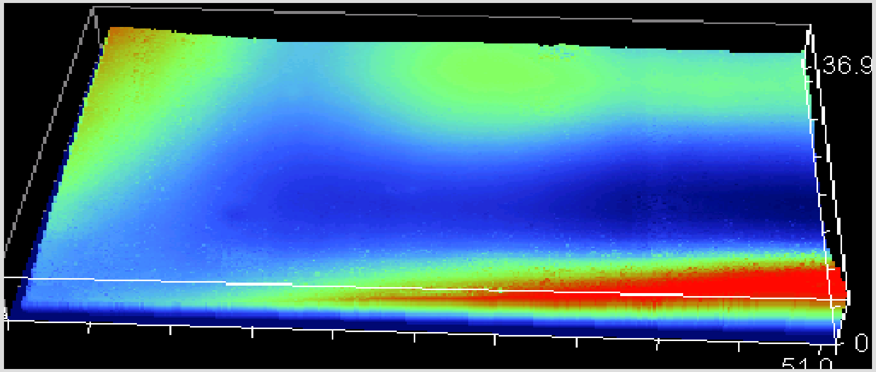
Software Transfer and Training

Entire CTP modeling team will participate in SECA industry team training

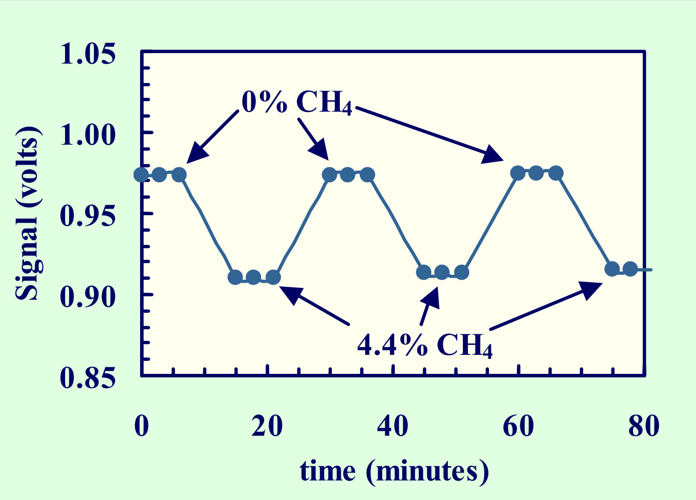
Models will be put on a consolidated computing hardware for easy access and use by industry team members

Power Electronics, Sensors, Controls & Diagnostics – CTP Activities

- Ohio State University
  - Laser Dilatometry & Photo Optometry



- Texas A&M University
  - Cost effective DC to AC converters for SOFC power systems
- Virginia Polytechnic Institute & State University
  - Cost effective DC/DC converters for SOFC power systems
- NexTech Materials, Ltd
  - Hydrocarbon & Sulfur Sensors for SOFC Systems



Summary

- Technology gaps have been identified and R&D programs have been developed to bridge these gaps.
- Active participation from SECA industry teams has led to prioritization of R&D activities.
- Long-term reliability of SOFC materials, components, and systems is being emphasized through R&D projects .
- Communication channels have been developed among SECA participants for timely dissemination of results.
- Advanced computational tools and materials technology developed under the CTP have been transferred to the SECA industry teams.

Acknowledgements

- Office of Fossil Energy, U.S. DOE
- NETL SECA Program Management for programmatic support
- CTP contributors from participating Universities, National Laboratories, and R&D organizations
- SECA industry teams for valuable guidance and suggestions

SECA Modeling – Tools for Stack/system Optimization

SECA

Electrochemical modeling tools allow for selection and optimization of gas flow patterns and stack geometries