

Low-Cost Manufacturing of Multilayer Ceramic Fuel Cells

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SECA Core Technology Workshop
Pittsburgh, PA
June 18, 2002

- ❑ **Overview of MLFC Program**

- ❑ **Results**

- **Fabrication Process Development**
- **Electrical and SOFC Testing**
- **Mechanical Property Testing**

- ❑ **Ceria-Based Electrolytes**

- ❑ **Closing Comments**

Low-Cost Manufacturing of Multilayer Ceramic Fuel Cells

DOE Contract No. DE-AC26-00NT40706

Program Manager: Bill Dawson

Principal Investigator: Scott Swartz

NETL Project Manager: Don Collins

MLFC Program Timeline

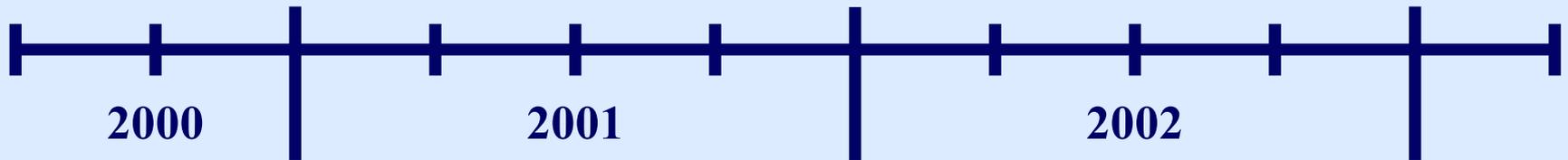
Manufacturing Analyses (Phase I)

Process Development (Phase II)

- * LSM Powder Production Process
- * Optimized Process for YSZ Coating Suspensions
 - * Fabrication Process for Cathode Supported Cells
 - * Fabrication Process for Anode Supported Cells

Testing (Phase III)

- Demonstrated High Performance *
- Scale-Up of Cell Fabrication *
- Samples for Property Testing at ORNL *
- Screen Printing Process for Cathode Coatings *
- Initiated Long-Term Cell Testing at GTI *



Program Plan

Phase I (3 months)

**Manufacturing Cost
and Risk Assessment**

**Michael A. Cobb & Co.
Advanced Materials Technologies
Gas Technology Institute**

Phase II (12 months)

**Development of Fabrication
Processes for Planar Cells**

**NexTech Materials
Oak Ridge National Laboratory
University of Missouri-Rolla**

Phase III (18 months)

**SOFC Testing, Destructive
and Non-Destructive Testing**

**Northwestern University
Gas Technology Institute
Ohio State University**

NEXTECH

MATERIALS

Process Development

NexTech

**Tape Casting
(Anode or Cathode)**

**Colloidal Spray
(Electrolyte)**

Co-Sintering

**Screen Printing
(Cathode or Anode)**

ORNL

**Tape Casting
(Anode)**

**Screen Printing
(Electrolyte)**

Co-Sintering

**Screen Printing
(Cathode)**

UMR

**Tape Casting
(Cathode)**

Sintering

**Spin Coating
(Electrolyte)**

**Screen Printing
(Anode)**

Phase I Lessons

□ Keys to Cost Reduction

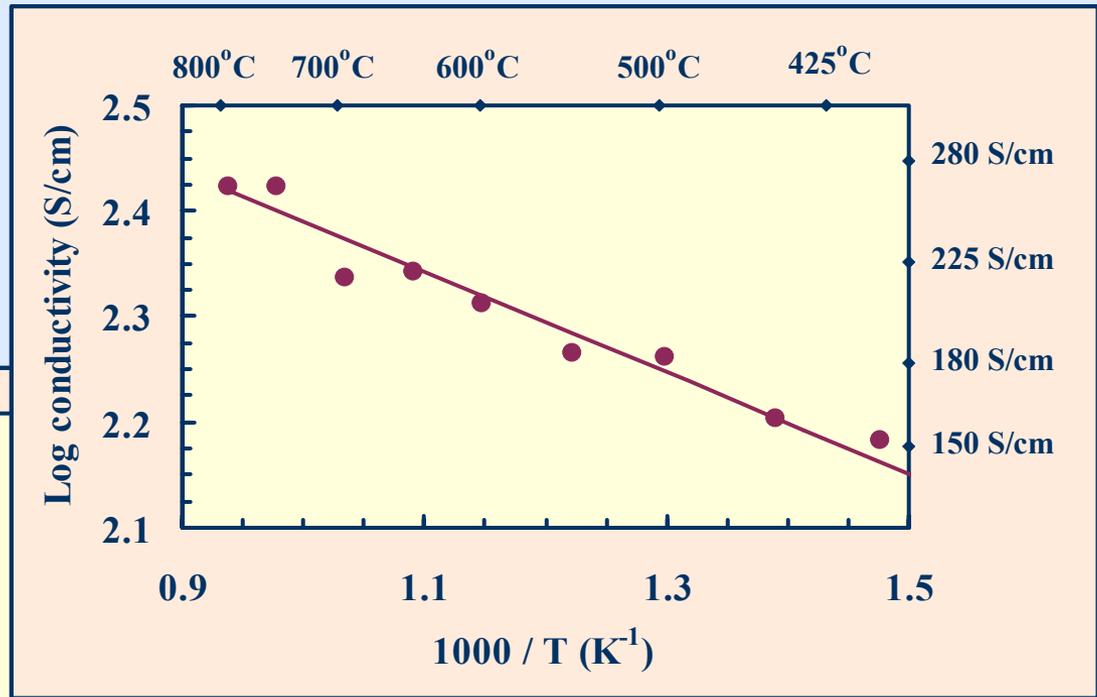
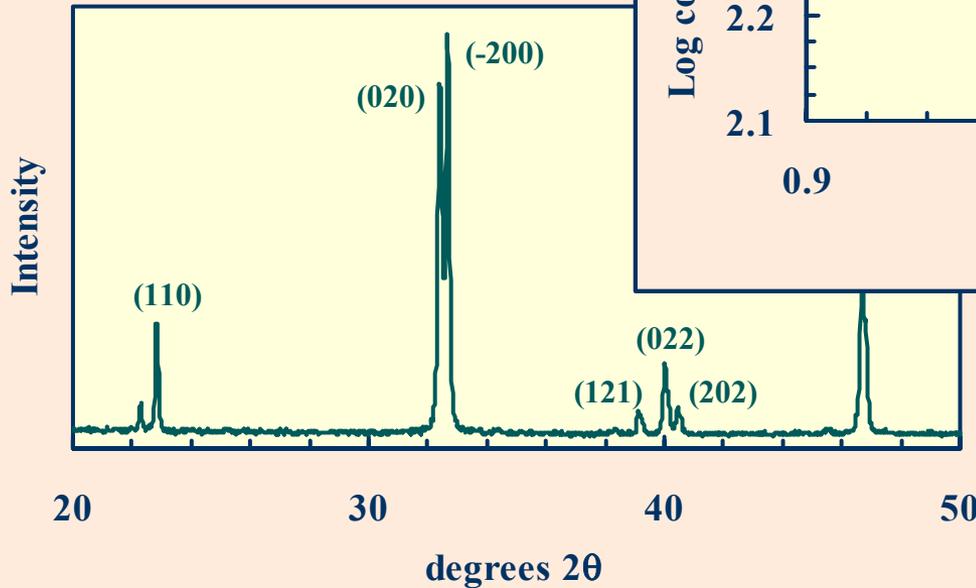
- Lower operating temperatures (interconnects)
- Increased power densities
- Low-cost raw materials (especially YSZ)
- Increased manufacturing yields
- Efficient materials utilization (advanced designs)

□ Development Risk Elements

- Seals
- Thermal expansion mismatches
- Gas flow through porous electrodes
- Scale up to large electrolyte areas

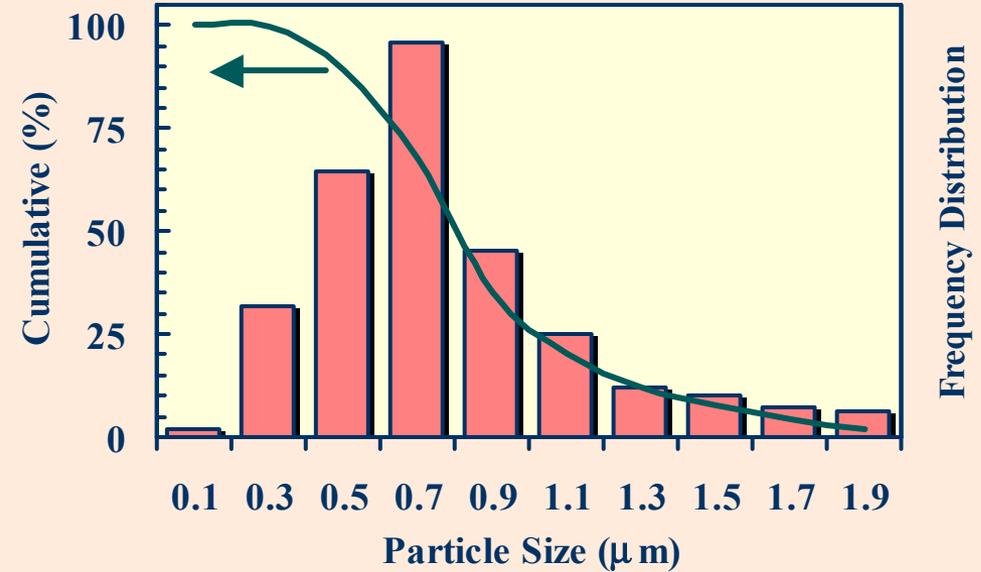
LSM Powder Production

Composition

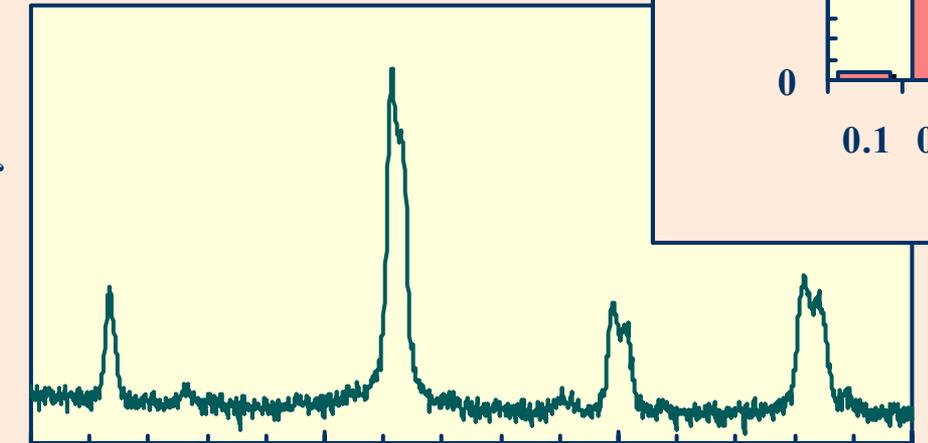


LSF Powder Production

Composition



Intensity



20

30

40

50

degrees 2θ

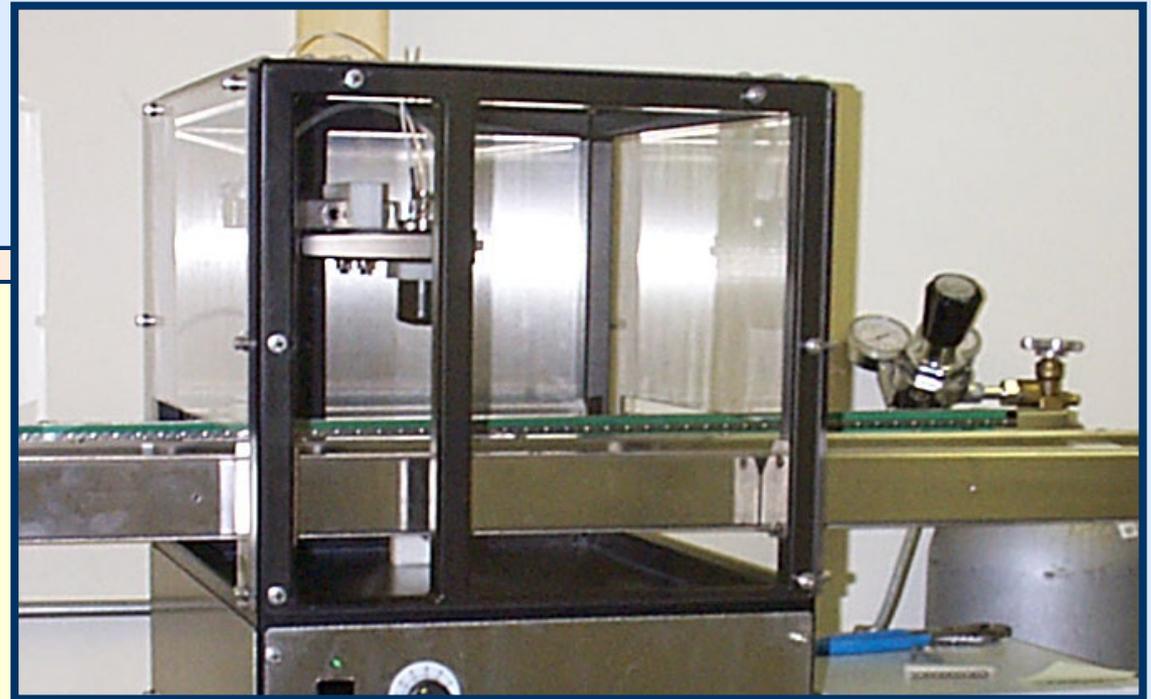
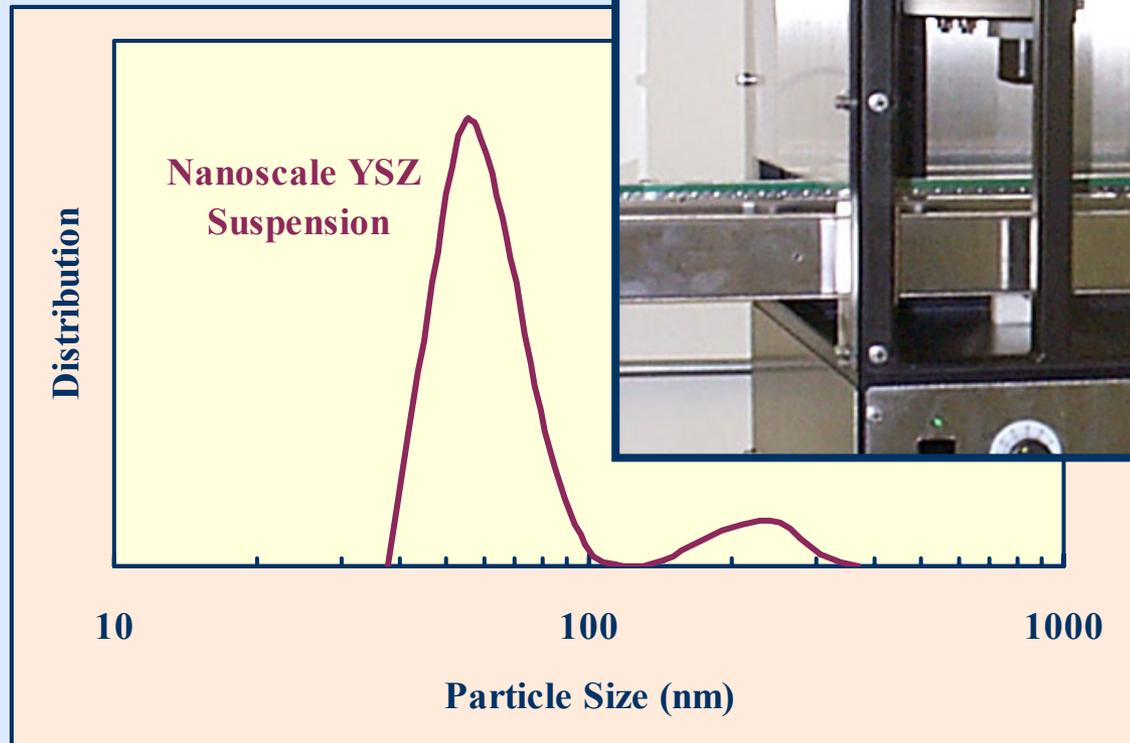
Tape Casting

Process Variables

- Particle Size and Surface Area
- Formulation (binders, fugitives, etc.)
- Cleanliness (dust, hair, etc.)
- Tape Casting Layer Thickness
- Lamination Conditions
- Dicing Methods
- Binder Burnout Cycle
- Pre-Calcination Conditions



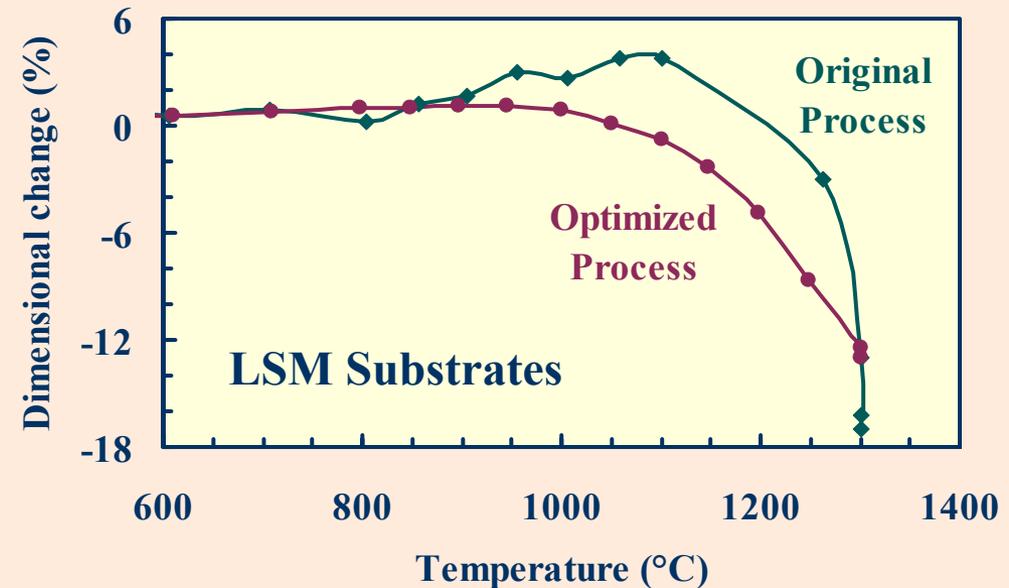
YSZ Coating Process



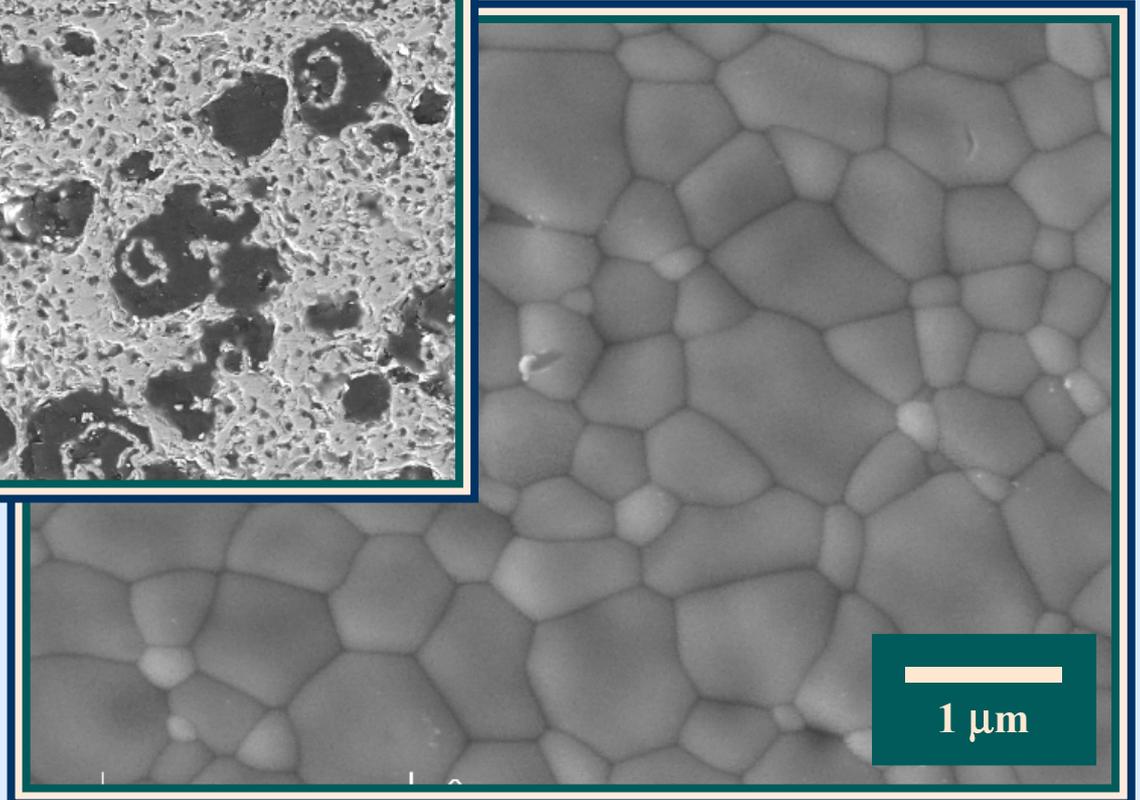
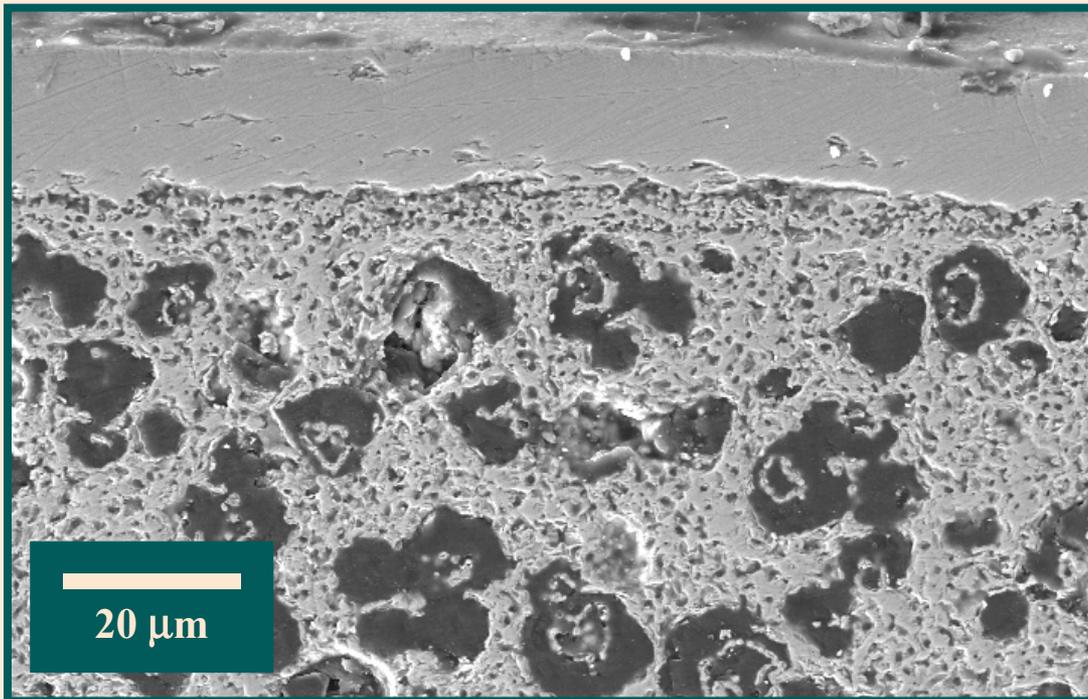
Co-Sintering

Process Variables

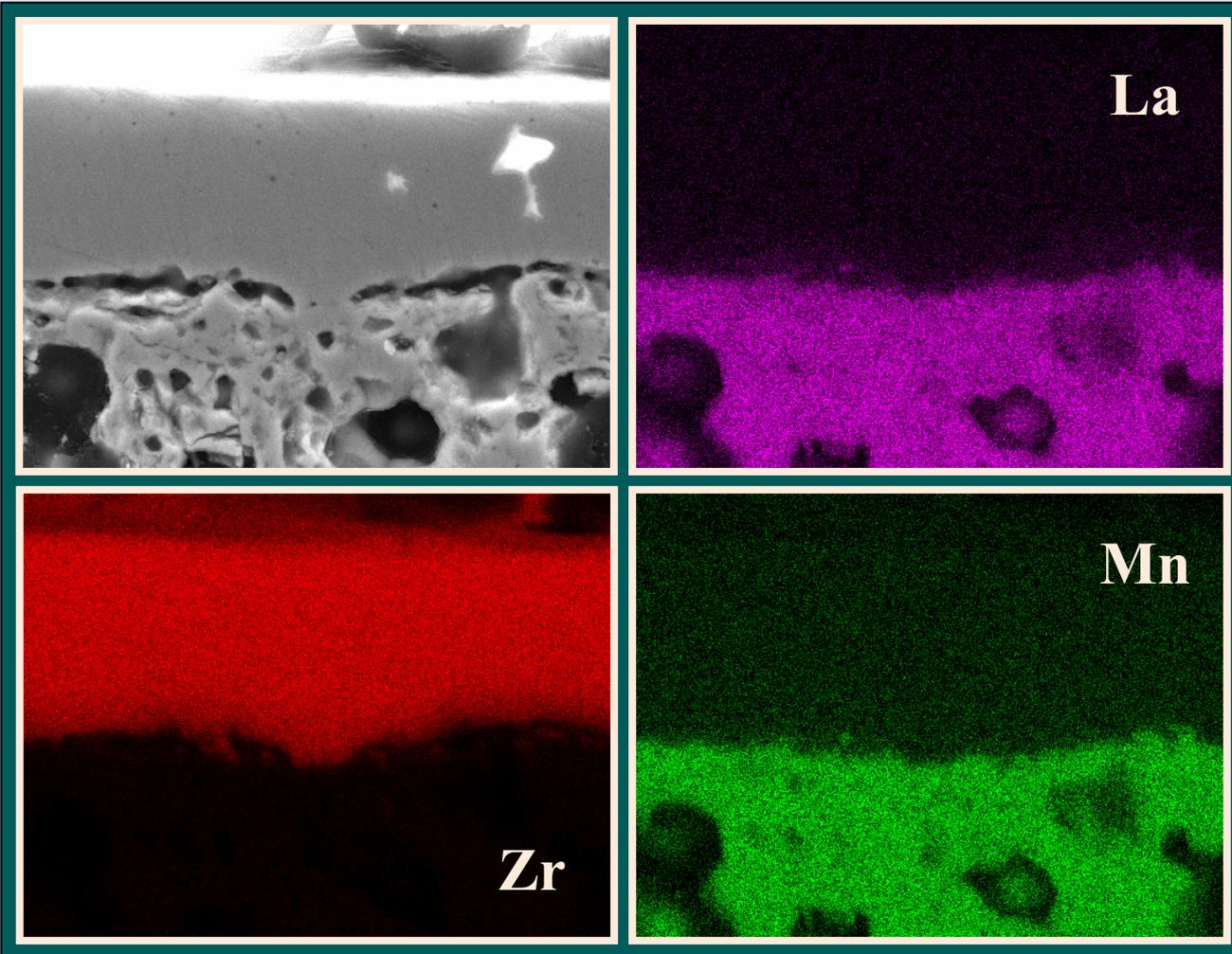
- Tape Formulations
- Coating Process
- Pre-Calcination
- Setter Tiles
- Control of Component Shrinkages
- Heating and Cooling Rates
- Constrained Sintering
- Creep Flattening



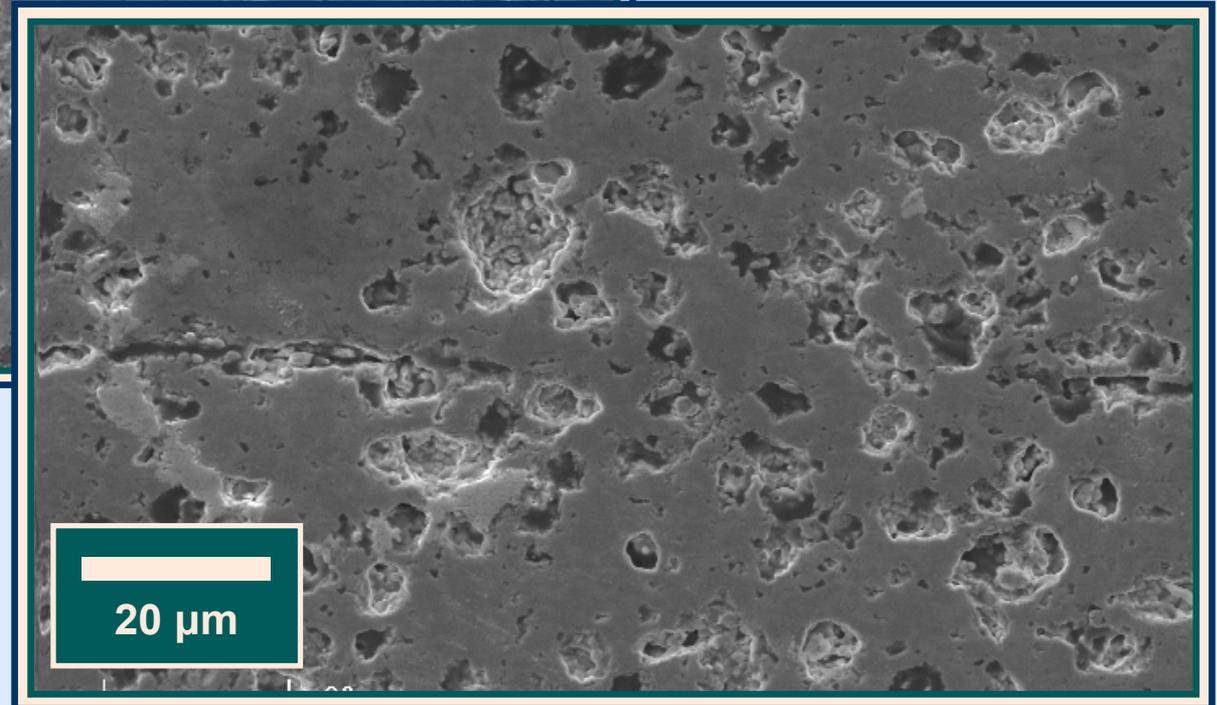
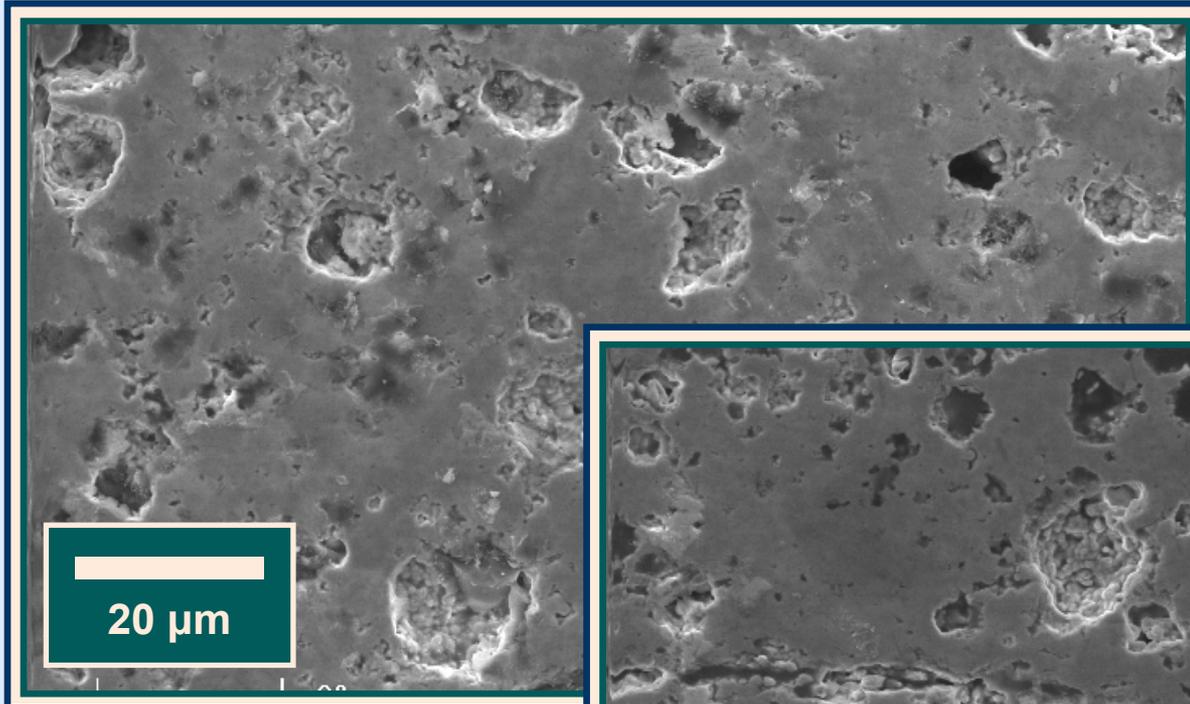
Cathode Supported Cells



Cathode Supported Cells

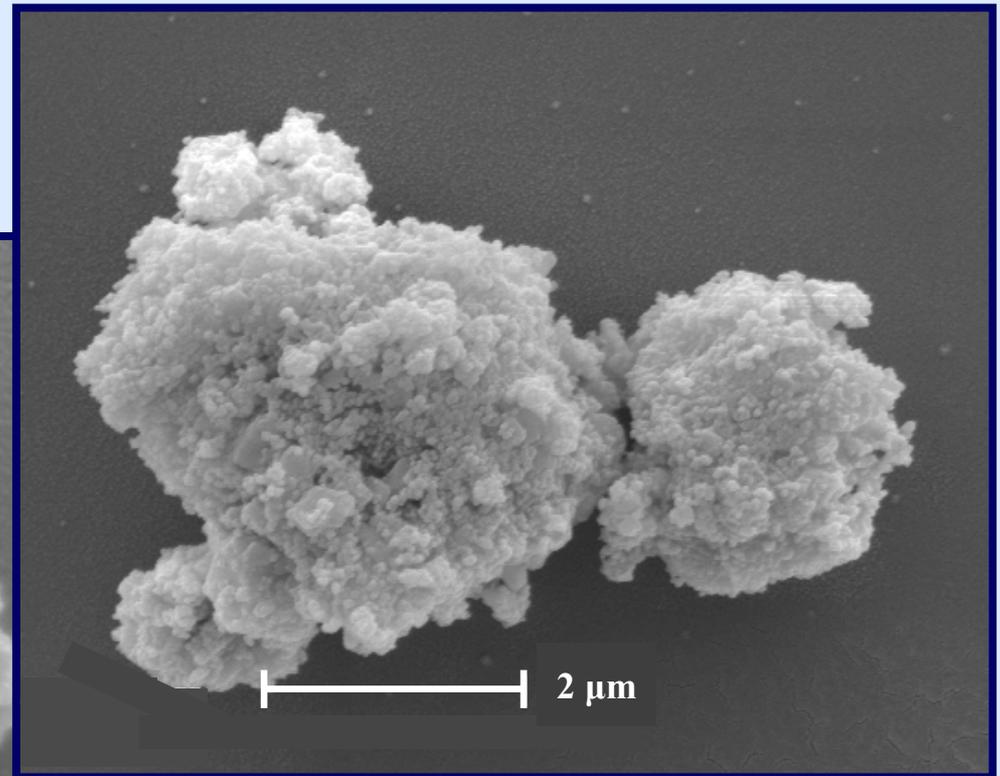
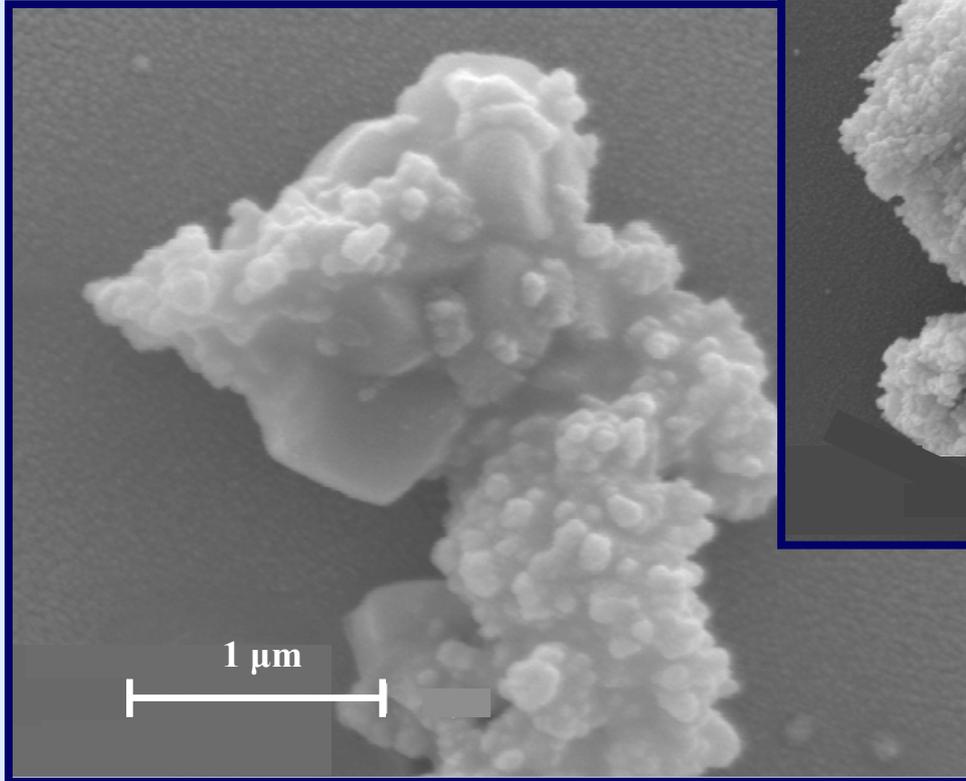


Anode Microstructure



Nano-Composite Anodes

Conventional Method



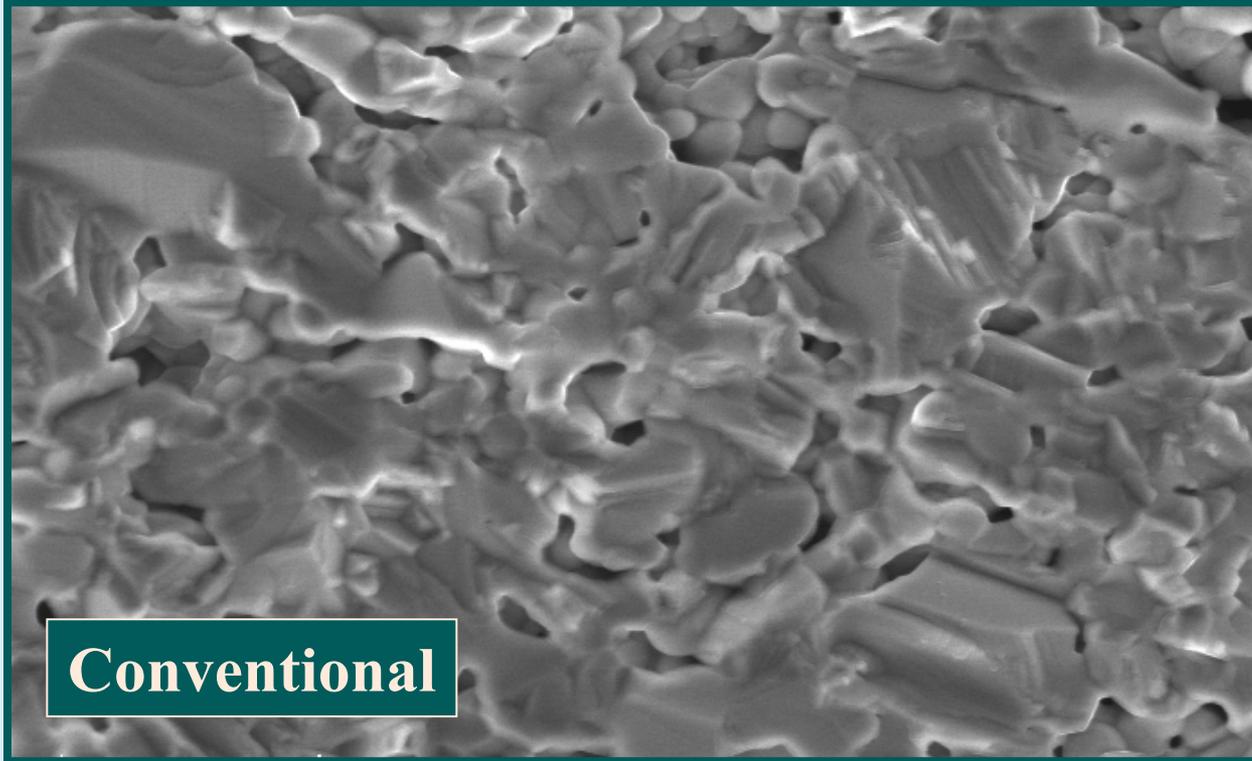
New Method

U.S. Patent Pending

NEXTECH

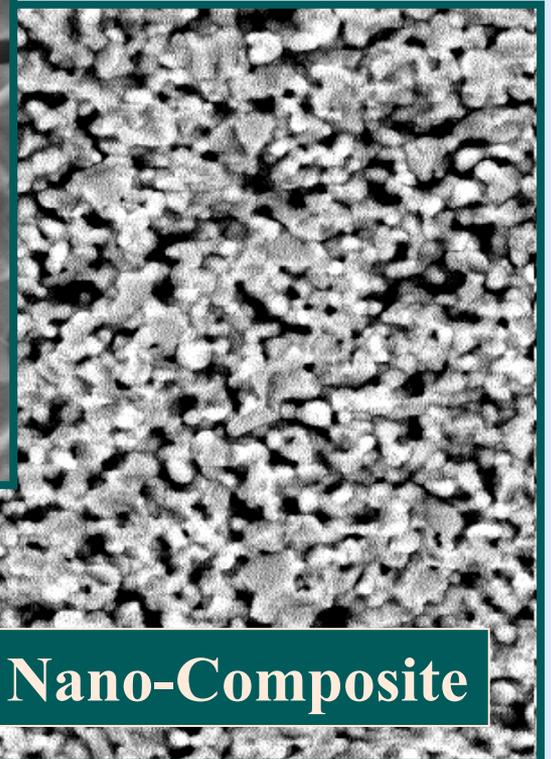
MATERIALS

Nano-Composite Anodes



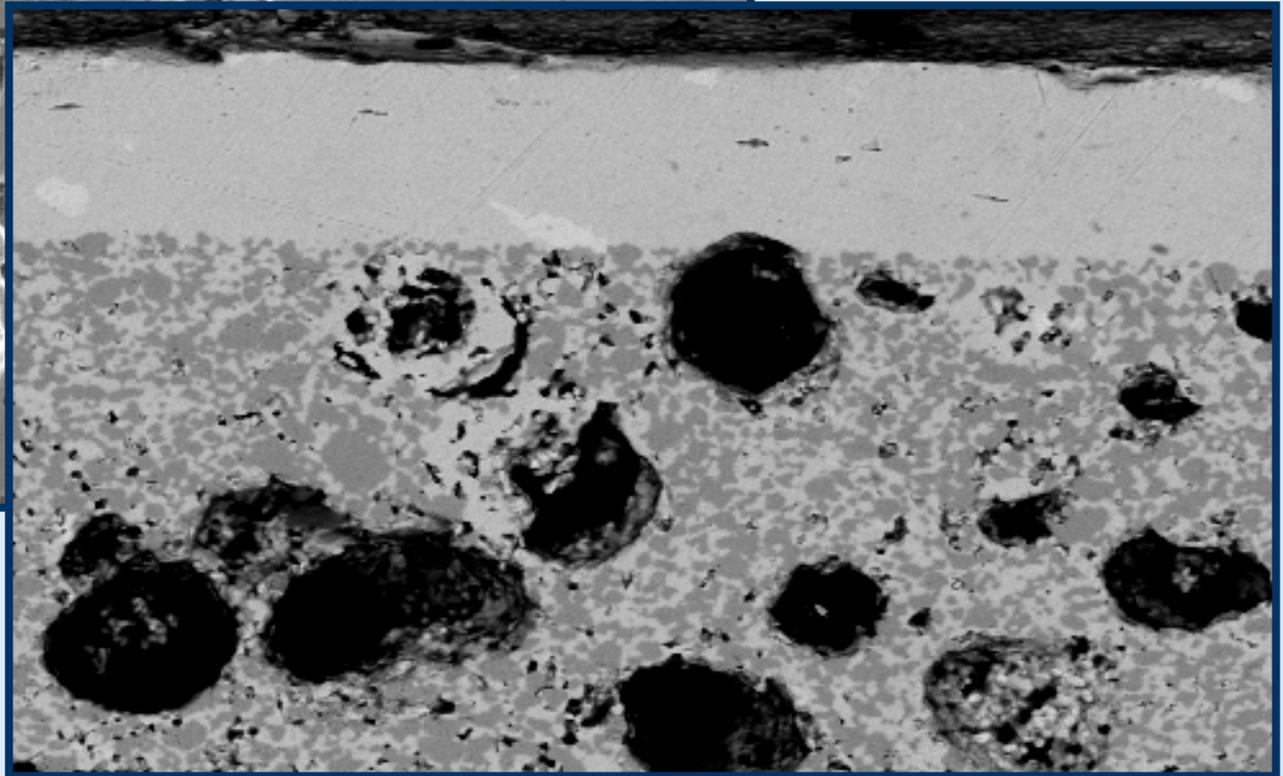
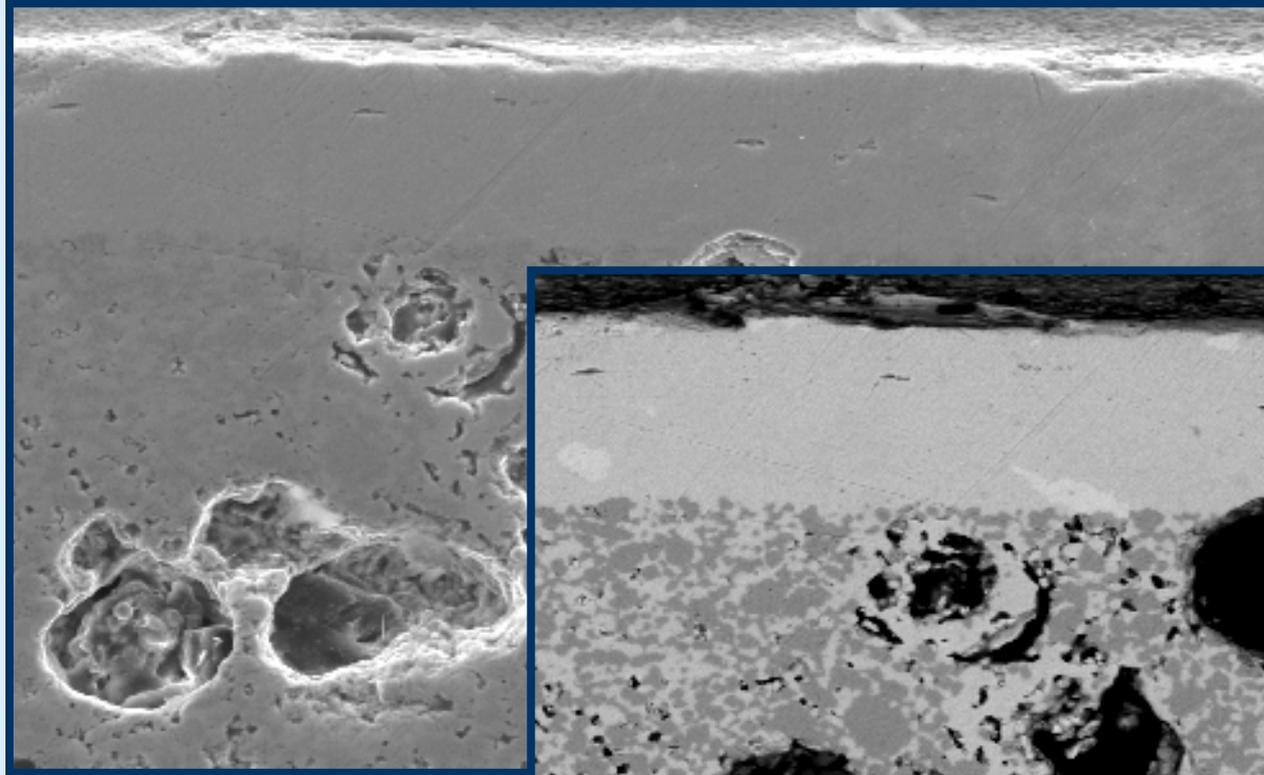
Conventional

5 μm



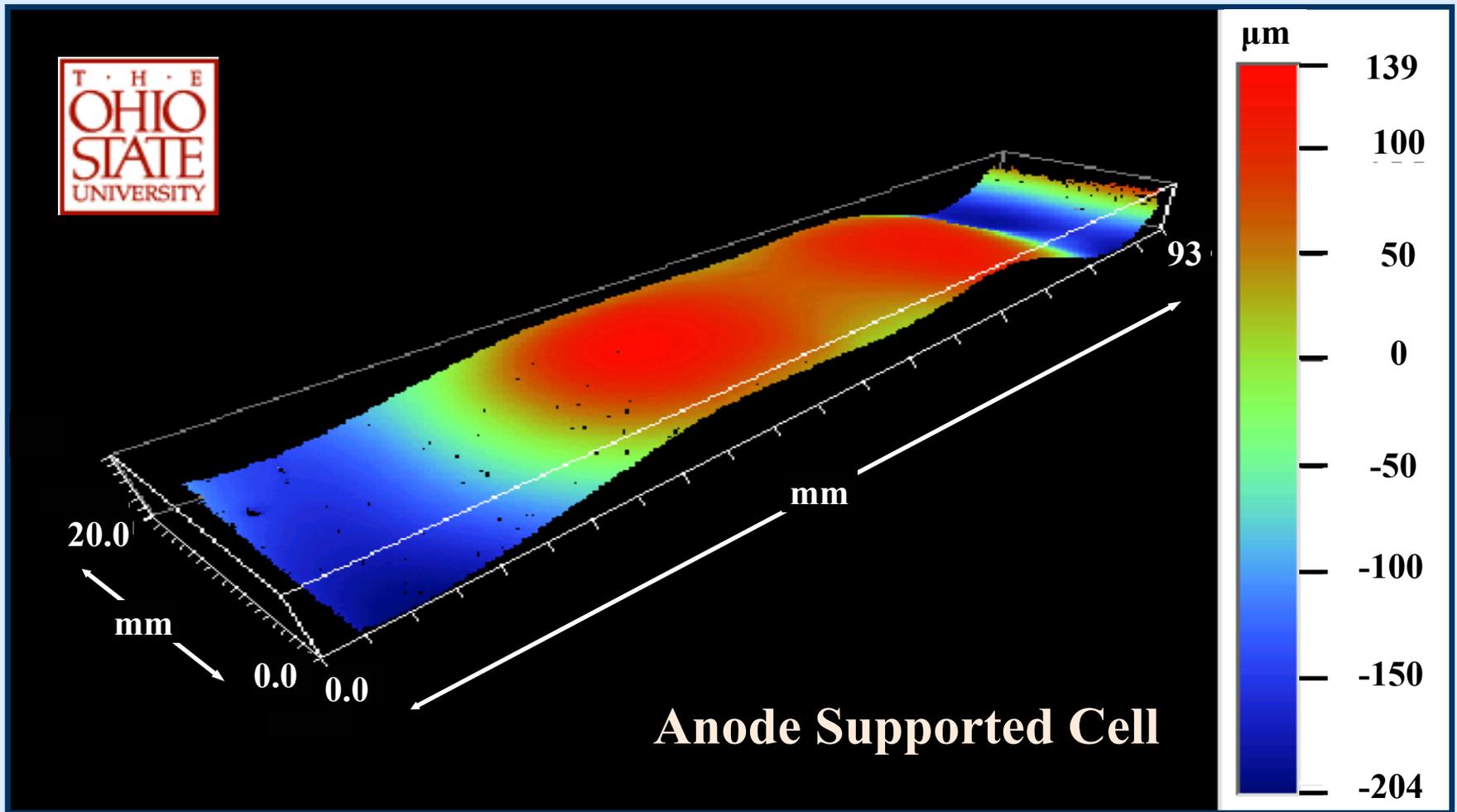
Nano-Composite

Anode-Supported Cells

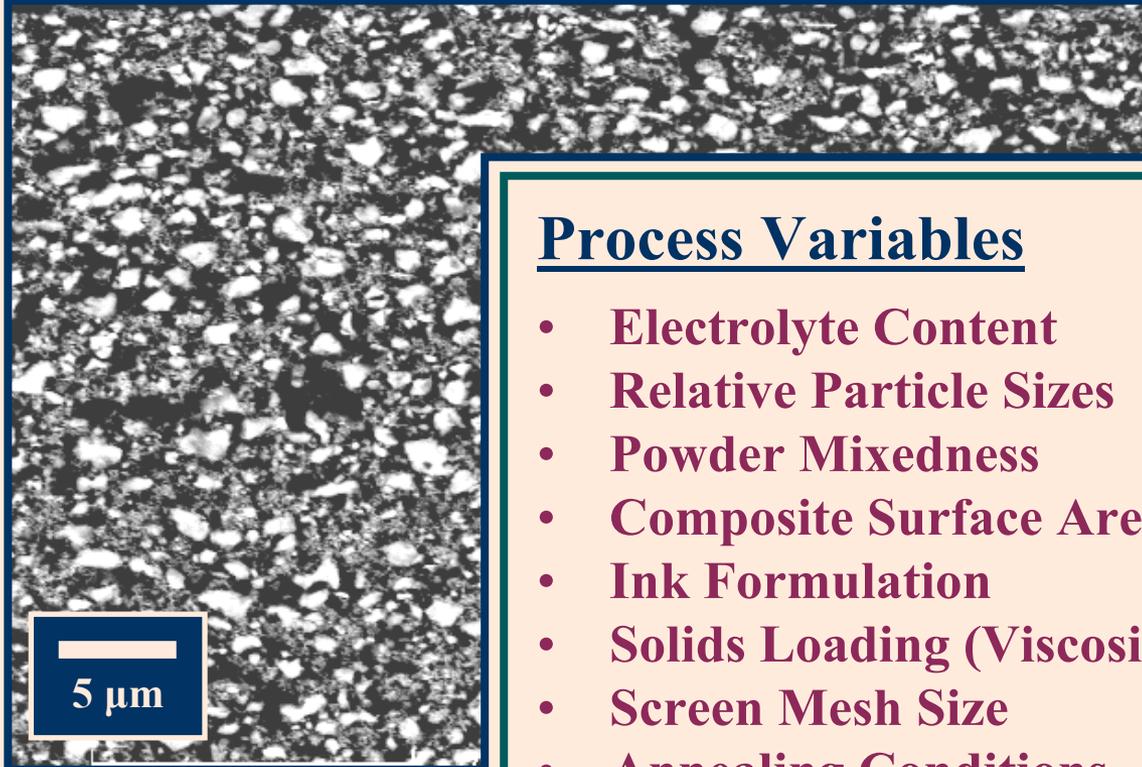


10 μm

Flatness Measurements

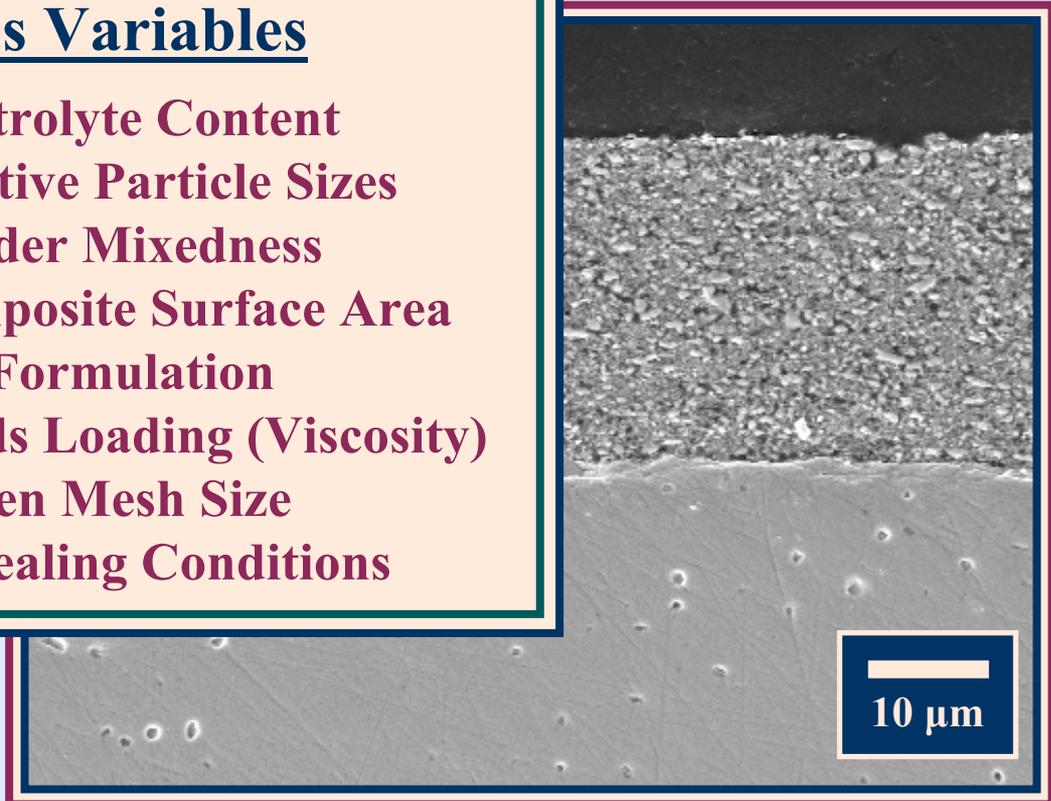


Cathode Screen Printing

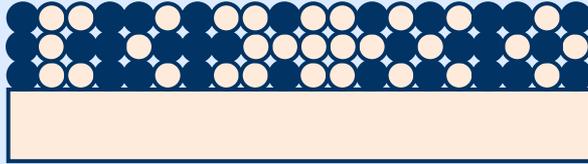


Process Variables

- Electrolyte Content
- Relative Particle Sizes
- Powder Mixedness
- Composite Surface Area
- Ink Formulation
- Solids Loading (Viscosity)
- Screen Mesh Size
- Annealing Conditions

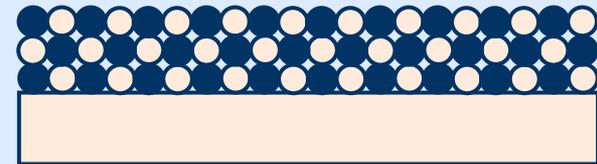
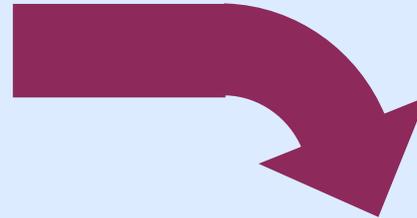


Composite Cathodes



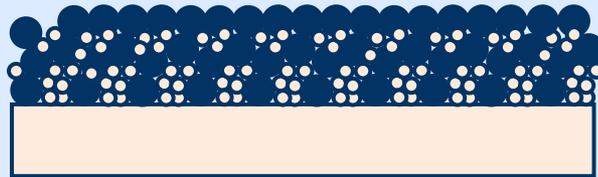
Conventional Ink Routes

- **Improved Low-Temperature Performance**
- **Inhomogeneous Microstructure**
- **Limited Increase In TPB Area**



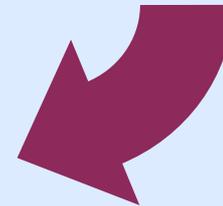
Improved Homogeneity

- **Greater TPB Area**
- **More Uniform Shrinkage**
- **Low Connectivity Current Paths**

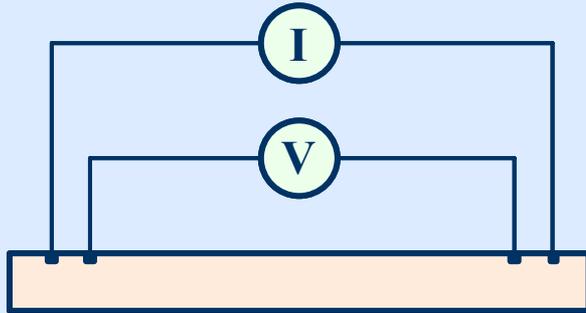


Nanoscale Composites

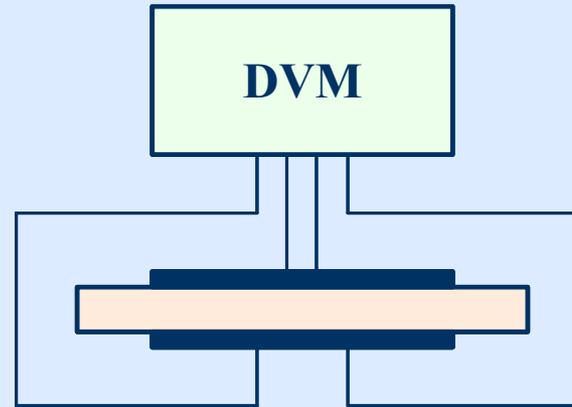
- **Maximized TPB Area**
- **Uniform Distribution of Electrolyte**
- **High Connectivity Current Paths**



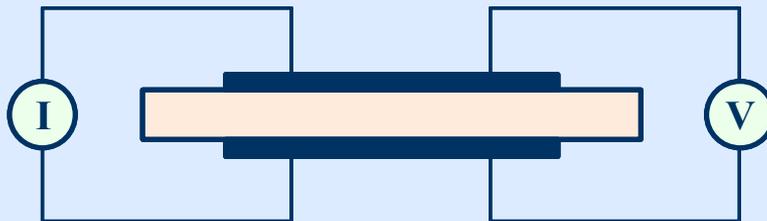
Electrical Testing



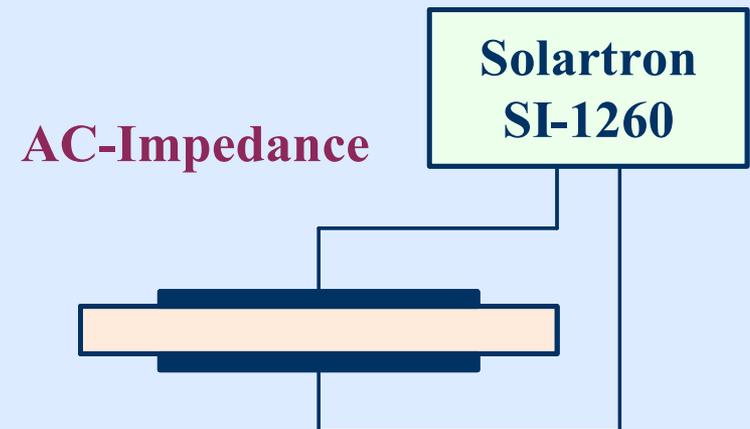
Four-Point Conductivity



Four-Point Resistance

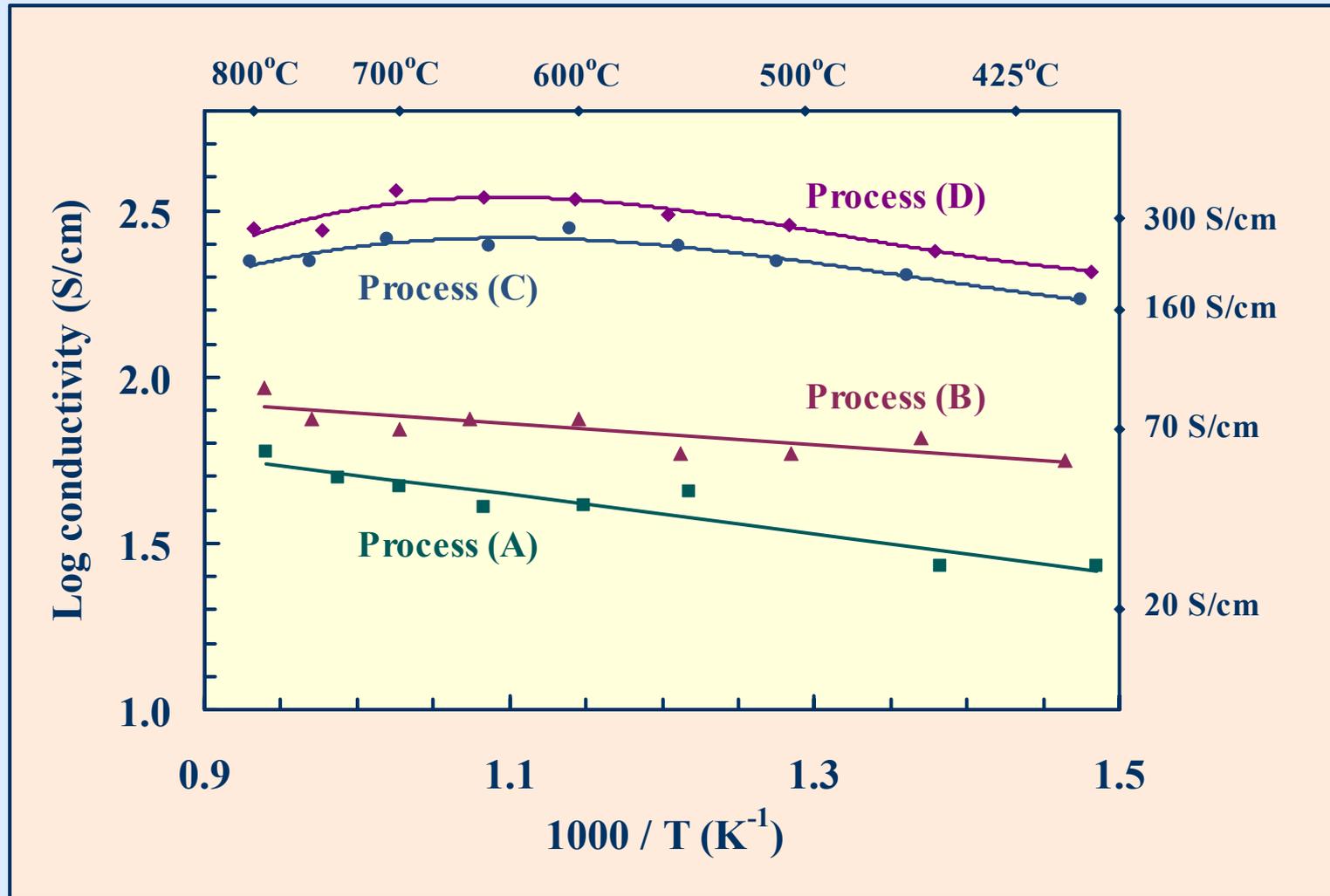


I-V Measurements

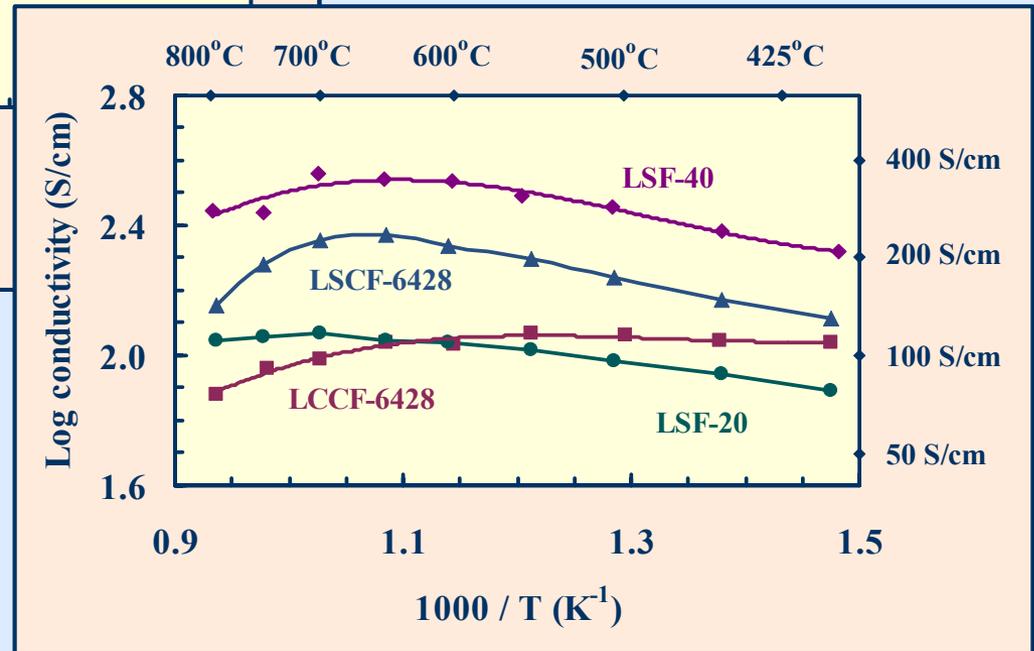
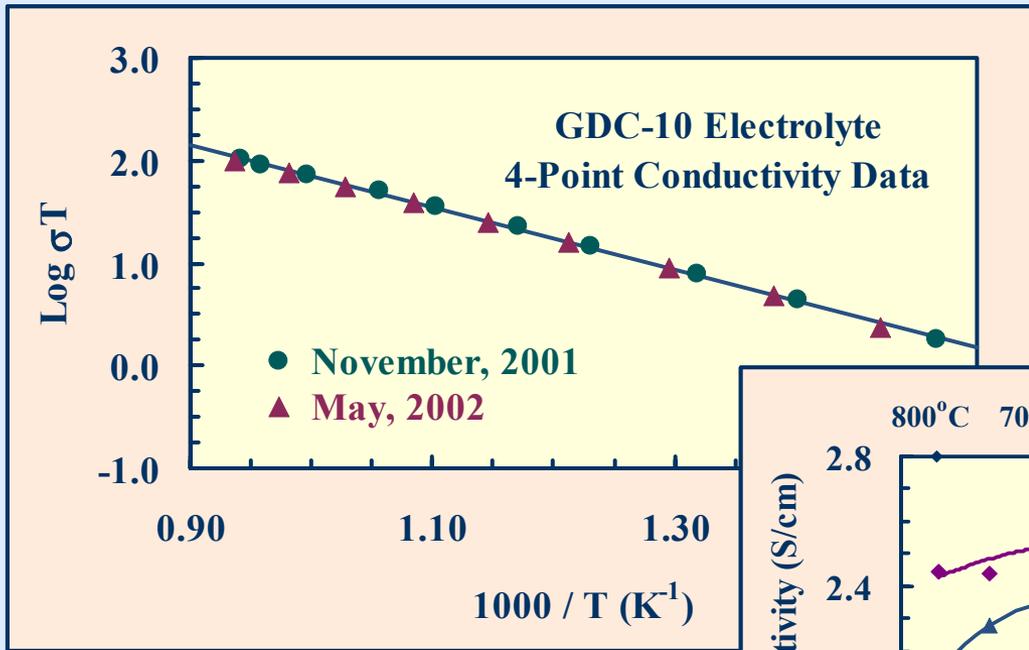


AC-Impedance

Effect of Processing (LSF-40)



Electrical Testing



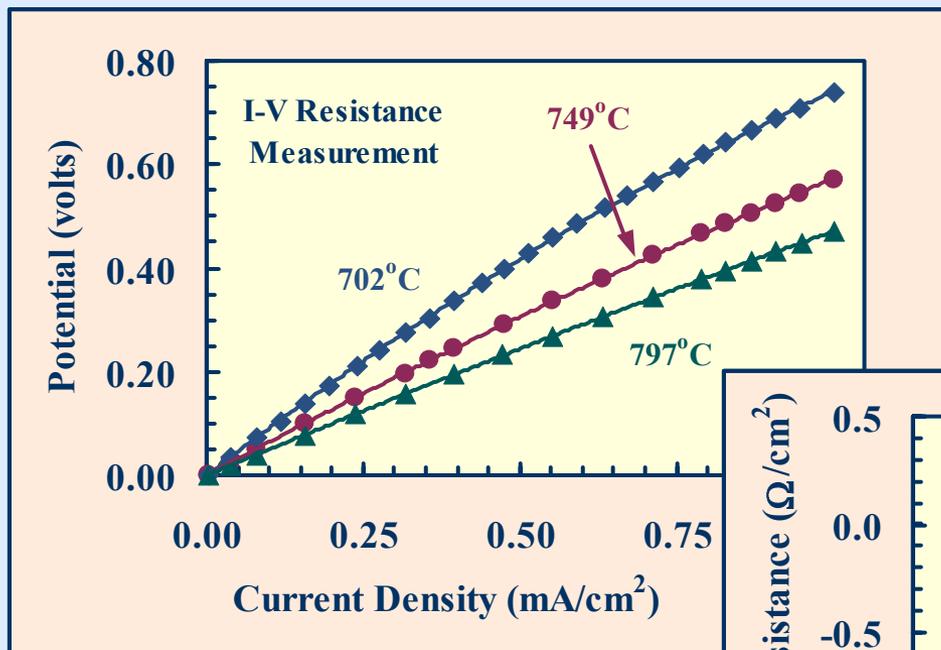
LSF-20: $(\text{La}_{0.80}\text{Sr}_{0.20})\text{FeO}_3$

LSF-40: $(\text{La}_{0.60}\text{Sr}_{0.40})\text{FeO}_3$

LSCF-6428: $(\text{La}_{0.60}\text{Sr}_{0.40})(\text{Co}_{0.20}\text{Fe}_{0.80})\text{O}_3$

LCCF-6428: $(\text{La}_{0.60}\text{Ca}_{0.40})(\text{Co}_{0.20}\text{Fe}_{0.80})\text{O}_3$

Electrode Resistance



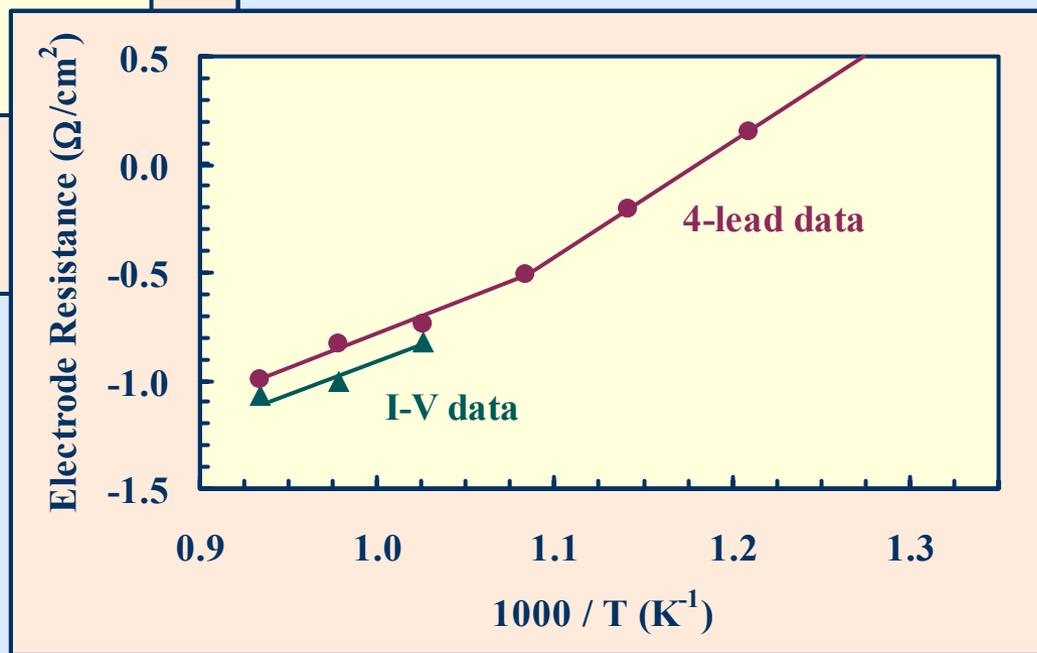
GDC Electrolyte Disc

300 μm thick

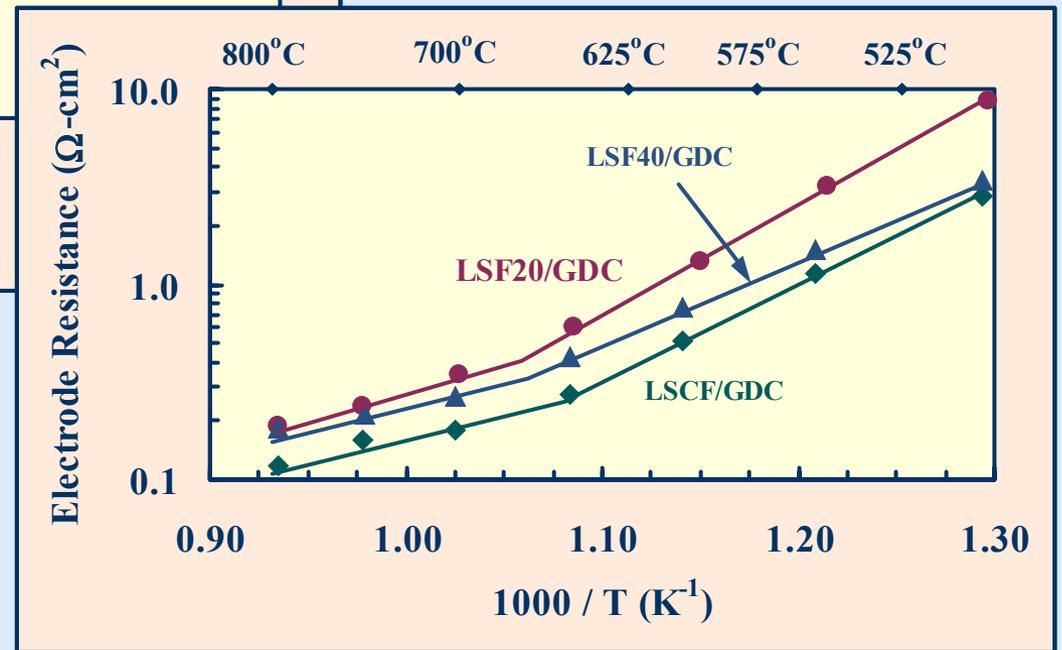
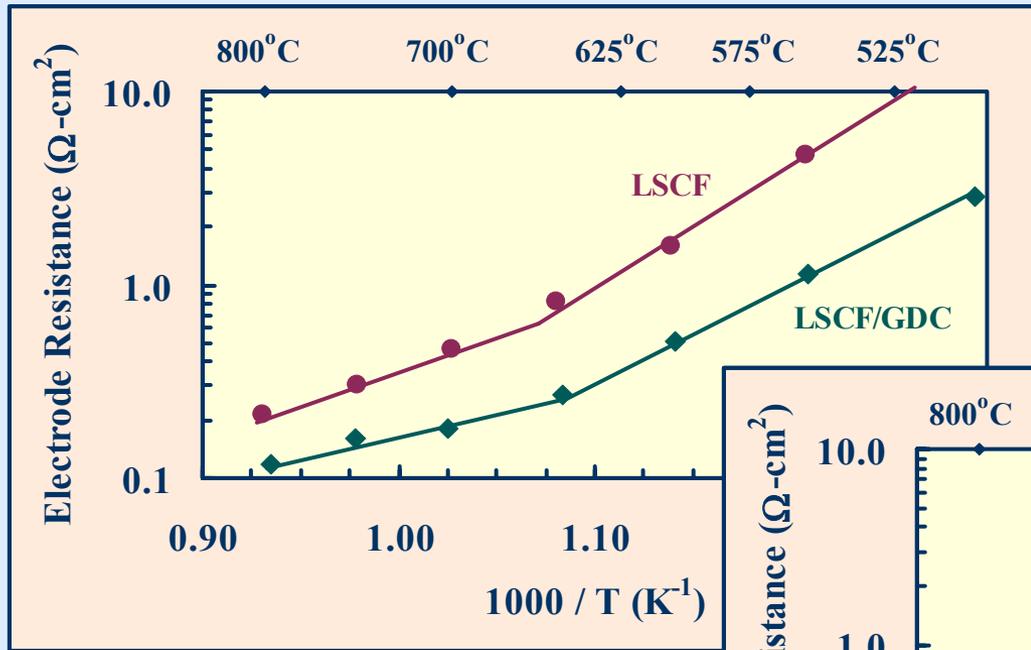
Screen-Printed Electrodes

LSCF/GDC Composition

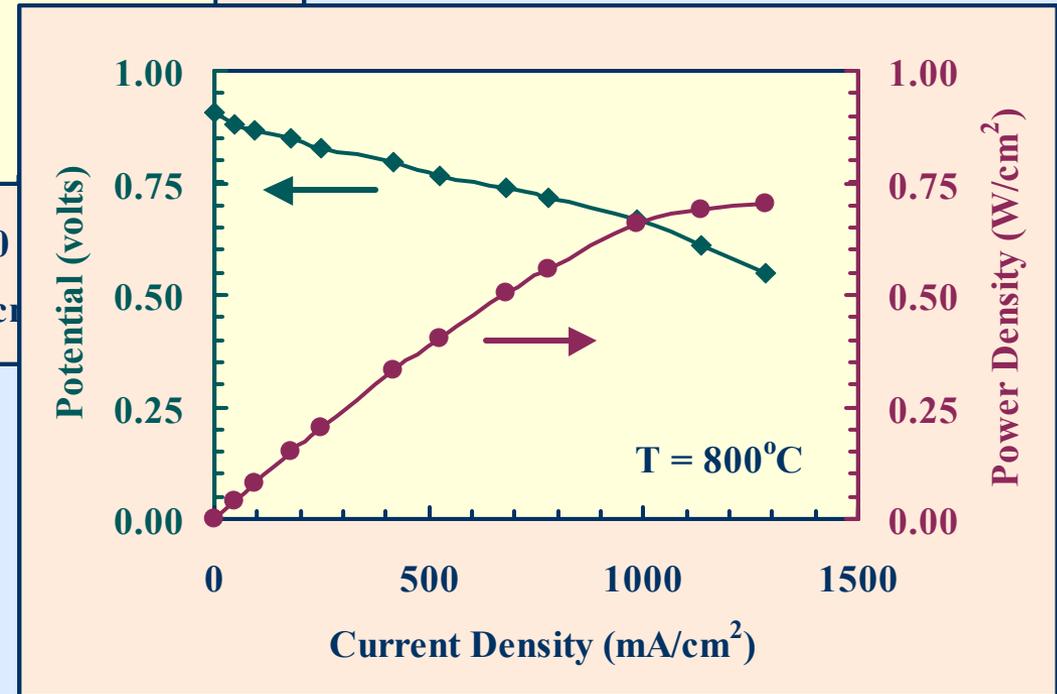
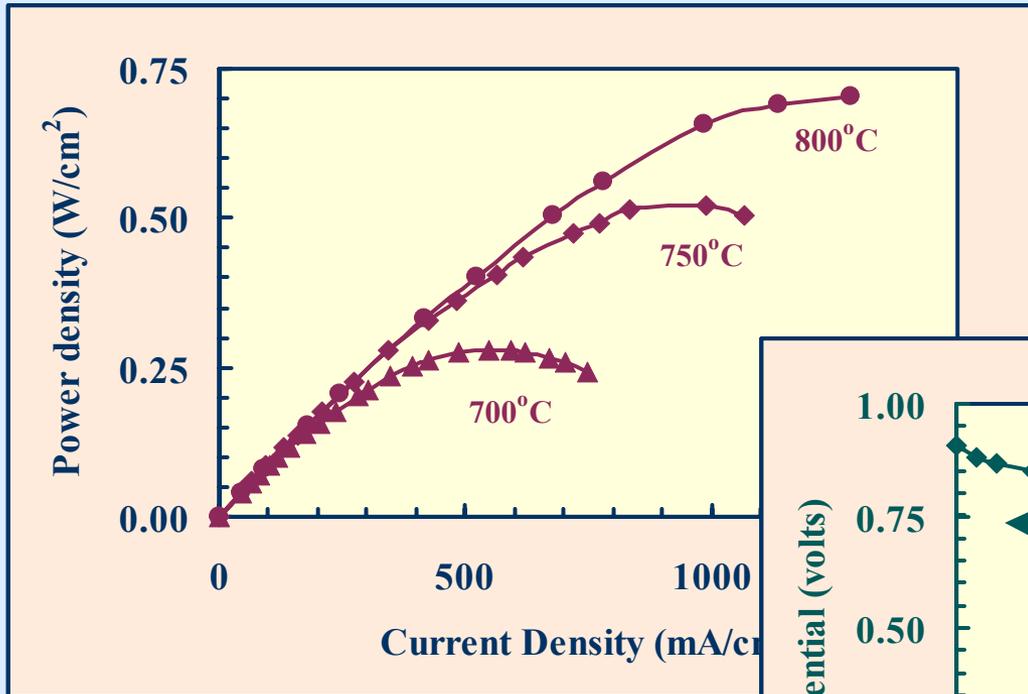
1.27 cm^2 area, ~ 50 μm thick



Electrical Testing



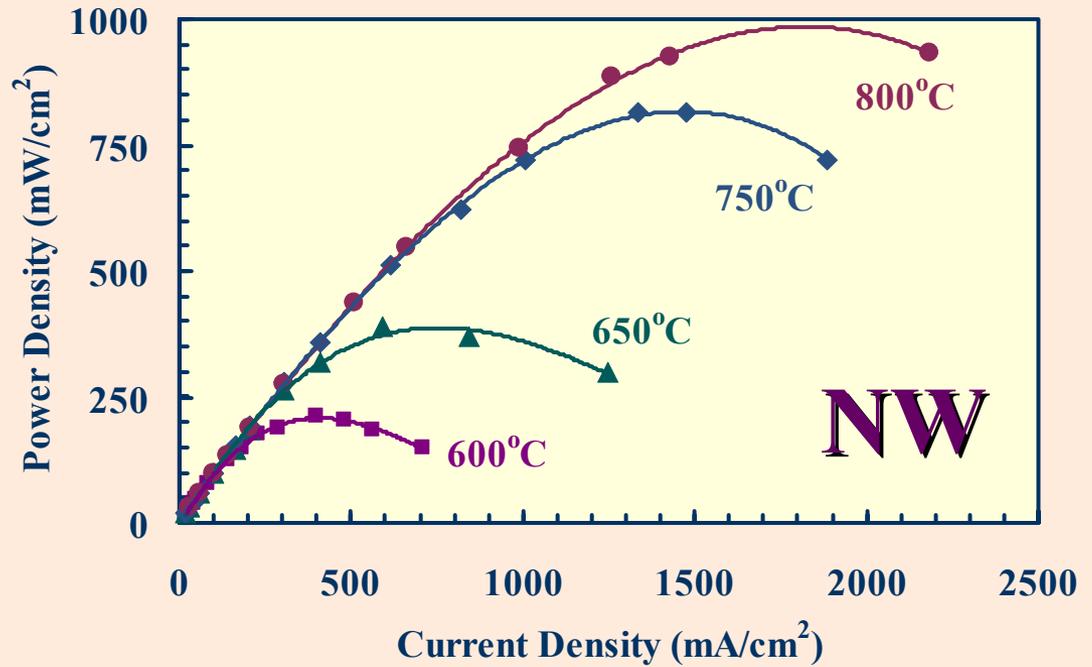
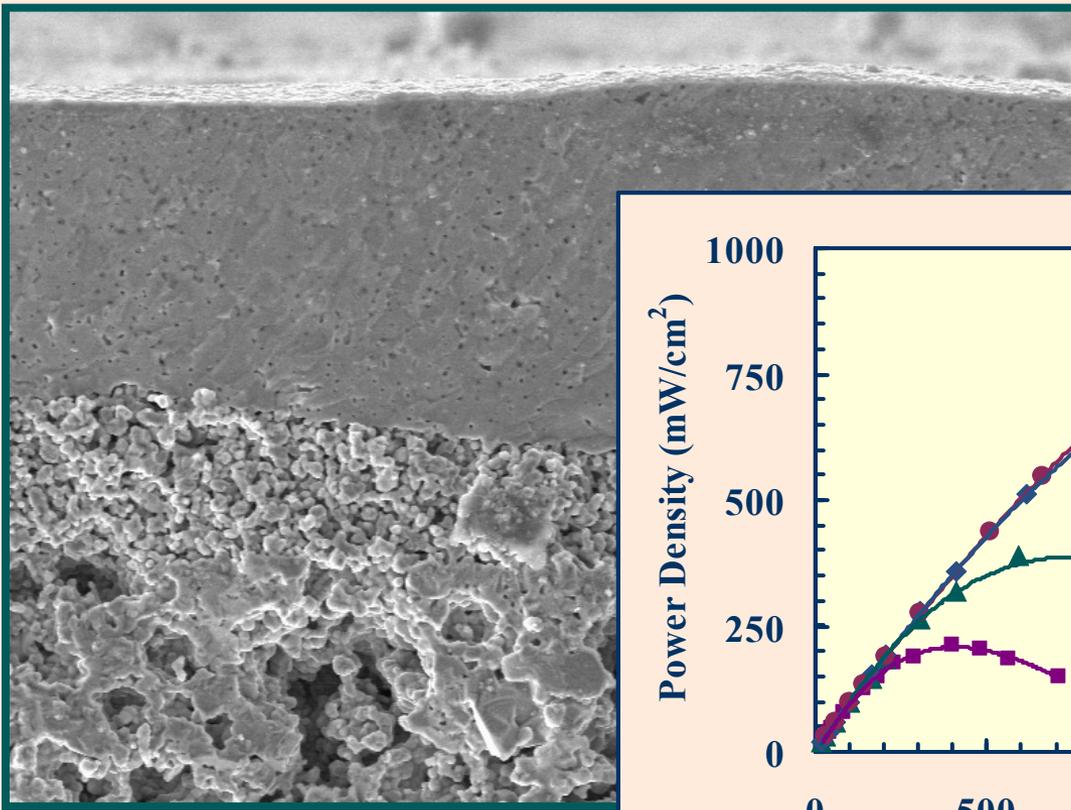
SOFC Testing



NexTech
Anode-Supported Cell
LSCF/GDC Cathode

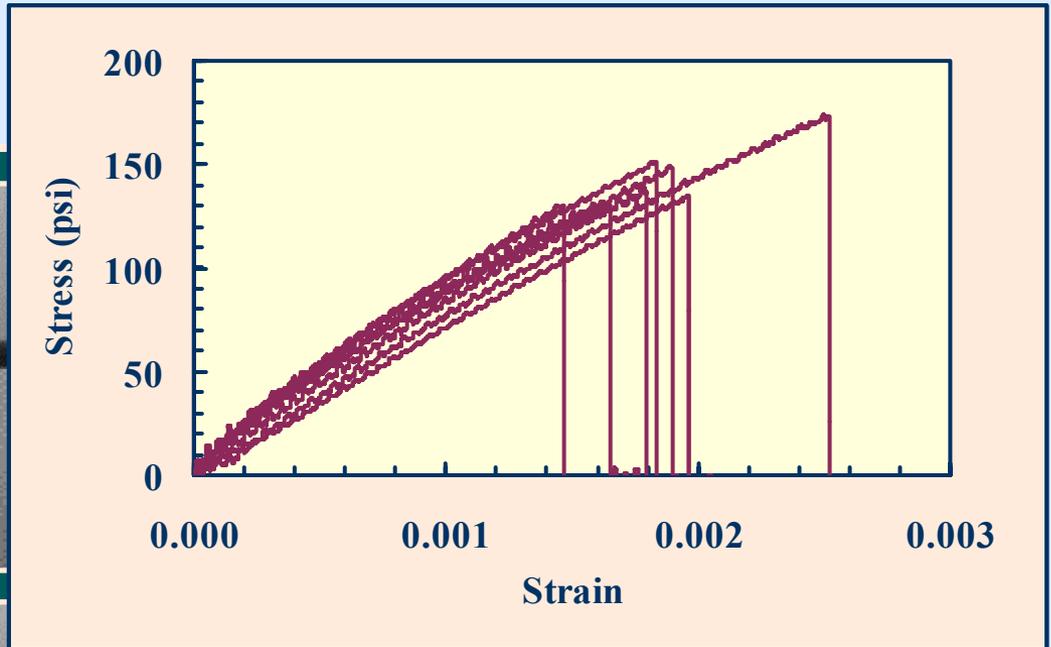
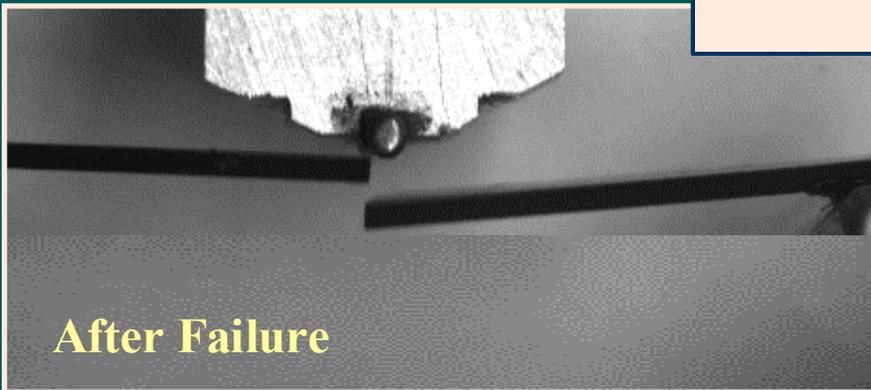
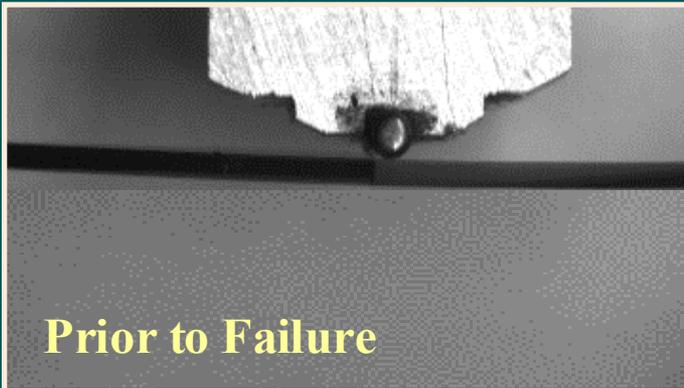
ornl

SOFC Testing



Mechanical Properties

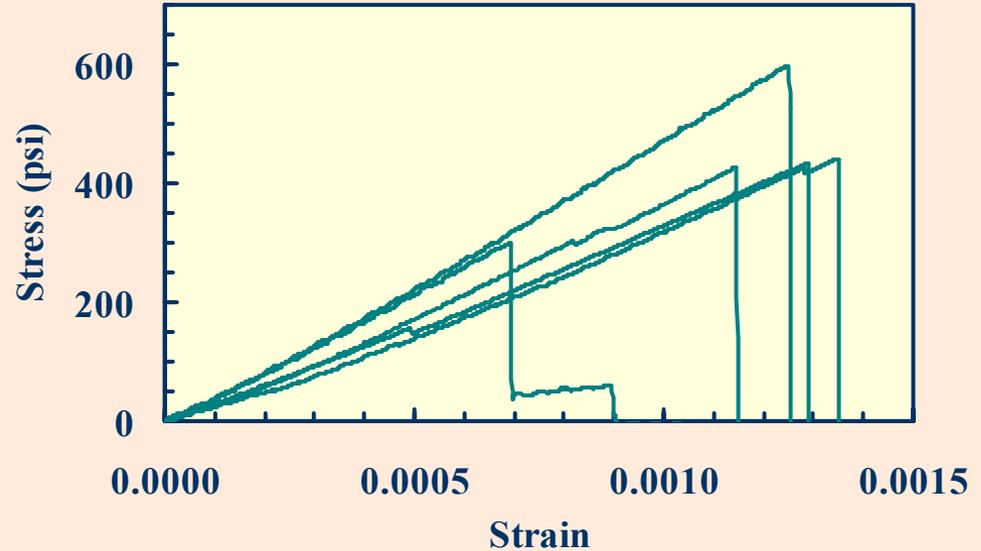
LSM Cathode Substrates



Flexural Modulus: 83,546 psi
Flexural Failure Stress: 142.5 psi
Flexural Failure Strain: 0.00185

Mechanical Properties

NiO/YSZ Anode Substrates



Flexural Modulus: 364,000 psi
Flexural Failure Stress: 440.2 psi
Flexural Failure Strain: 0.00115

Ceria-Based Electrolytes

DOE Contract No. DE-AC26-00NT40706

Principal Investigator: Matt Seabaugh

NETL Project Manager: Lane Wilson

Objectives:

- Improve performance of ceria-based electrolytes
- Fabricate anode-supported, thin-film ceria SOFCs
- Achieve high power density at $T < 700^{\circ}\text{C}$

Accomplishments

- Improved processes for ceria electrolytes
- Higher-conductivity ceria formulations
- Lower-cost ceria formulations
- Co-sintering process for anode-supported cells
- Screen-printed composite cathode coatings

Technical Approach

Processing Strategies

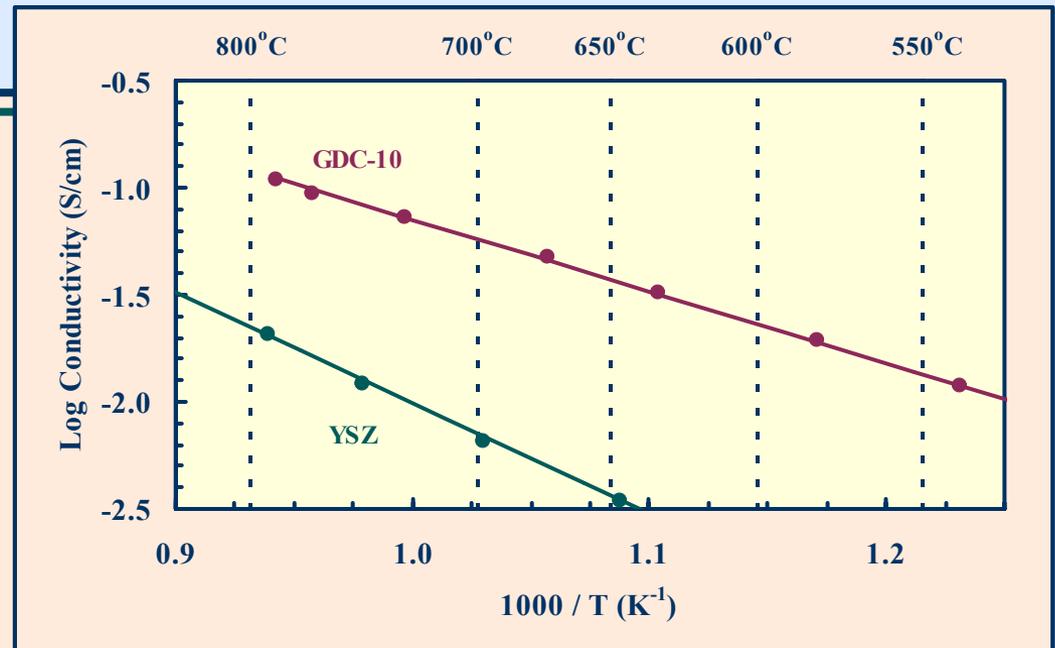
- Eliminate impurities
- Low-temperature sintering
- Maintain fine grain size
- Anode-supported cells

Compositional Strategies

- Multiply-doped ceria (👉)
- Use of inexpensive dopants
- Reduce lattice strain - increase conductivity

👉 Previously demonstrated by:

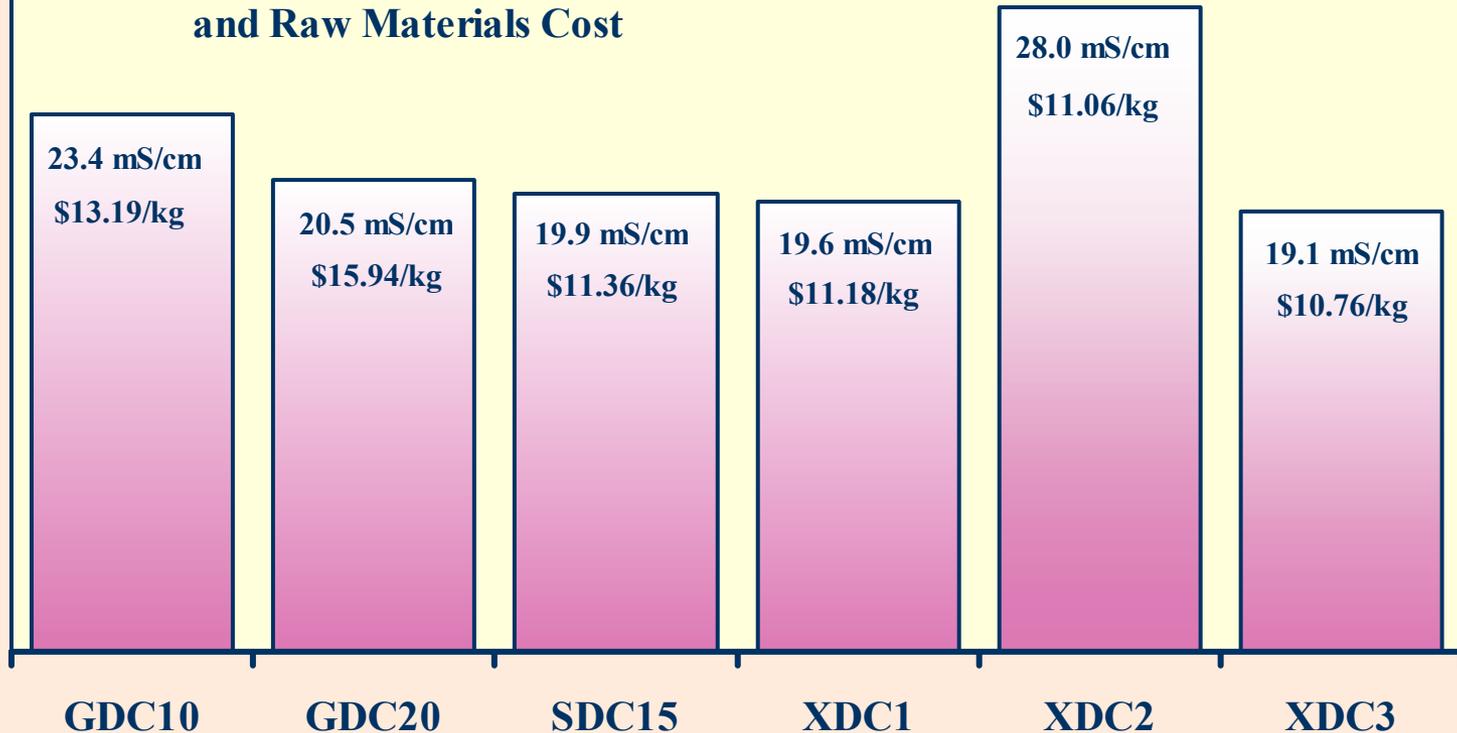
- van Herle, et al. (EPFL)
- Mori, et al. (NIRIM)



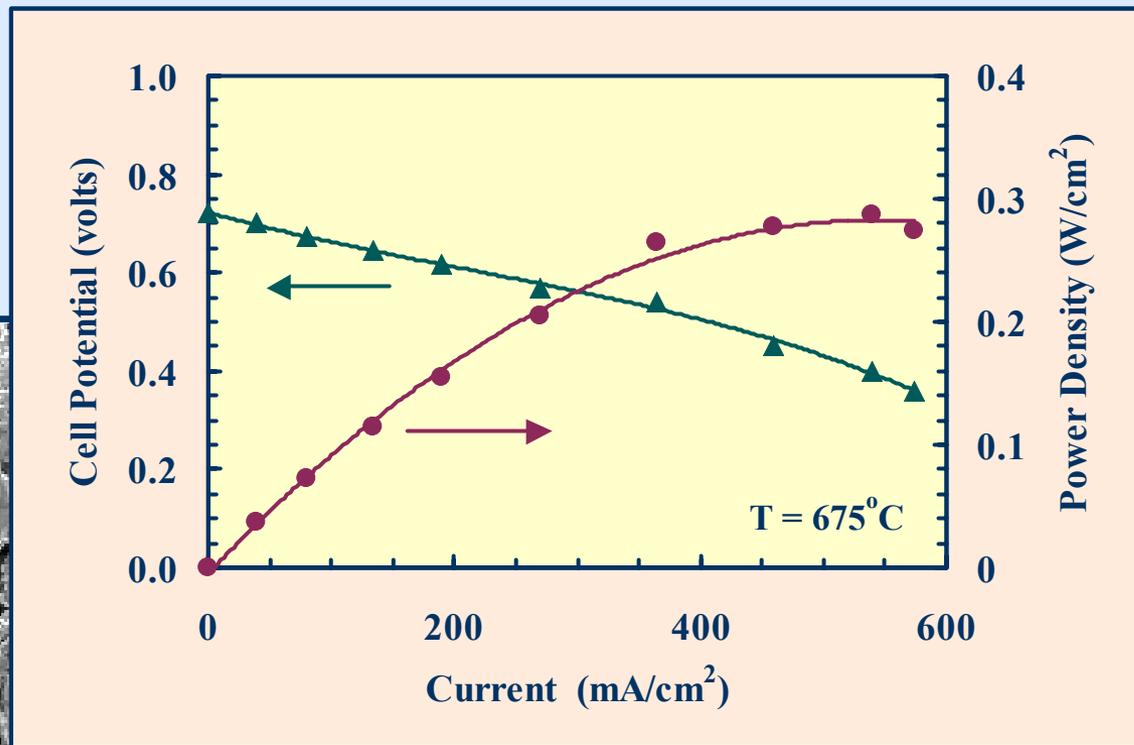
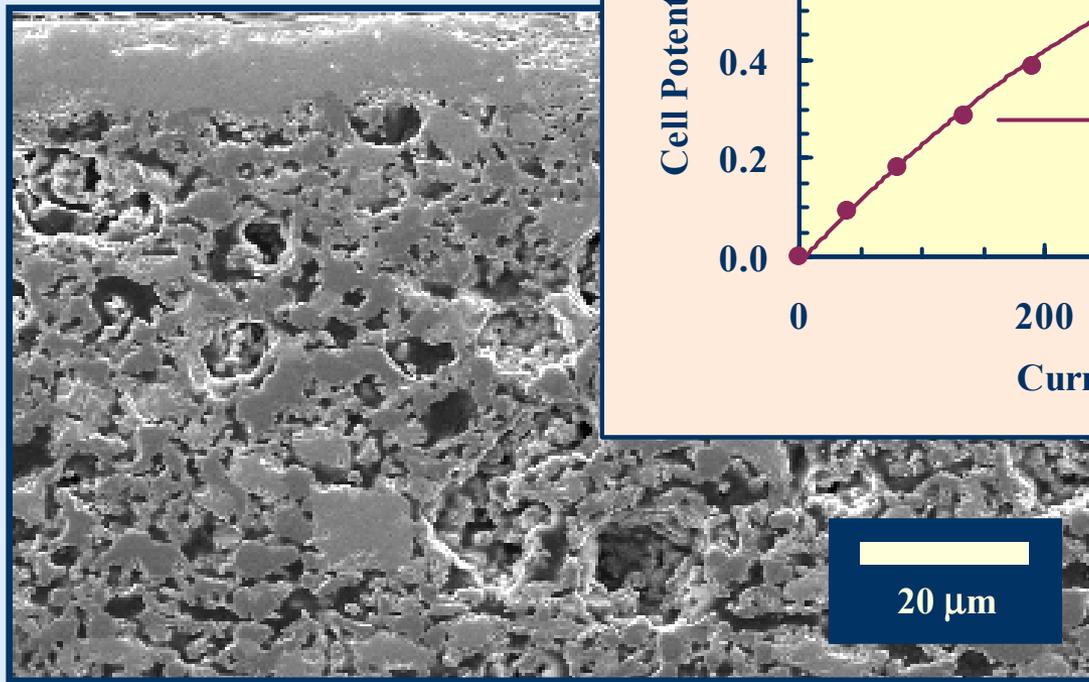
Ceria-Based Electrolytes

Lower-Cost Ceria Electrolytes

Ionic Conductivity at 600°C
and Raw Materials Cost



Thin-Film Ceria SOFCs



Cathode Development

- Evaluate nano-composite approaches
- Optimization of formulations and processing
- Integrate ac-impedance into measurement protocol

SOFC Testing (NexTech and Northwestern)

- Effect of cathode material
- Effect of anode microstructure
- Functional interlayers

Additional Collaboration with Ohio State and ORNL on Mechanical Property Measurements

Long-Term Single-Cell Testing (GTI)

Acknowledgments

**Thanks to DOE, NETL,
and the State of Ohio!**

Team Members

- **Harlan Anderson (UMR)**
- **Tim Armstrong (ORNL)**
- **Scott Barnett (Northwestern)**
- **Bob Remick and Chuck Sishla (GTI)**
- **Mike Cobb and Kirby Meacham (Cobb & Co.)**
- **Jim Stephan (Advanced Materials Technologies)**
- **John Lannutti and Mark Walter (Ohio State University)**
- **Russ Bennett and Gary Kapp (EMTEC)**

NexTech Colleagues

- **Bill Dawson**
- **Matt Seabaugh**
- **Buddy McCormick**
- **Kathy Hasinska**
- **Chris Holt**
- **A-M Azad**
- **Jerry Jayjohn**