Continuous Process for Low-Cost, High-Quality YSZ Powder

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Project Details and Team

- **U.S. Department of Energy (SECA)**
  - DOE Contract Number: DE-FC26-02NT41575
  - Project Monitor: Shawna Toth
  - Phase I: 10/1/2002 through 9/30/2003
  - Phase II: 10/1/2003 through 9/30/2005
- **State of Ohio (Third Frontier Program)**
  - *Building the Domestic Infrastructure for Solid Oxide Fuel Cells*
  - ODOD Contract Number: TECH 03-035
- **NexTech's Team**
  - Principal Investigator: Scott Swartz
  - Lead Engineer: Michael Beachy
  - Scientific Support: Matt Seabaugh
  - Technical Support: NexTech's Fuel Cell Group

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Continuous Process for Low-Cost, High-Quality YSZ Powder

Outline

- Technical Issues Addressed
- R&D Objectives and Approach
- Results to Date
- Applicability to SOFC Commercialization
- Activities for the next 12 Months

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Technical Issues being Addressed

- Low-cost scalable powder synthesis/production processes.
- Lower sintering temperatures.
- Effects of dopants and processing methods on conductivity and mechanical properties.
- Long-term degradation of ionic conductivity of zirconia-based electrolytes.
- Tailoring of the YSZ electrolyte powder for different SOFC fabrication processes.
- Batch-to-batch reproducibility.

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R&D Objectives

Development of a low-cost synthesis process for YSZ electrolyte powder tailored for SOFC fabrication processes

- **Process Development Goals**
  - Homogeneous precipitation
  - Utilization of low-cost precursors
  - Continuous – where possible
  - Aqueous
  - Agile
- **Powder Quality Metrics**
  - Surface area: ~10 m^2/gram
  - Average particle size: <0.5 microns
  - Sinterability: $\rho \sim 98\%$ theoretical at $T_S < 1300^oC$
  - Ionic conductivity: $\sigma > 0.05$ S/cm at 800°C

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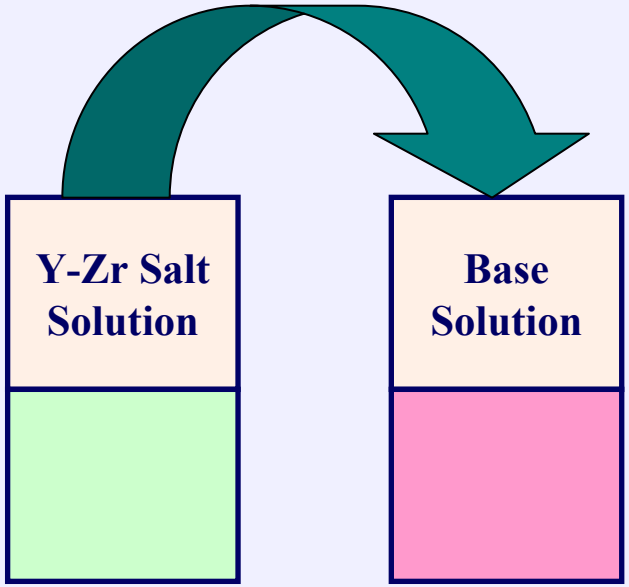
Continuous Precipitation

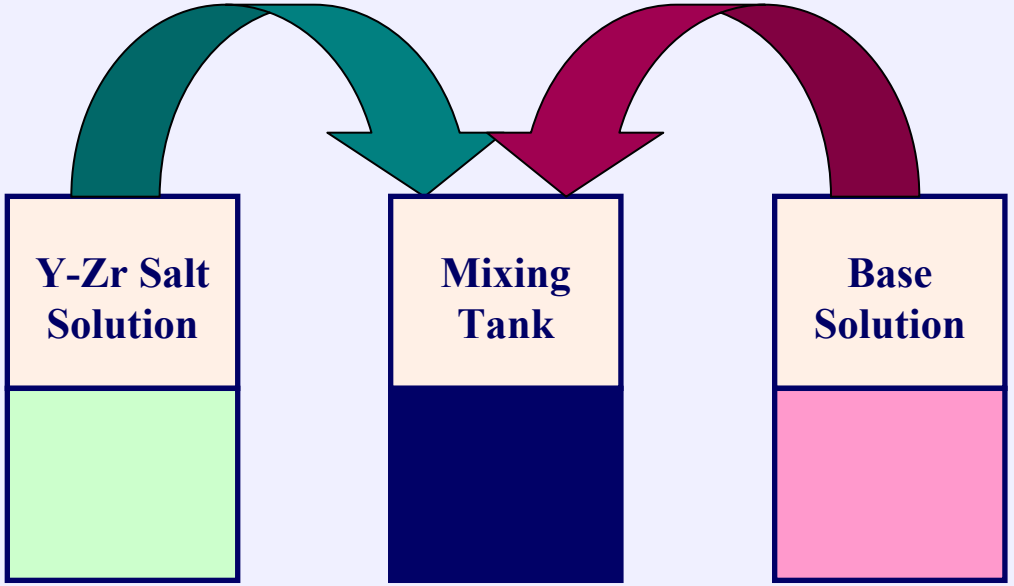


Standard Precipitation

pH varies continuously during process

Homogeneous Precipitation

pH remains constant throughout process



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Powder Processing Approach

- Precipitation
- Filtration/Washing
- Drying/Sieving
- Calcination
- Milling
- Drying/Sieving

Synthesis Process Variables

- Batch Size (typically 3-5 kg)
- *Precipitation Conditions*
- Chemical Purity (e.g., silica content)
- Dopants – sintering aids
- *Solvent System (water or alcohol)*
- *Drying Methods*
- Calcination – control of surface area
- *Milling Methods – particle size control*

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Powder Evaluation Protocol

- **Powder Characterization**
  - Particle Size Distribution (centrifugal analysis)
  - Surface Area (multi-point BET)
  - Chemical Analysis (ICP)
- **Sintering Performance Studies**
  - Samples: pressed pellets or tape-cast substrates
  - Temperature range: 1100 to 1400°C
  - Density measurements by Archimedes method
- **Characterization of Sintered YSZ Ceramics**
  - Ionic conductivity (four-point method)
  - Long-term conductivity testing
  - Mechanical properties
  - Microstructural analyses

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Process Development Challenges

- Assuring “apples to apples” comparisons.
- Drying and milling processes are more efficient at larger production scales.
- Labor-intensive process at current scale of production.
- Complex relationships between precipitation variables on downstream processes.
- Difficulty in achieving absolute control of surface area and particle size simultaneously.
- Lack of accelerated tests for long-term stability.

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Results to date (including Phase I)

- Established homogeneous precipitation process for synthesis of YSZ powders.
- Established calcination and milling methods to meet surface area and particle size targets.
- Achieved state-of-the-art performance levels, relative to commercially available YSZ powders:
  - Improved low-temperature sinterability (at same surface area)
  - Achieved identical ionic conductivity values
- Demonstrated potential for achieving manufacturing cost of less than $25/kg target.
- Identified cost drivers for process.

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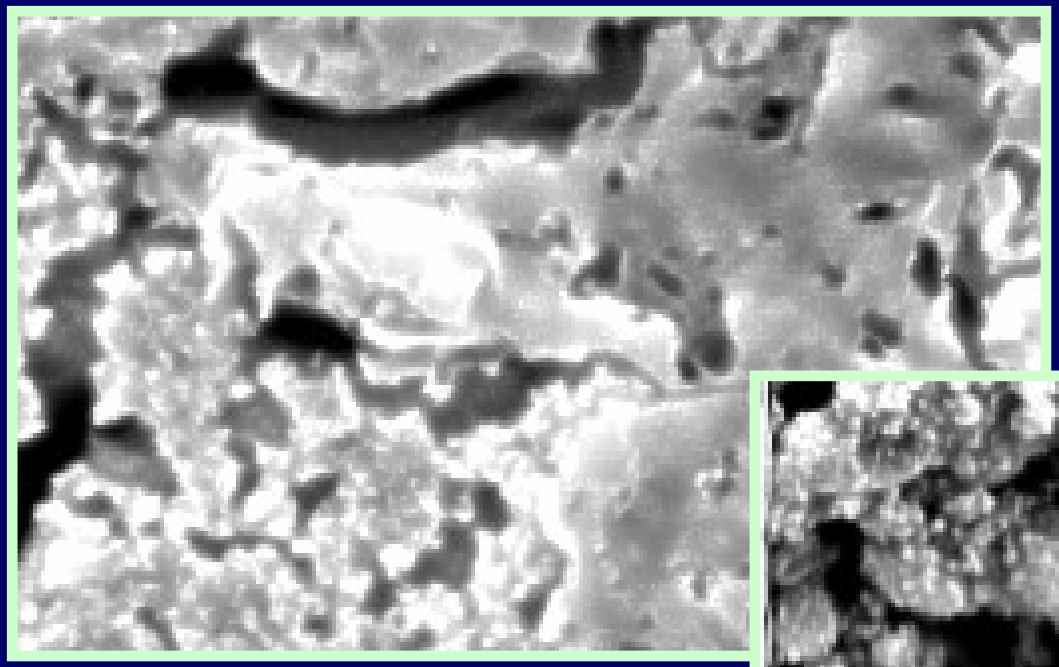
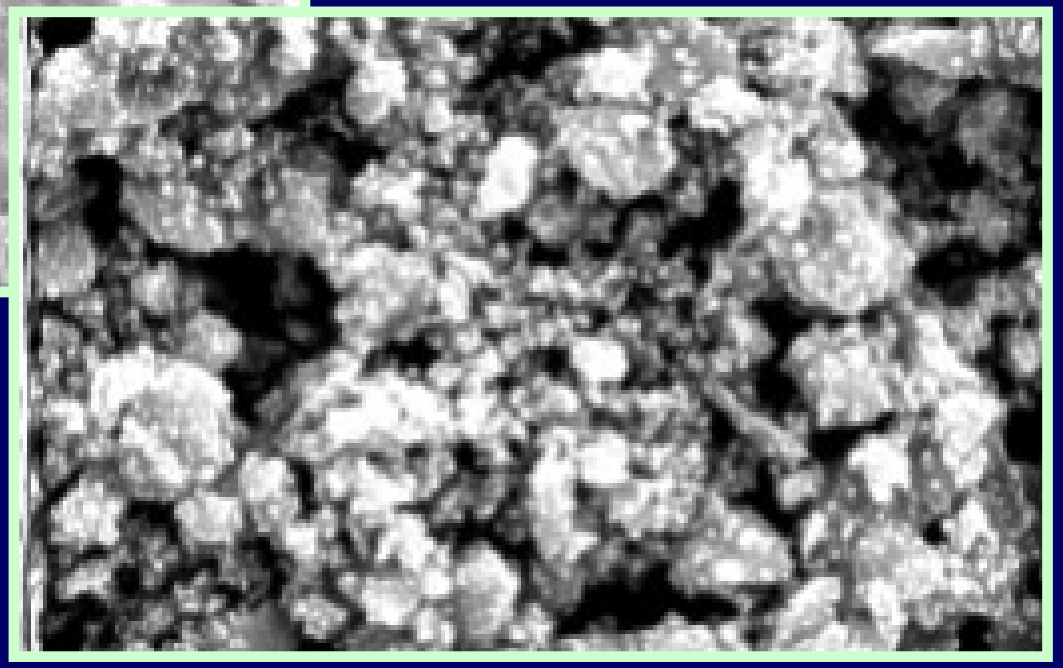
Non-Optimized Process



As-Precipitated

Calcined and Milled



1 μm

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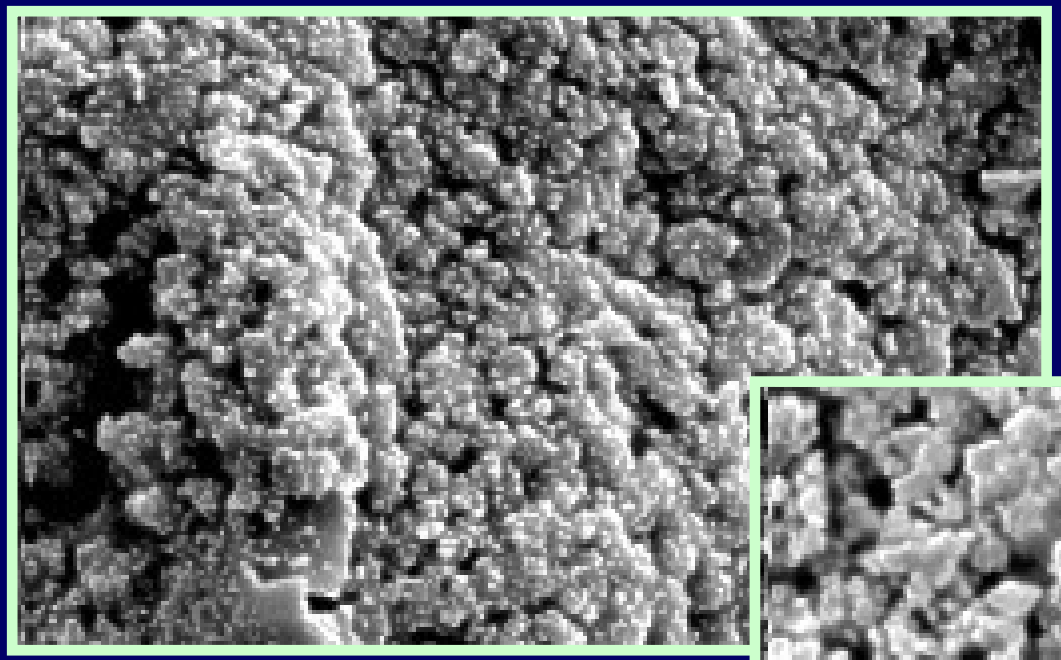
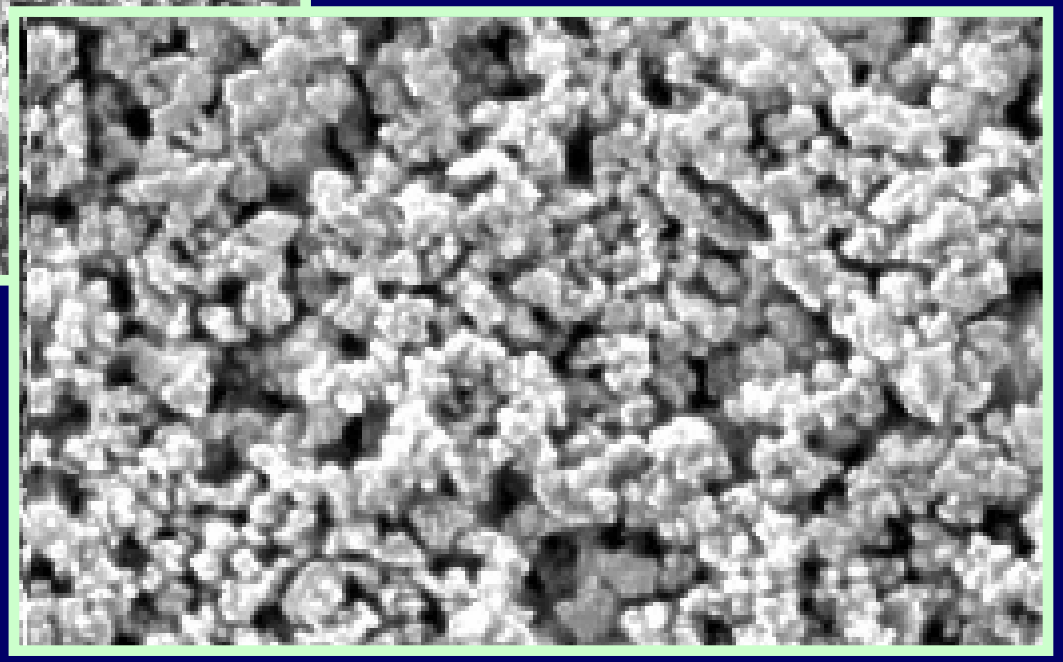
Optimized Process



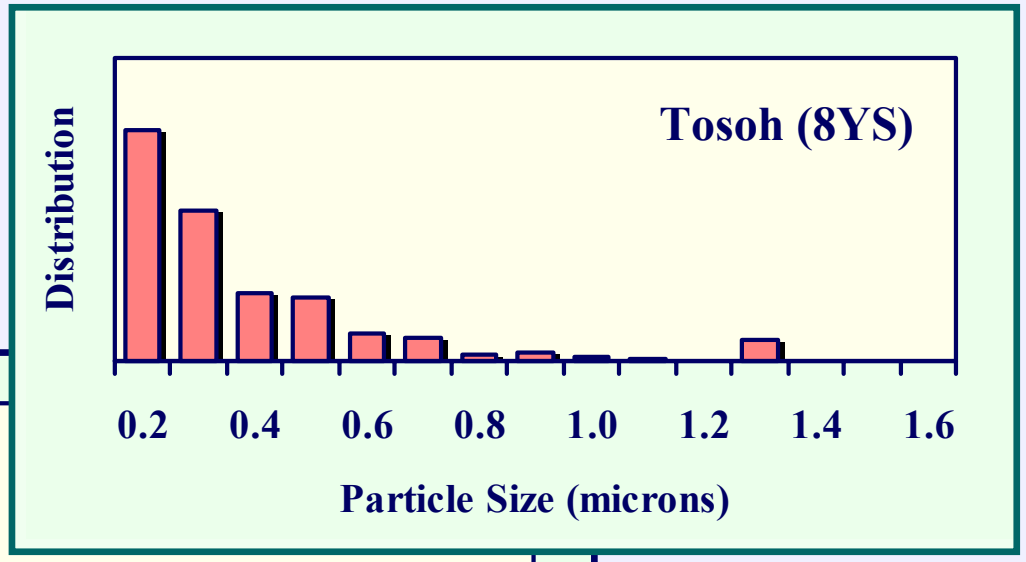
As-Precipitated

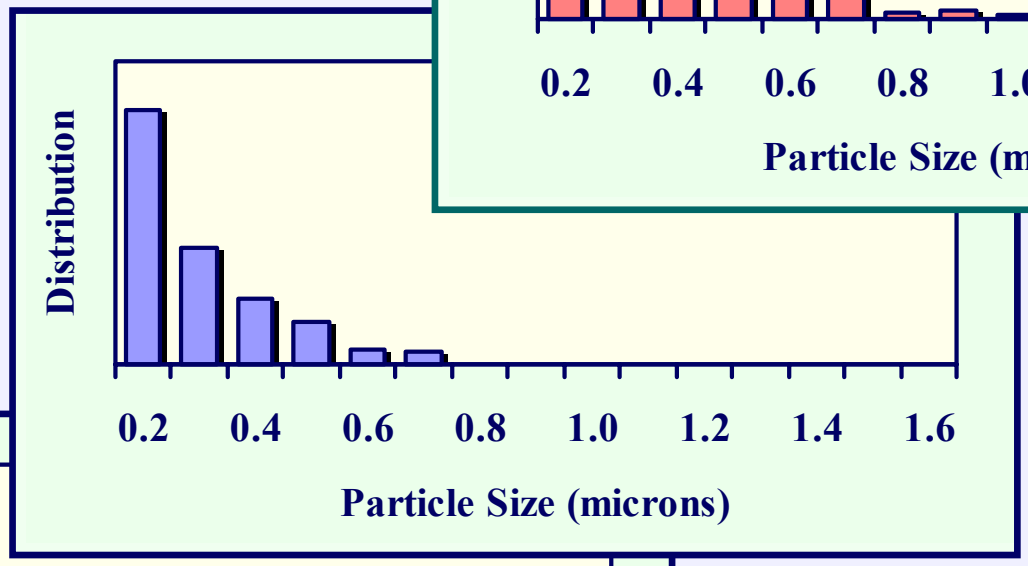
Calcined and Milled



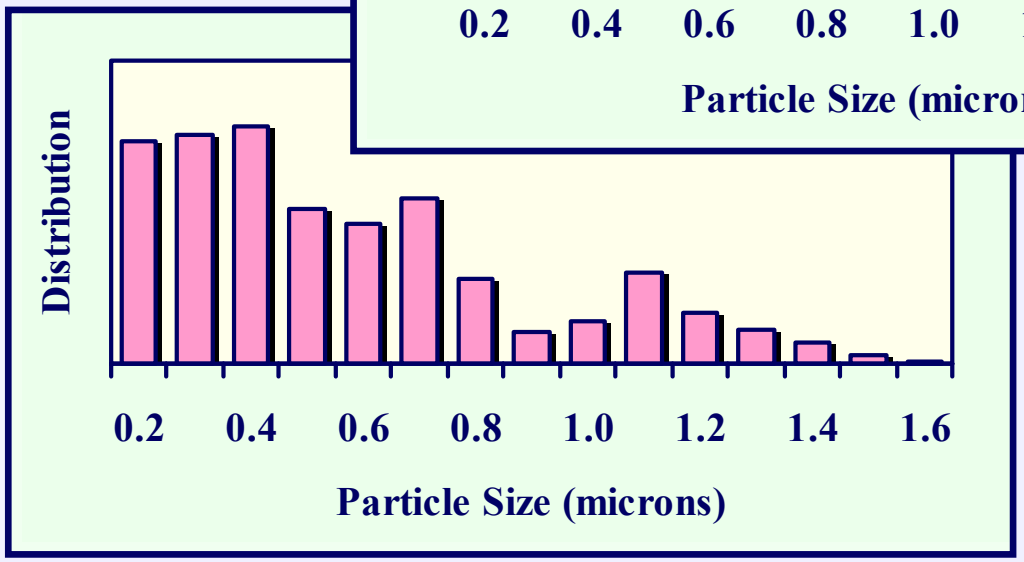
1 µm

Particle Size Distributions





Optimized Milling Methods



Non-Optimized Milling Methods

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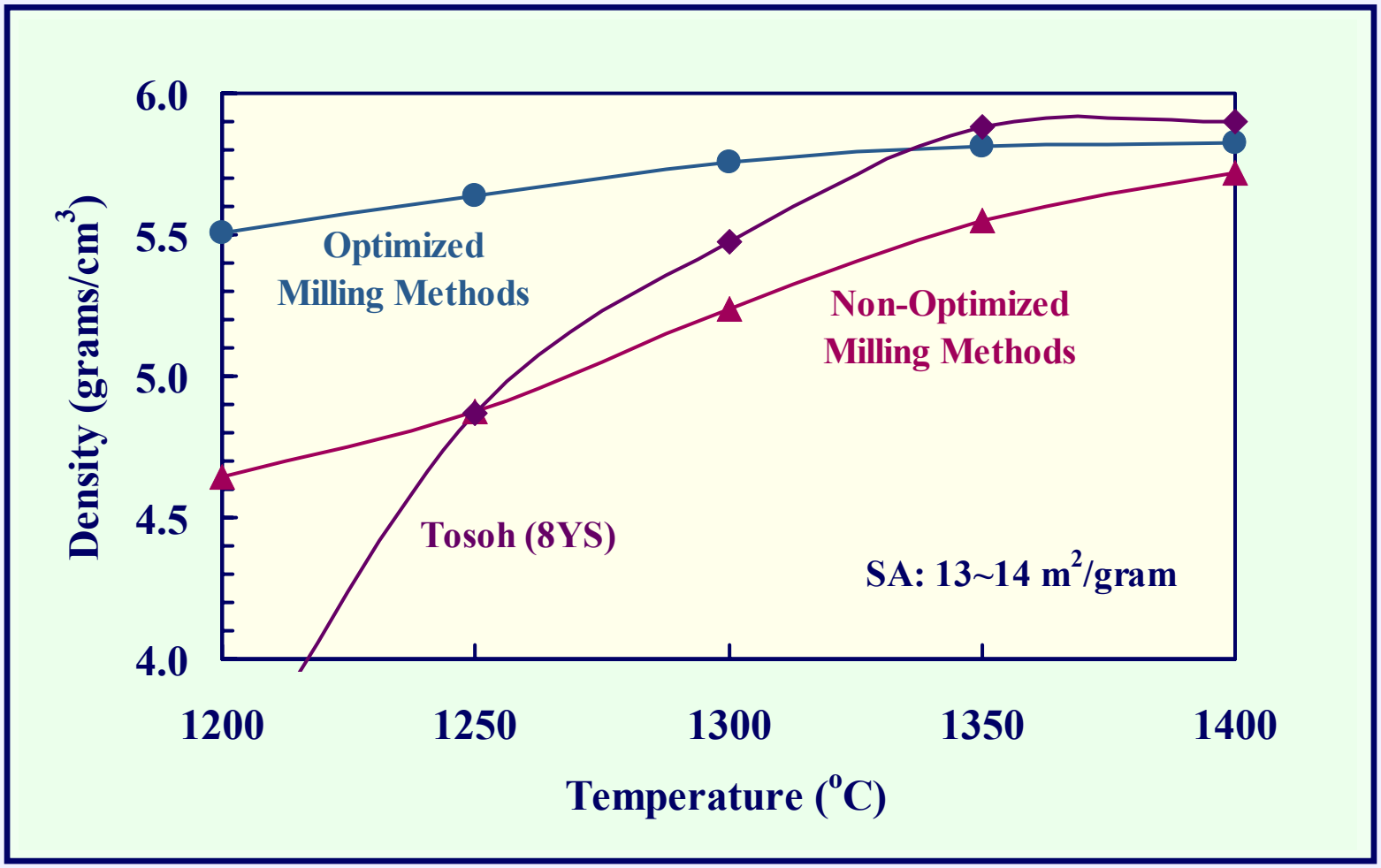
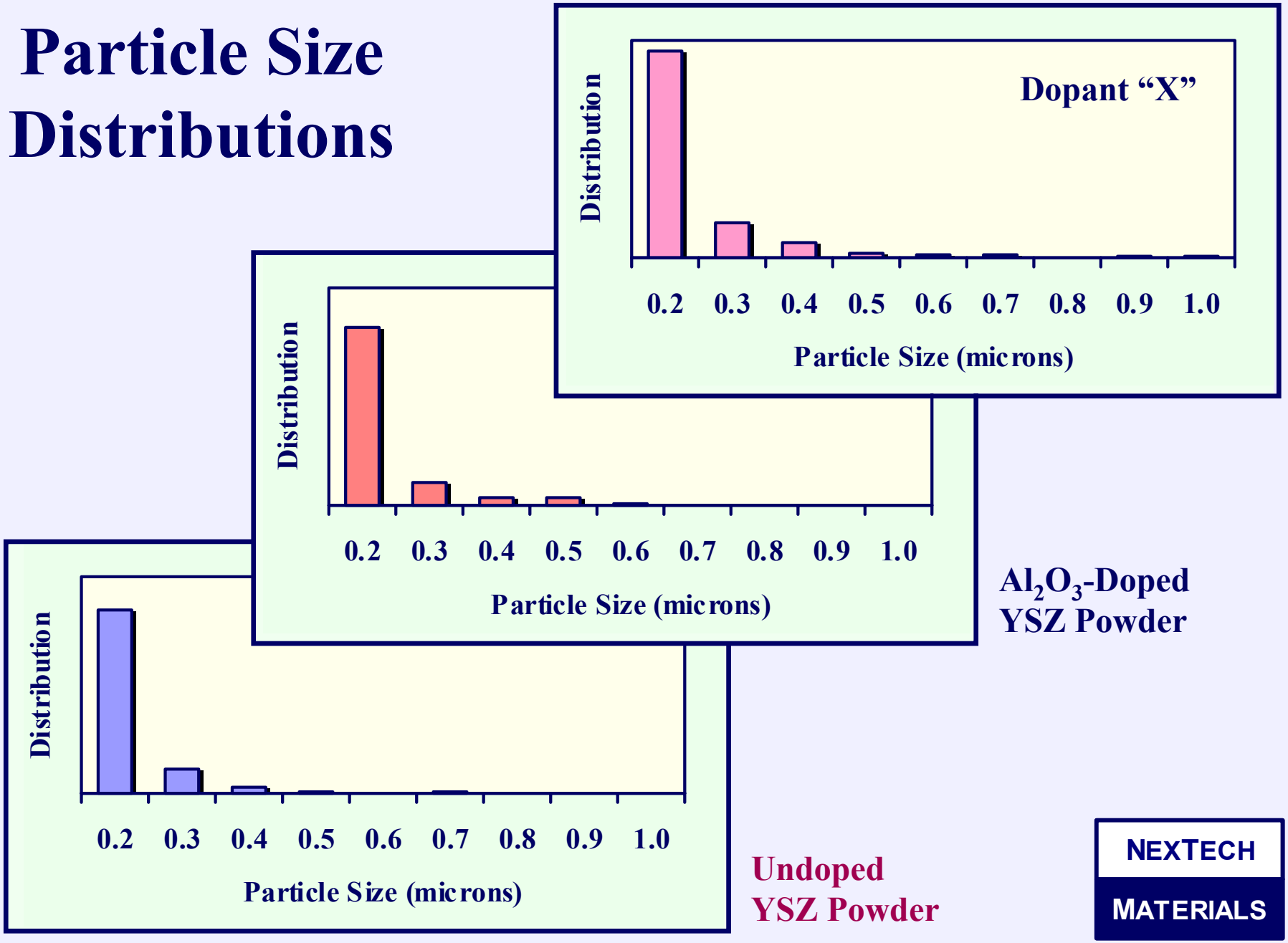
Effect of Milling Methods on Sintering Performance



Particle Size Distributions



Al_2O_3-Doped YSZ Powder

Undoped YSZ Powder

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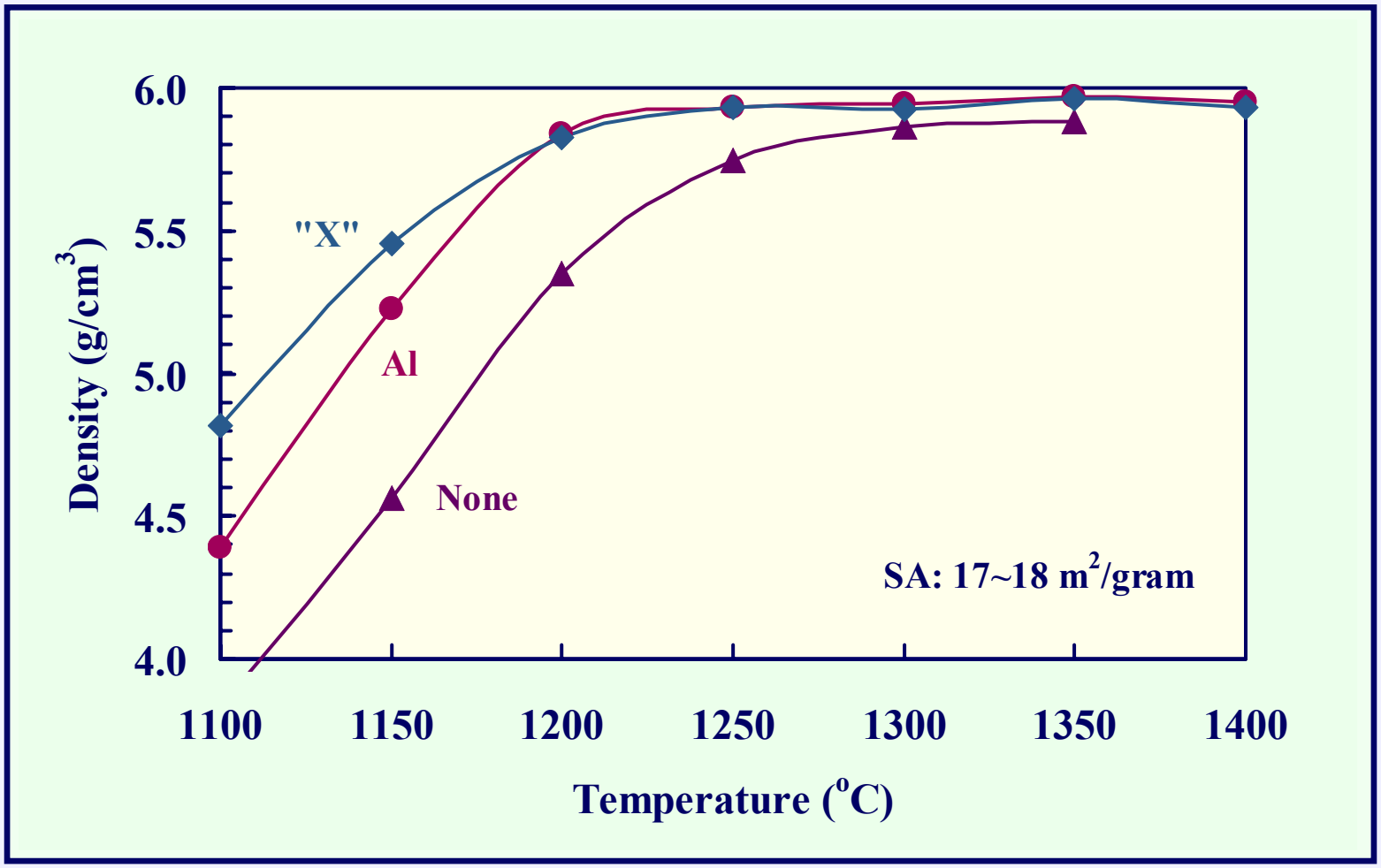
Effect of Dopants on Sintering Performance



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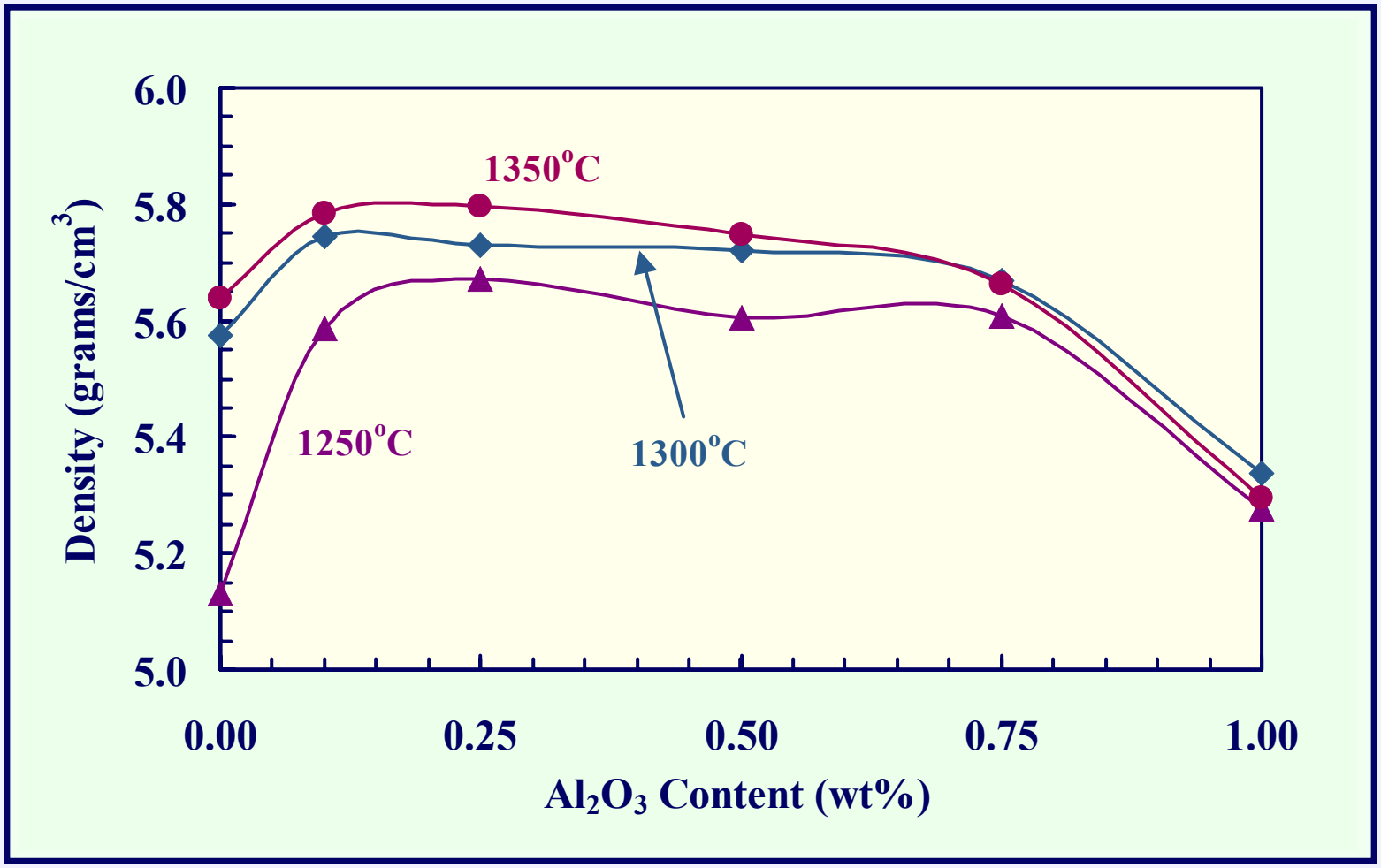
Effect of Al_2O_3 Dopant Content on Sintering Performance



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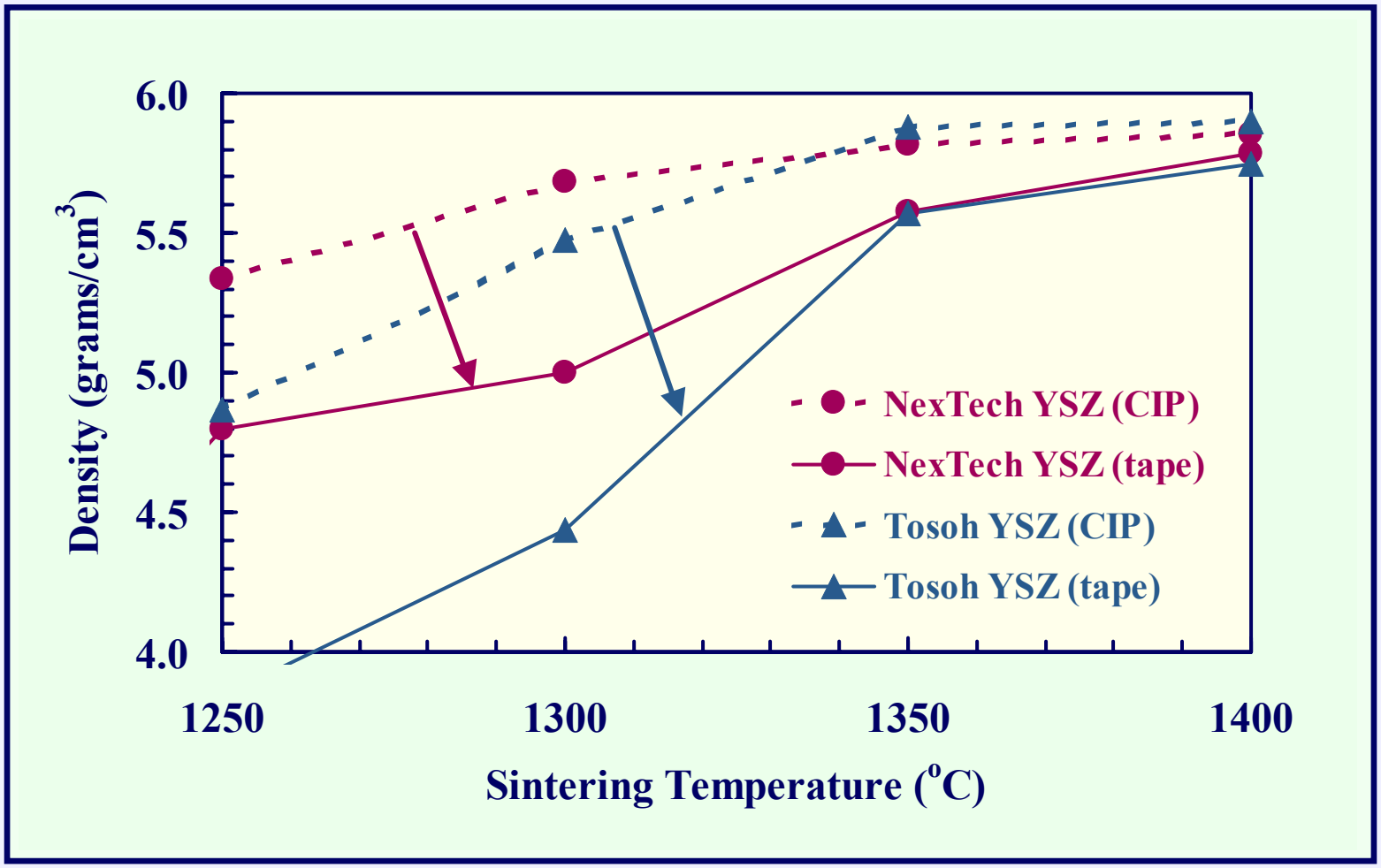
Effect of Fabrication Method on Sintering Performance



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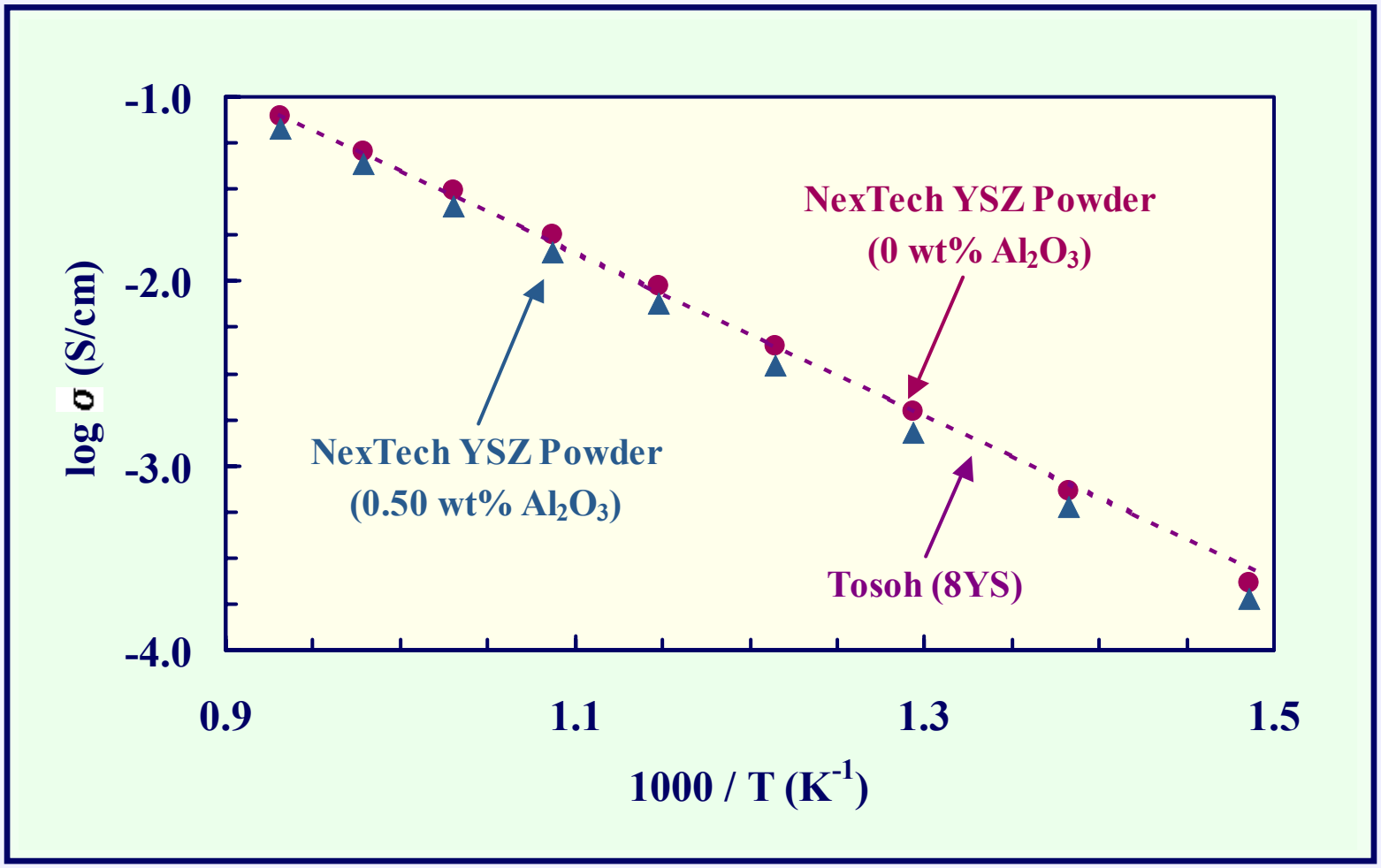
Ionic Conductivity Measurements



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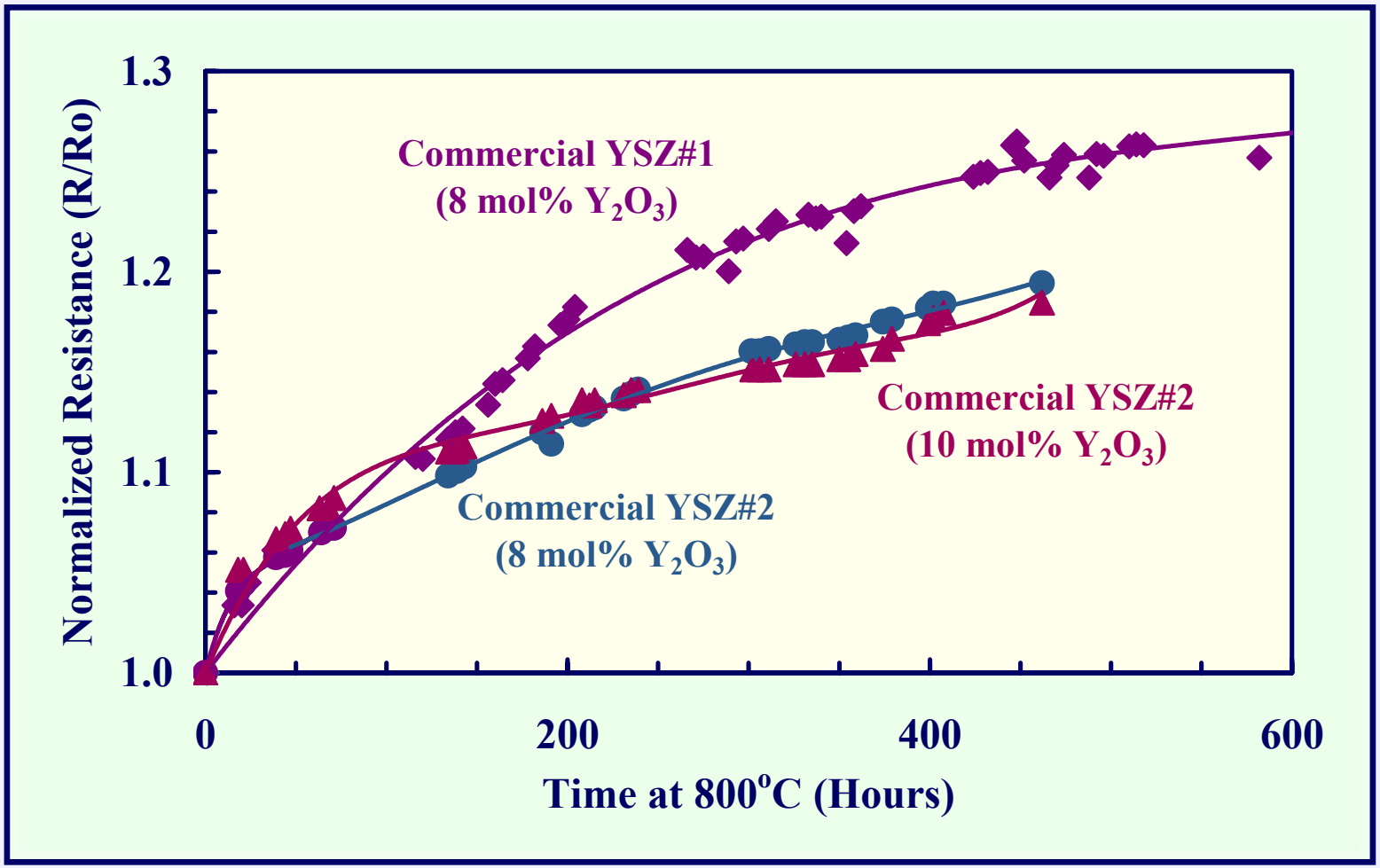
Aging of Ionic Conductivity



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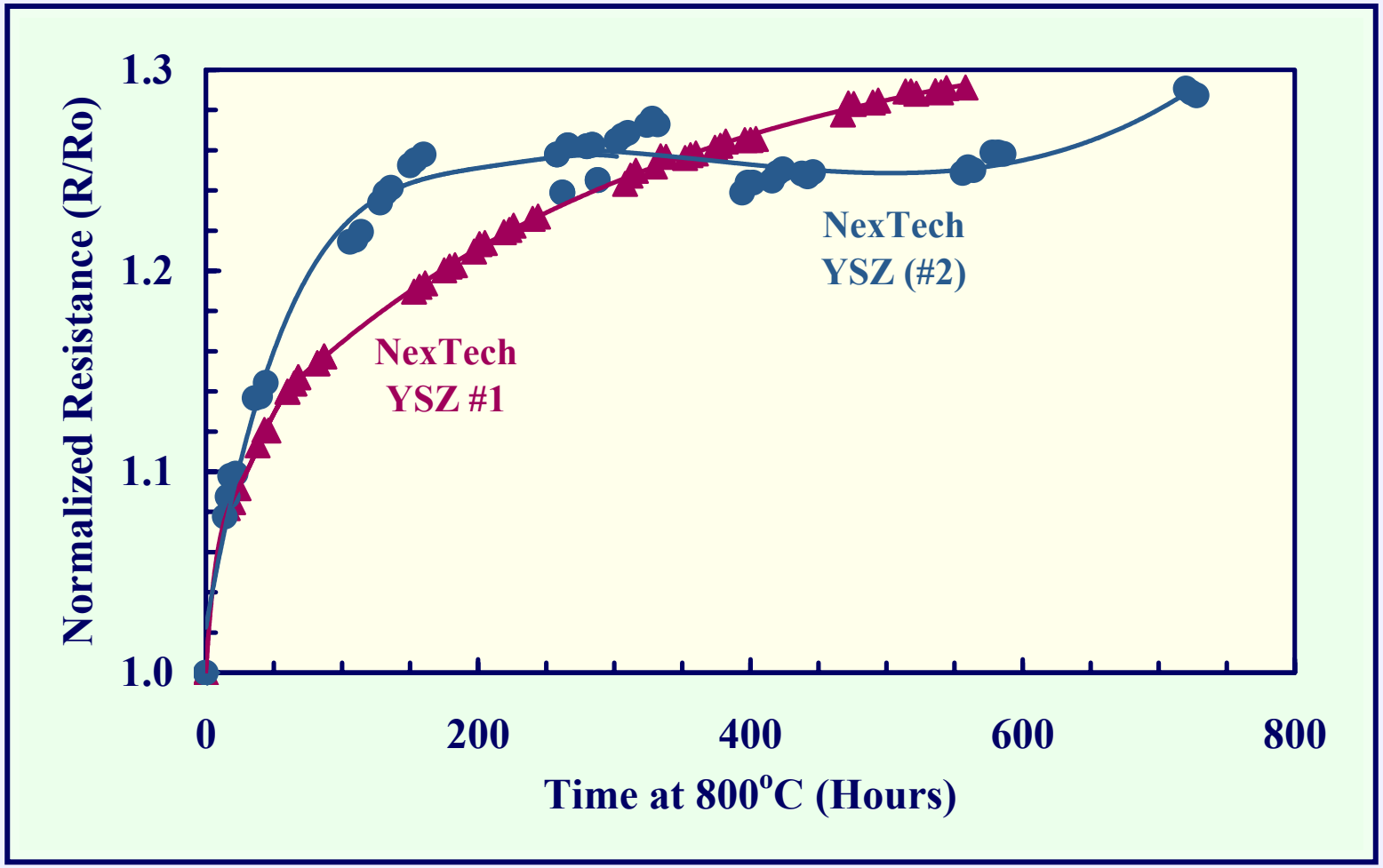
Aging of Ionic Conductivity



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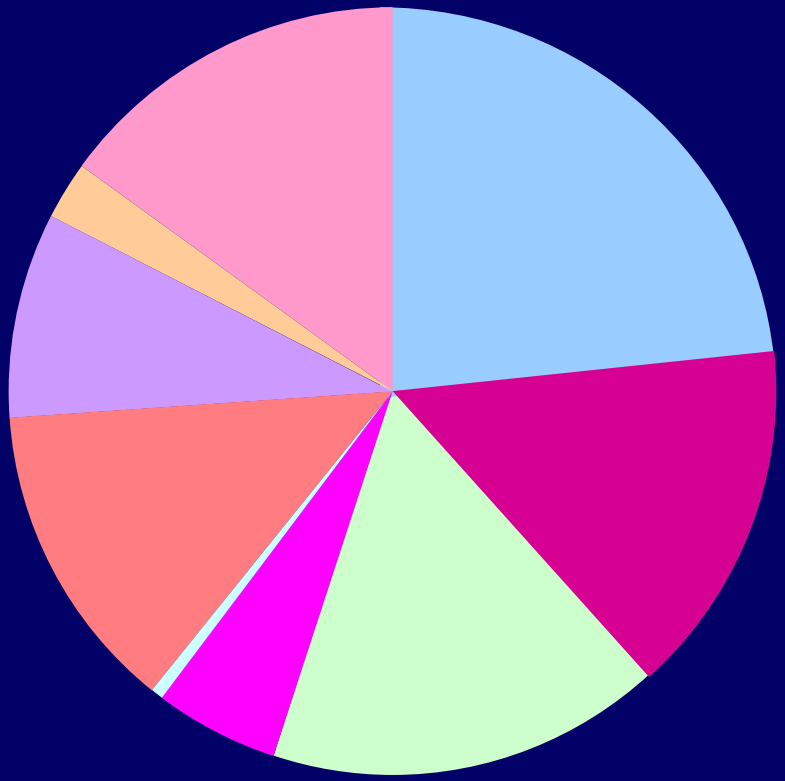
Manufacturing Cost Estimate

- **Basis of Calculations:**
  - Plant size: 500 MT/year
  - Fixed capital investment: $11.2 M
  - Cost per kilogram of YSZ: $23.56



- Raw Materials: $5.27
- Utilities: $3.37
- Labor: $3.75
- Maintenance & Repairs: $1.39
- Operating Supplies: $0.17
- Plant Overhead: $2.93
- Depreciation: $1.90
- Local Taxes and Insurance: $0.57
- General Expenses: $3.37

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Applicability to SOFC Commercialization

YSZ powder must be tailored for different manufacturing processes used for anodes and electrolyte layers.

SECA Industry Team	Electrolyte Fabrication	Anode Fabrication
Delphi/Battelle	Tape Casting	Tape Casting
GE	Tape Calendaring	Tape Calendaring
Cummins/SOFCo	Tape Casting	Screen Printing
SWPC	Plasma-Spray	Plasma Spray
Fuel Cell Energy	Screen Printing	Tape Casting
Accumentrics	Dip Coating	Extrusion

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Applicability (continued)

Agile processing will allow tailoring to requirements of SOFC fabrication methods and different developers.

- **Tape Casting Methods:** Tight control of particle size distribution is important; relatively low surface areas needed for high green density.
- **Co-Sintering Processes:** Lower sintering temperatures are desired; control of sintering shrinkage rates is essential.
- **Colloidal Deposition:** Dispersion chemistry is critical; higher surface areas can be tolerated; tailored particle size distributions are beneficial.
- **Plasma-Spray Methods:** Large particle size and spherical powder morphology are required for optimum flow characteristics.
- **Extrusion:** Lower surface areas needed for dimensional control and green strength; particle size requirements vary by developer.

Batch-to-batch reproducibility is essential for all processes!

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Phase II Work Plan (Year 1)

- **Survey of SECA Industry Teams**
- **Process Development and Scale-Up**
  - Process refinements (especially washing and drying steps)
  - Chemical analyses through all processing steps
  - Scale-up to 10-20 kg batch sizes
  - Evaluation of batch-to-batch reproducibility
  - Electrical and mechanical property testing
- **Validation of Alumina Doping Strategies**
  - Evaluation of dopant incorporation methods
  - Chemical analyses
  - Comprehensive microstructural analyses
  - Electrical and mechanical property testing
  - Long-term testing

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Phase II Work Plan (Year 2)

- **Demonstration of Process Reproducibility**
- **Demonstrations in SOFC Fabrication Processes**
  - Preparation of composite (NiO/YSZ) anode powders
  - Tape casting of anode substrates
  - Co-sintering of anode-supported cells
  - Screen-printed anode coatings
  - *Special Requests*
- **Production of Evaluation Samples**
  - YSZ electrolyte powder
  - NiO/YSZ anode powder
  - Fabricated components
- **Manufacturing Cost Analyses**

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Acknowledgments