Novel Functional Graded Thermal Barrier Coatings in Coal-fired Power Plant Turbines

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PRAXAIR SURFACE TECHNOLOGIES

- **Subcontractor** (Praxair Surface Technologies)**:** James Knapp
- **Industrial collaborators** (Praxair Surface Technologies)**:** John K. Anderson, Vlad Belov, Don Lemen, Li Li
- **Graduate students** (IUPUI)**:** Xingye Guo, Yi Zhang

Outline of Talk

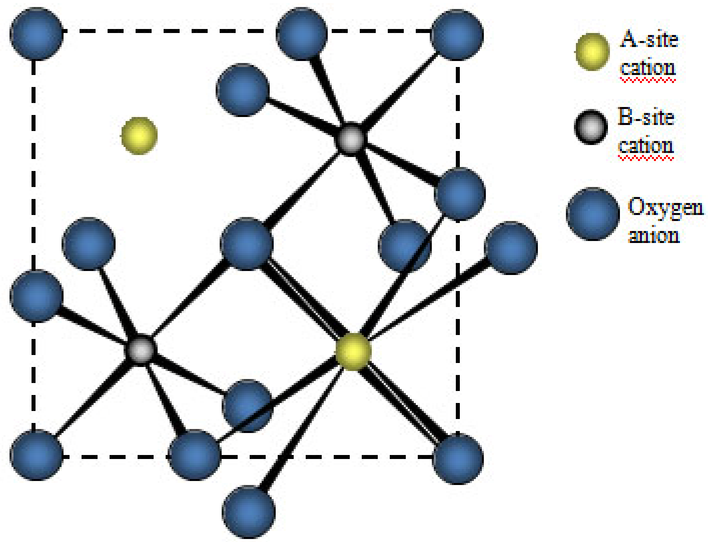
- **I. Introduction**
  - i. Pyrochlore oxide
  - ii. Double-layer thermal barrier coating
- **II. Results (September 1, 2012- June 1,2013)**
  - i. Powder fabrication and characterizations
  - ii. Design of double-layer structure
- **III. Summary / Future work**

Goals

- The objective of the project is to investigate a novel double-layer functional graded coating material, pyrochlore oxide, for thermal barrier coating (TBC) applications.
- The ultimate goal is to develop a manufacturing process to produce the pyrochlore oxide based coating with improved high-temperature corrosion resistance.

Pyrochlore

Pyrochlore-type rare earth zirconium oxides ($Re_2Zr_2O_7$, Re = rare earth) are promising candidates for thermal barrier coatings, high-permittivity dielectrics, potential solid electrolytes in high-temperature fuel cells, and immobilization hosts of actinides in nuclear waste.

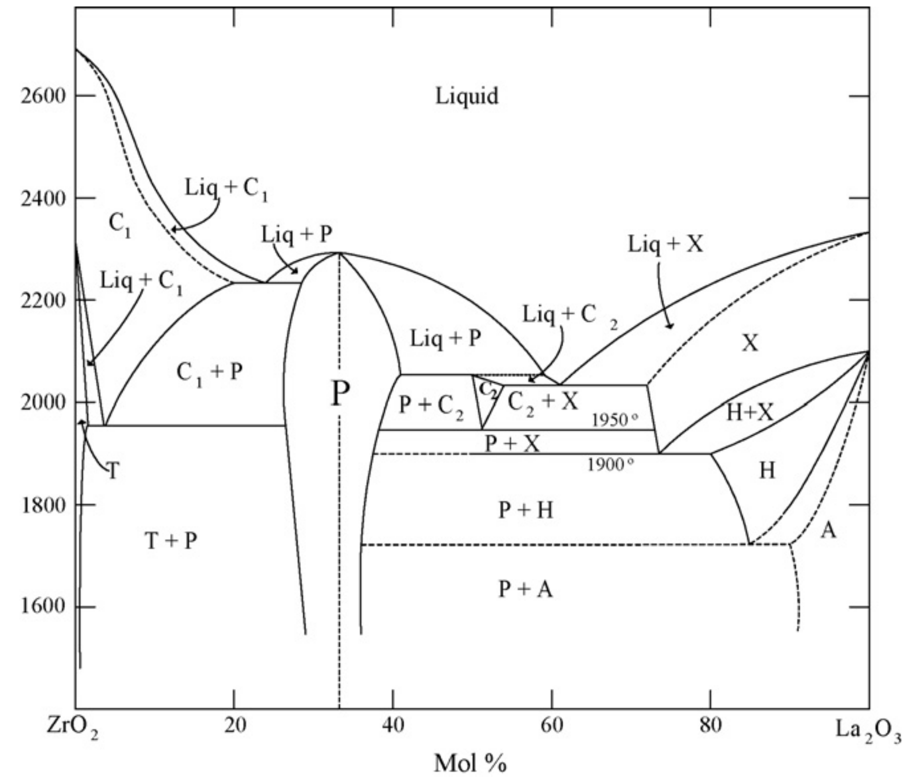


Pyrochlore crystal structure: $A_2B_2O_7$. A and B are metals incorporated into the structure in various combinations. (credit: NETL)

Why $La_2Zr_2O_7$?

Compared with YSZ, $La_2Zr_2O_7$ has

- Higher temperature phase stability. No phase transformation
- Lower sintering rate at elevated temperature
- Lower thermal conductivity
- Lower CTE (can be enhanced by CeO_2 doping)



Phase diagram of La_2O_3–ZrO_2

YSZ vs. $La_2Zr_2O_7$

Materials property	8YSZ	$La_2Zr_2O_7$
Melting Point (^oC)	2680	2300
Maximum Operating Temperature (^oC)	1200	>1300
Thermal Conductivity (W/m-K) (@ 800^oC)	2.12	1.6
Coefficient of Thermal Expansion ($x10^{-6}$/K) (@1000 ^oC)	11.0	8.9-9.1
Density (g/cm^3)	6.07	6.00
Specific heat (J/g-K) (@1000 ^oC)	0.64	0.54

$La_2Zr_2O_7$ Fabrication Methods

- Solid-state reaction [1]
- Coprecipitation–calcination method [2]
- Sol-gel method [3]

However, currently only small quantity powders were made at lab scale. There is an urgent need to develop a scalable method to produce large quantity of $La_2Zr_2O_7$

1. *Cao, Vassen et al, Spray-drying of ceramics for plasma-spray coating, J. Eur. Ceram. Soc.,20,2433-2439 (2000)*
2. *Zhou et al, Preparation and thermophysical properties of CeO2 doped La2Zr2O7 ceramic for thermal barrier coatings, J. Alloy Compd, 438, 217-221 (2007)*
3. *Kido, Komarneni, Roy, Preparation of La2Zr2O7 by Sol - gel route, J. Am. Ceram. Soc.,74, 422–424 (1991)*

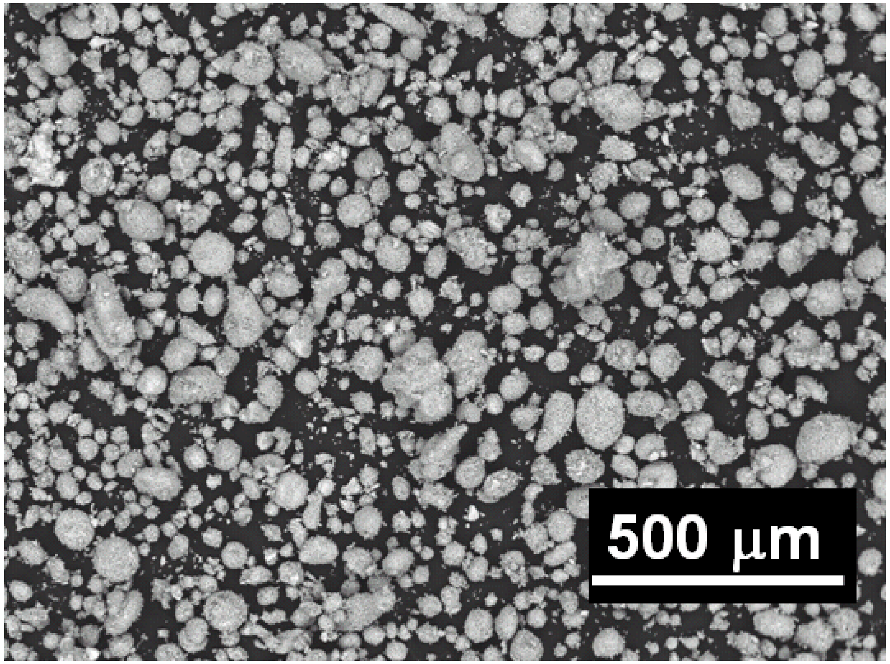
Layered Coating System

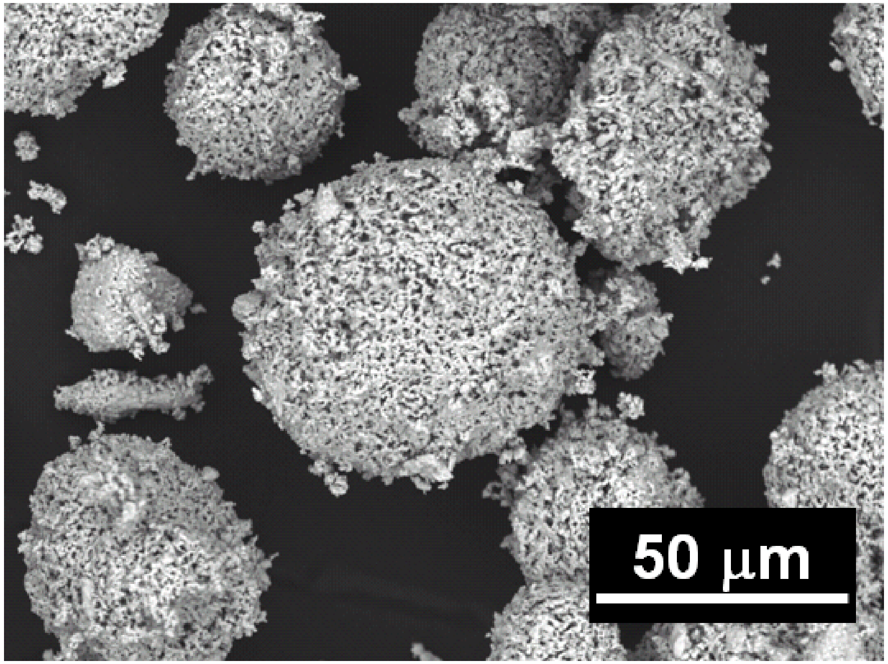
- The coefficient of thermal expansion of $La_2Zr_2O_7$ ($10x10^{-6}$/K) is lower than those of both substrate and bondcoat (about $15x10^{-6}$/K). As a result, the thermal cycling properties may be a concern
- The layered topcoat is believed to be a feasible solution
- In this work, we develop a double-layer, functionally graded, pyrochlore oxide based TBC system

Scalable Thermal Spray Powder Production

- The powders are produced using a reengineered spray drying and sintering technique at Praxair Surface Technologies.
- La_2O_3 and ZrO_2 particles are mixed into a water based slurry, and then spray dried or atomized into a powder form, using a rotary wheel style spray drier to convert the liquid slurry into dry particles of an agglomerated La_2O_3/ZrO_2 powder.
- These spray dried particles are sintered in a gas fired kiln to achieve powder particles of an appropriate particle structure.
- The sintered cake is de-agglomerated to break down the cake, and screened to a particle size distribution suitable for spray.
- The resulting powder is then blended in a "V" blender to make a homogenous mixture of the variety of particle sizes.
- Approximately 150 lb of $La_2Zr_2O_7$ powders are produced in a single batch.

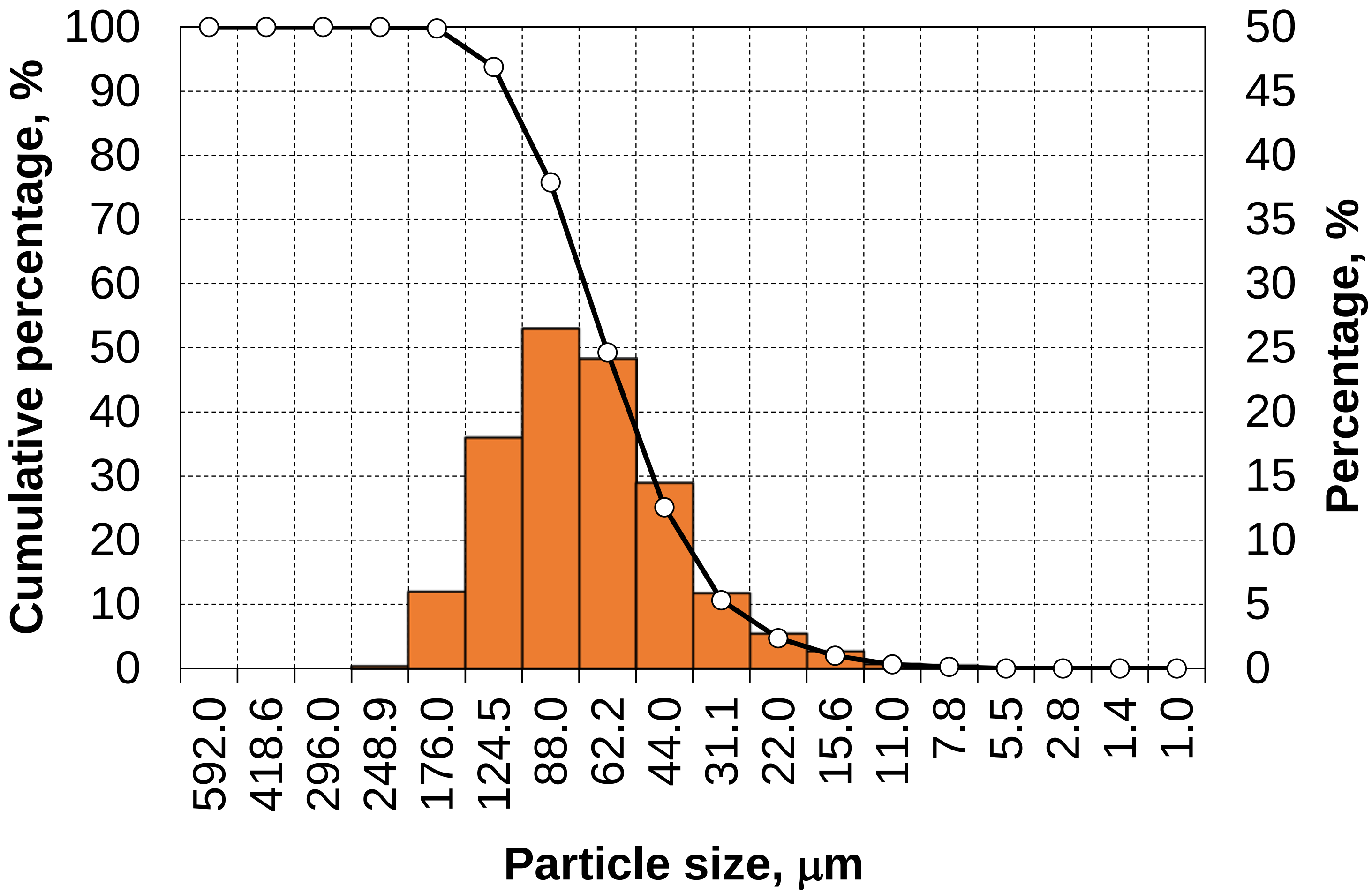
$La_2Zr_2O_7$ Powder Morphology





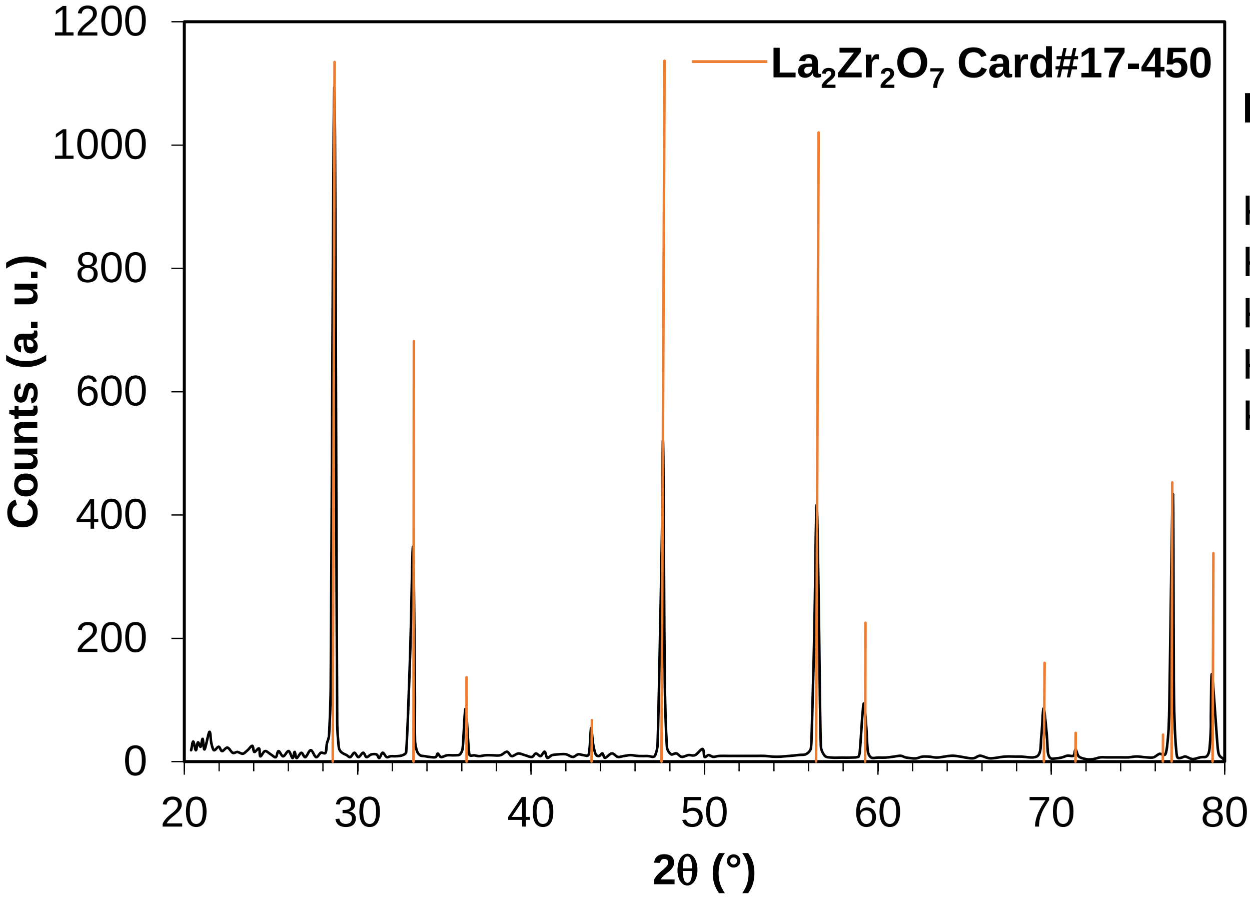
- Spherical shape with porous surface
- Good flowability and high density
- Particle size between 30 – 100 μm

Powder Size Distribution (PSD)



Microtrac standard range particle analyzer's percent passing data show that the average powder size, D_{50}, is ~65 μm

X-ray Diffraction (XRD) Analysis



PANalytical/Philips X'pert MRD

$K_{\alpha1}$ wavelength: 1.5405600 Å
$K_{\alpha2}$ wavelength: 1.5443900 Å
$K_{\alpha2}$ / $K_{\alpha1}$ intensity ratio: 0.5000
K_{α} wavelength: 1.5405600 Å
K_{β} wavelength: 1.3922200 Å

XRD data show that the powder composition is $La_2Zr_2O_7$

Chemical Composition - ICP-MS

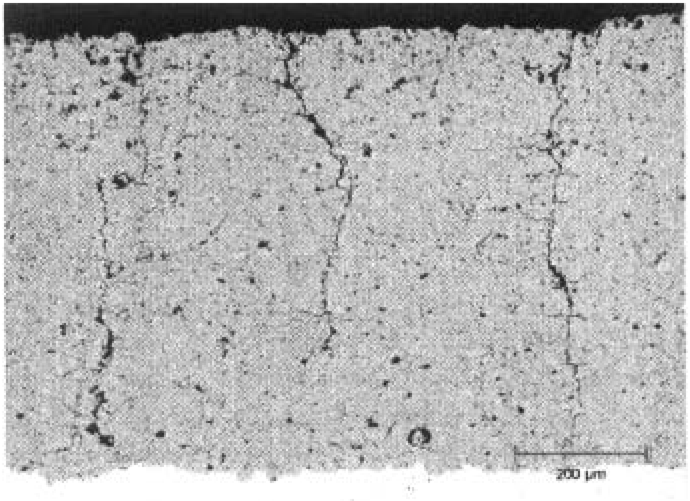
All elements measured in weight percent unless otherwise specified. Sampling Method per ASTM B215.

Chemistry	Test Method	Test Lab	Min	Max	Result	OK
Aluminum Oxide	ICP	NSL Analytical Services		0.2	<0.1	Yes
Ferric Oxide	ICP-MS	NSL Analytical Services		0.5	0.1	Yes
Hafnium Oxide	ICP	NSL Analytical Services		2.5	0.8	Yes
Lanthanum Oxide	By Difference	NSL Analytical Services			57	Yes
Other Oxides Total	ICP-MS	NSL Analytical Services		1.5	0.4	Yes
Silicon Dioxide	ICP	NSL Analytical Services		1.0	0.7	Yes
Titanium Dioxide	ICP-MS	NSL Analytical Services		0.5	0.0	Yes
Uranium + Thorium	ICP-MS	NSL Analytical Services		0.05	0.02	Yes
Zirconium Oxide	ICP	NSL Analytical Services			41	Yes

- Inductively coupled plasma – mass spectrometry (ICP-MS) technique was used to measure the powder compositions
- The measurements confirms $La_2Zr_2O_7$ composition

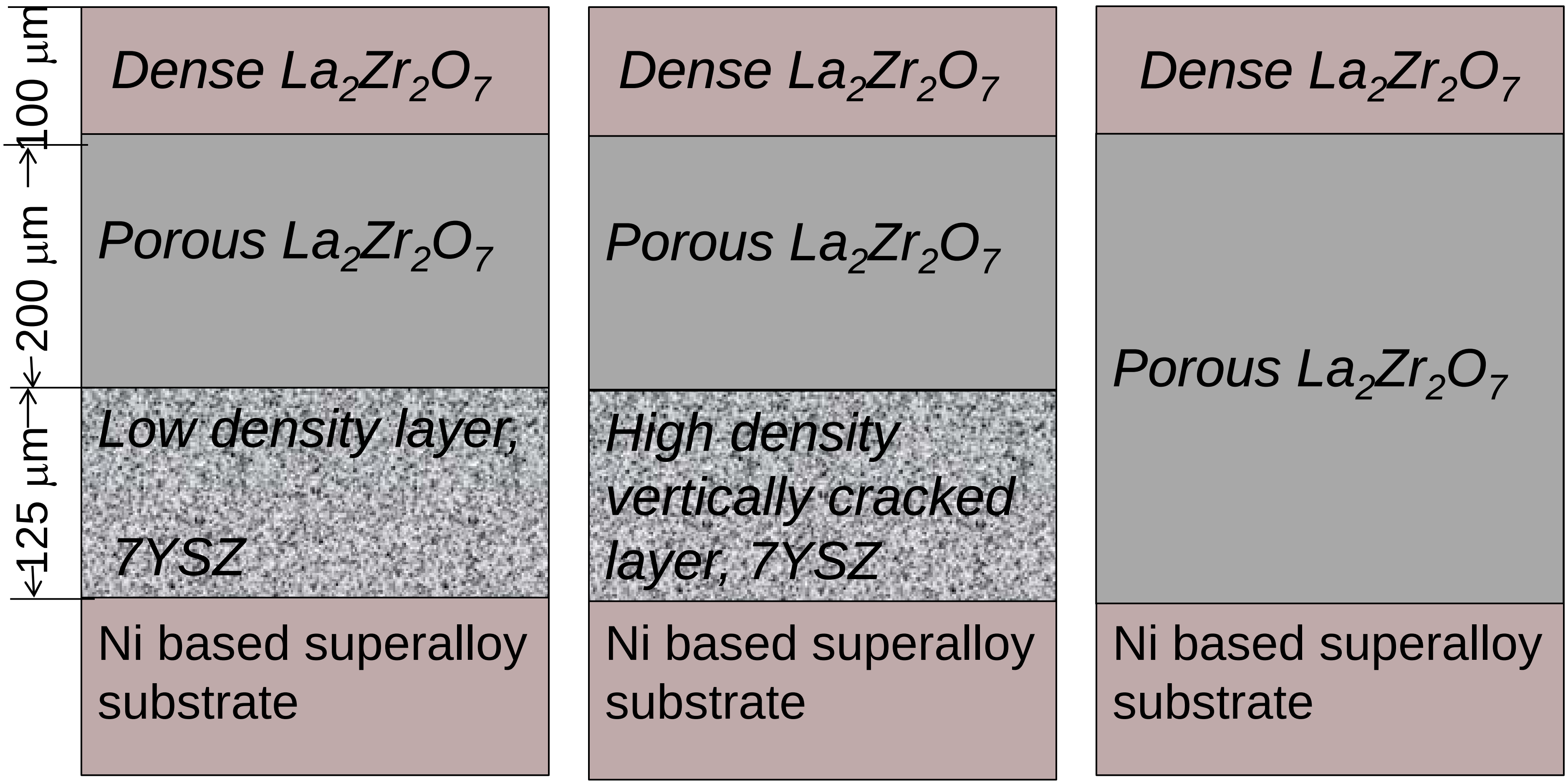
Vertically Cracked Intermediate Layer

- Intermediate layer (e.g., Zircoat™) between topcoat and bondcoat, characterized by the intentional vertical cracks, provides improved tolerance to the strain caused by the CTE mismatch
- The Intermediate layer will be applied as a (1) low-density or (2) dense vertically cracked structure.



Cross-section of APS Zircoat™ containing approximately 16-24 cracks per linear centimeter. (Feuerstein, Hitchman, Taylor, Lemen, Process and Equipment for Advanced Thermal Barrier Coatings, in *Advanced Ceramic Coatings and Interfaces III* (2009)

Design of Layered TBCs



(1) (2) (3)

Air Plasma Spray (APS)

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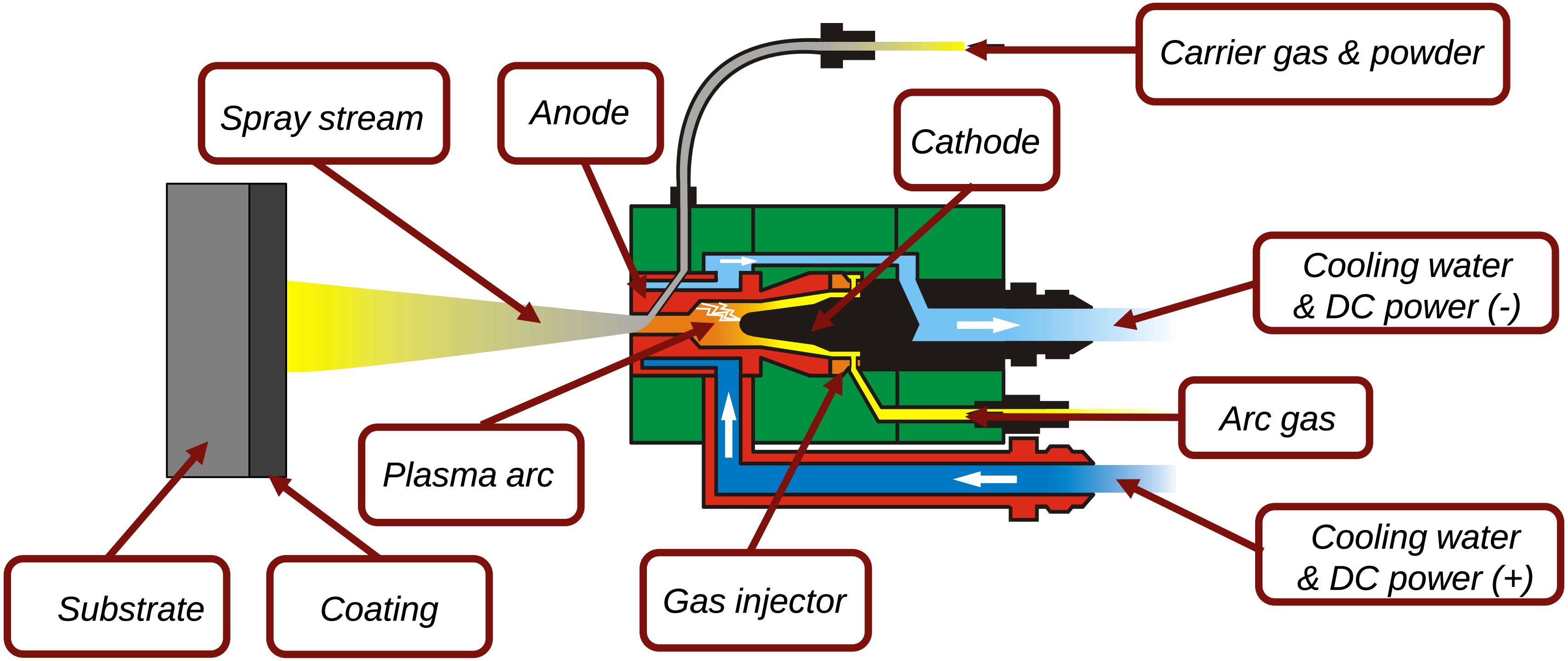
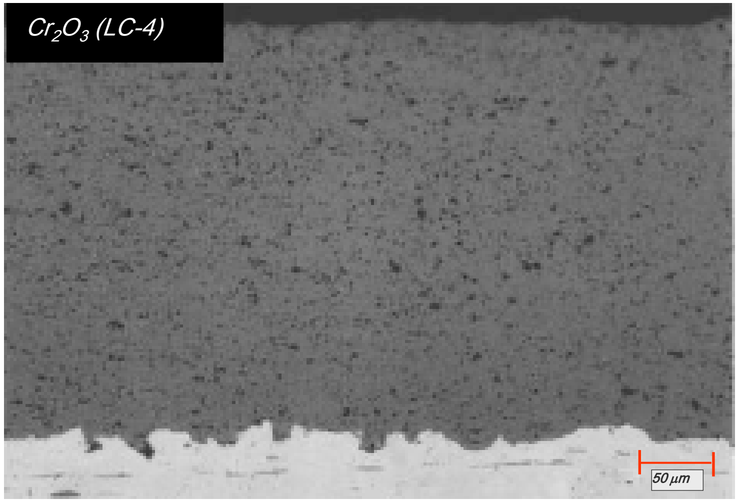
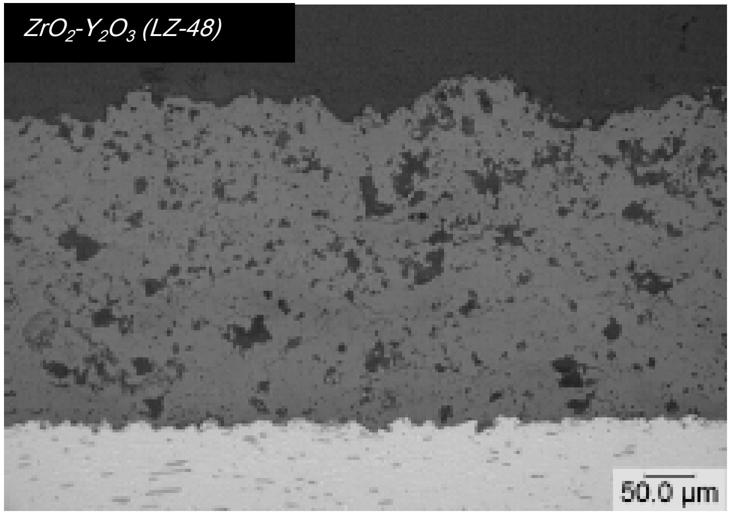


- Non-transferred electric arc excites a gas mixture generating a rapid expanding ionized stream
- High temperature at lower velocity coating process. The most versatile process capable of spraying any material that does not sublime.
- Mostly used for spraying ceramic materials
- ID coating capable down to approximately 3”
- PST shrouded plasma produces metallic coatings with very low oxide

Process Control Parameters

Typical Coating Microstructure





- Torch build
  - Powder dwell time and velocity
- Torch gas
  - Particle velocity
- Powder carrier
  - Particle melting
- Shield gas
  - Coating density and oxide
- Auxiliary carrier gas
  - Particle velocity
- Torch current
  - Particle melting
- Standoff
  - Coating density and oxide
- Powder feed rate
  - Unmelted particles
- Powder particle size distribution
  - Deposition efficiency

Plasma Spray Torches

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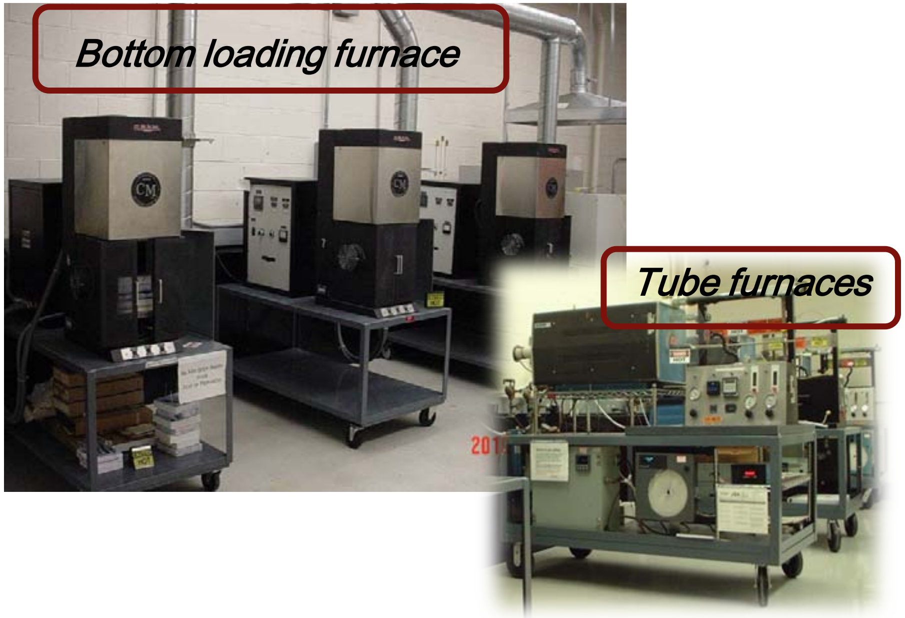
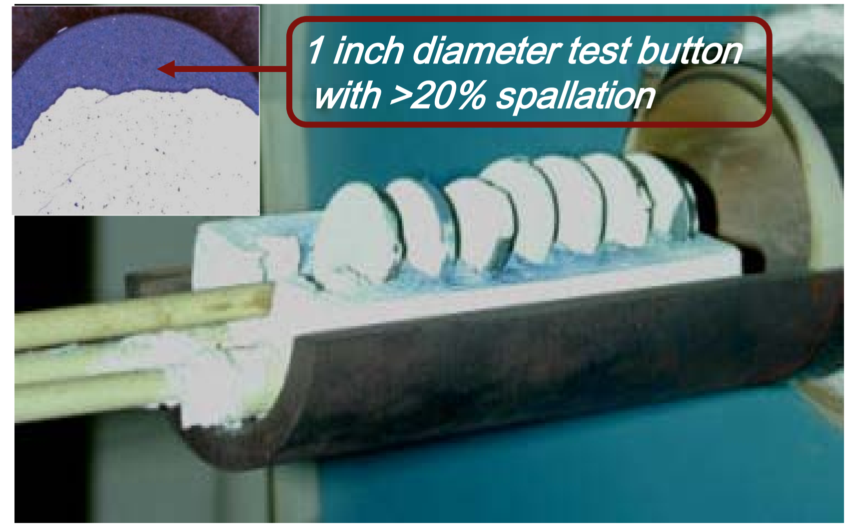
- PST 1108, 1130, 1125
- PST SG100
- TAFA PlasJet
- Metco F4
- Metco 3, 7, 9
- MetTech
- Metco Triplex
- Progressive 100HE

PST 1108

PST 1130

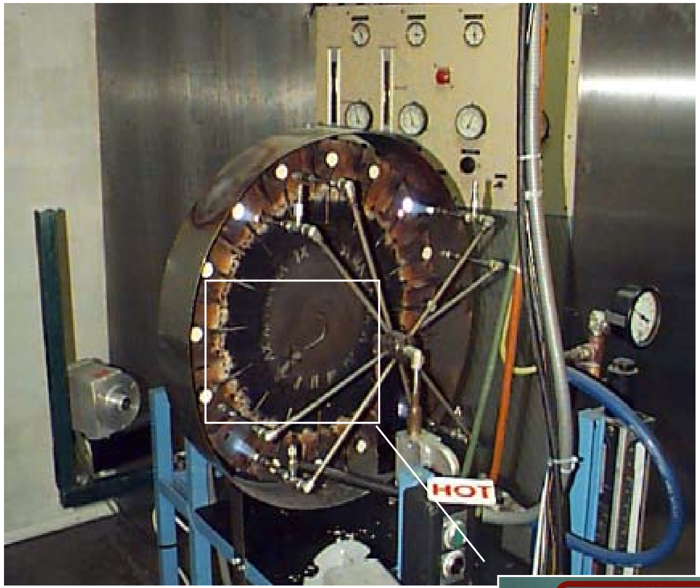
Furnace Cycle Oxidation Testing

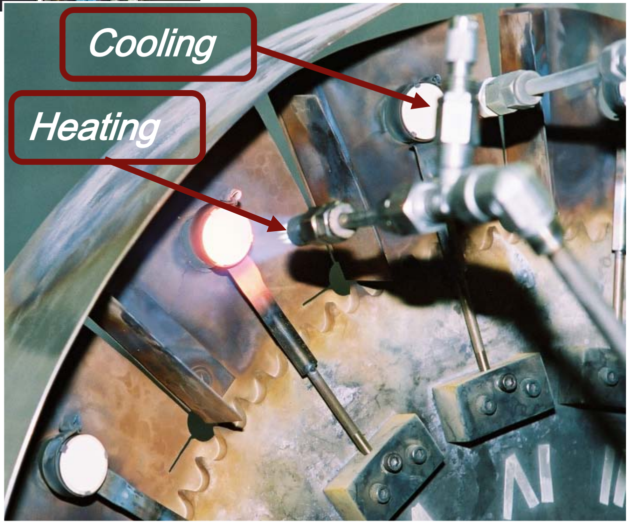




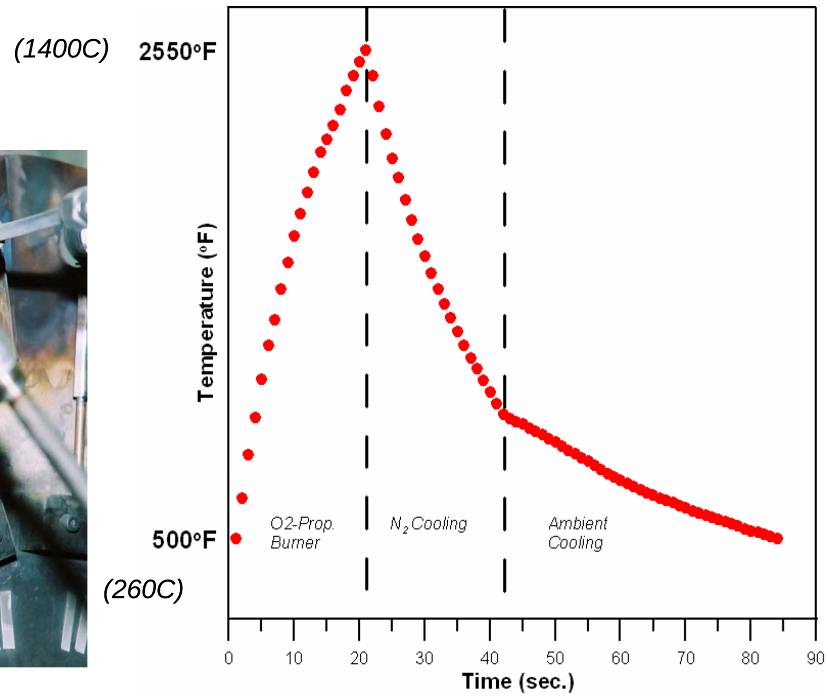
- Furnace Cycle Oxidation
  - *Isothermal test of TBC, with periodic excursions to room temperature (e.g. 1135-100°C)(2075-212°F)*
  - *50 Min at Temp – Air Quench 10 min*
  - *CM bottom-loading furnaces*

Thermal Shock Testing





- JETS (*Jet Engine Thermal Simulation*)
  - Thermal gradient test of TBC
  - Ceramic layer durability assessment
  - Measured by percentage of cracking on specimen edge



Summary

- PST has successfully developed a unique manufacturing process to scale up the production of high-purity, large-quantity lanthanum zirconate powders (>150 lb) in one single batch
- The powders morphology and chemistry are characterized
- Double-layer coating systems are designed

Future Work

- Fabricate the double-layer TBC systems
- Characterize and evaluate the TBC materials and their corrosion resistances at elevated temperatures and in corrosive environments