Application of Ceria Layers to Increase Low-Temperature SOFC Power Density

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Abstract

The main aim of this project has been to achieve solid oxide fuel cells (SOFCs) that can provide high power densities at relatively low temperatures; i.e., down to 600 °C or lower. In order to achieve this, we have developed thin YSZ and YSZ/YDC electrolyte SOFCs and have used interfacial layers to reduce interfacial resistances.

The interfacial resistance of Ni-Y_2O_3-stabilized ZrO_2 (Ni-YSZ) anodes have been reduced by inserting mixed-conducting layers of TiO_2-doped YSZ (YZT) or Y_2O_3-doped CeO_2 (YDC) between the Ni-YSZ and YSZ electrolytes.

The effects of interlayer type, thickness, and YSZ surface condition on the interfacial resistance were investigated using Impedance Spectroscopy at temperatures ranging from 600 to 750 °C and three differenct Ps (H_2). Two arcs, denoted as high frequency arc (HFA) and low frequency arc (LFA), were normally found in the impedance spectra. The HFA, which has been associated with the charge transfer process, was reduced by the addition of either interlayer.

The decrease in the size of HFA apparently results from the large reaction area within the porous interlayers. The LFA, which is believed to be associated with mass transport processes, decreased with increasing YSZ surface roughness, but increased with increasing interlayer thickness. The LFA was associated with a thermally activated process with an activation energy of ≈ 0.45 eV for Ni-YSZ/YDC, and ≈ 0.72 eV for Ni-YSZ/YZT. Under 97 percent H_2 + H_2O, the lowest interfacial resistances of Ni-YSZ with a 0.5μm thick YDC interfacial layer were 0.13 Ω-cm^2 at 750 °C and 0.29 Ω-cm^2 at 600 °C.

SOFCs with electrolytes that provide high open-circuit voltage (OCV) and low ohmic loss down to 550 °C are described. The electrolytes were bi-layers consisting of a 4 to 8 μm thick Y-doped ceria (YDC) layer with a 1. to 1.5 μm thick Y-doped zirconia (YSZ) layer on the fuel side. The cathode/supports were $La_{O.85}Sr_{0.15}MnO_3$-YSZ. The anodes consisted of thin YDC and Ni-YSZ layers. The YDC/YSZ electrolyte SOFCs yielded 85 to 98 percent of the theoretical OCV, compared with ≈50 percent for YDC electrolyte SOFCs. The cathode overpotential, which was a main factor limiting SOFC power density, was lower for YDC/YSZ than YSZ electrolytes. The maximum power density at 600 °C, 210 mW/cm^2, is higher than previously reported SOFCs.

SOFC power densities typically drop rapidly as the operating temperature is decreased, because of electrolyte ohmic losses and/or electrode overpotentials. In this paper, we describe fuel cells utilizing 8 μm-thick yttria-stabilized zirconia (YSZ) electrolytes to provide low ohmic loss. Adding thin porous yttria-doped ceria (YDC) layers on either side of the YSZ yielded much reduced interfacial resistances at both the LSM cathodes and Ni-YSZ anodes. The cells provide higher power densities than previously reported at below 700 °C, e.g., 300 and 480 mW/cm^2 at 600 and 650 °C, respectively (measured in 97 % H_2 + 3 % H_2O and air), and also provide high power densities at higher temperatures, e.g., 760 mW/cm^2 at 750 °C.

Increased Solid Oxide Fuel Cell Power Density Using Interfacial Ceria Layers

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Introduction

- SOFC power density is reduced at low temperatures due to:
  - electrode overpotentials and/or
  - electrolyte ohmic losses
- Improve low-T performance using Yttria Doped Ceria (YDC)
- Low electrolyte loss using thin YSZ-YDC electrolytes
- Low overpotentials obtained by using:
  - LSM/YDC cathode interface and
  - Ni-YSZ/YDC anode interface
- Can low-T SOFCs internally reform hydrocarbons?
  - demonstrate low-T SOFC operation on 97% methane
  - no C deposition

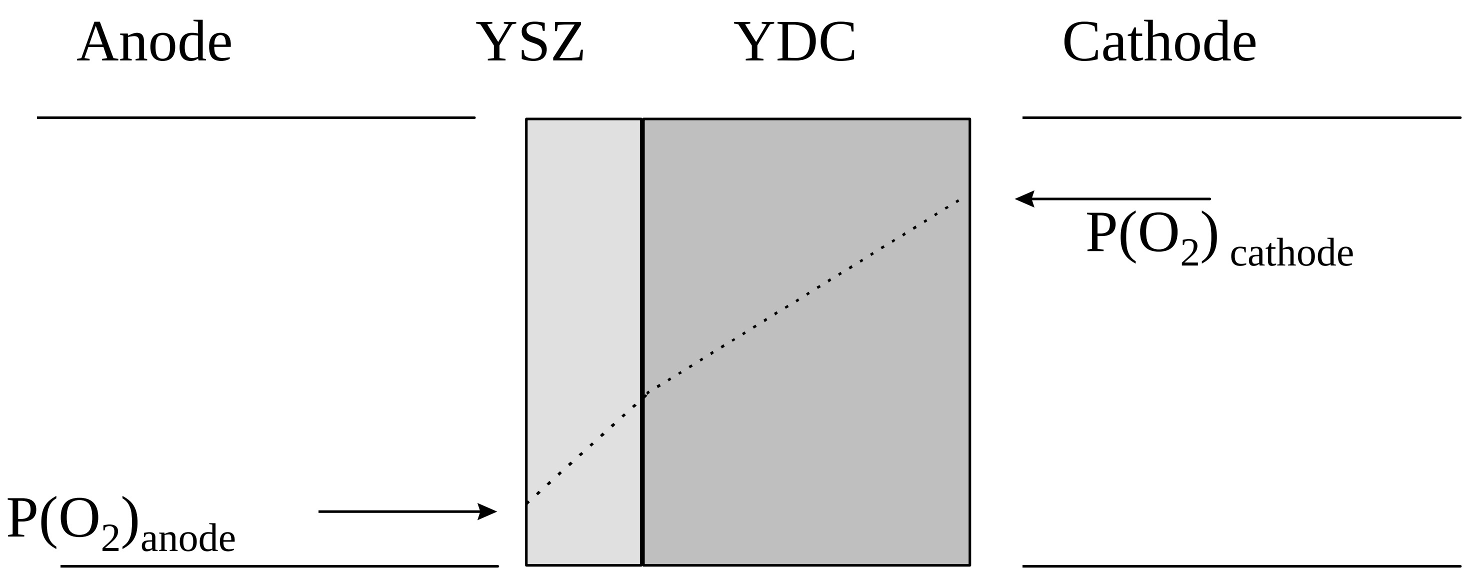
Experimental

- LSM cathodes by pressing and sintering
- Reactive sputtering from metal targets:
  - dense YSZ and YDC electrolyte layers using DC substrate bias
  - porous YDC interfacial layers and Ni-YSZ anodes
- Standard single-cell tests and impedance spectroscopy
  - T = 550 - 800^{o}C
  - cathode: air
  - anode: 97% H_2 + 3% H_2O
    97%CH_4 + 3% H_2O

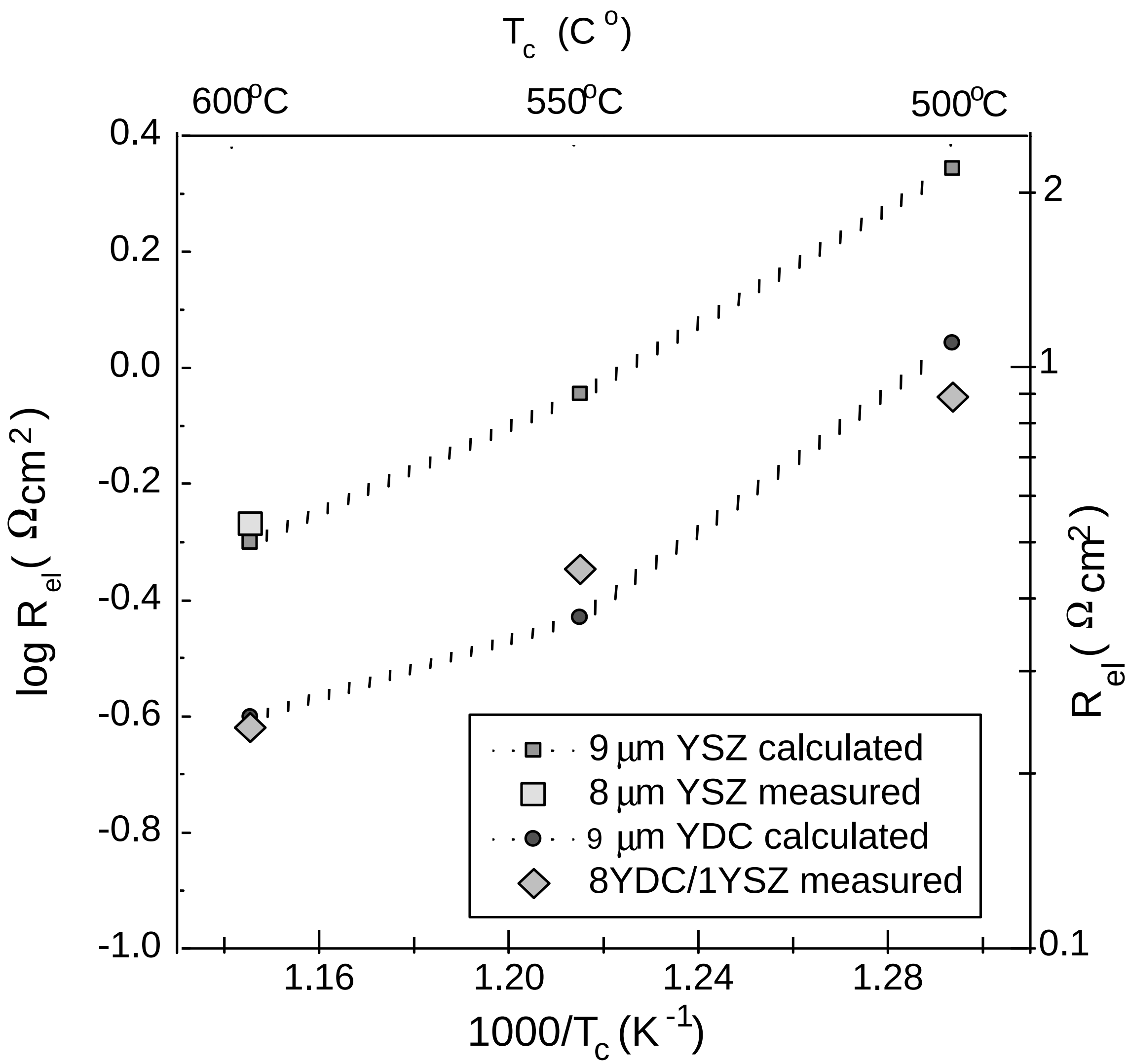
Decrease electrolyte resistance using YSZ-YDC bi-layer

Thin anode-side YSZ layer:

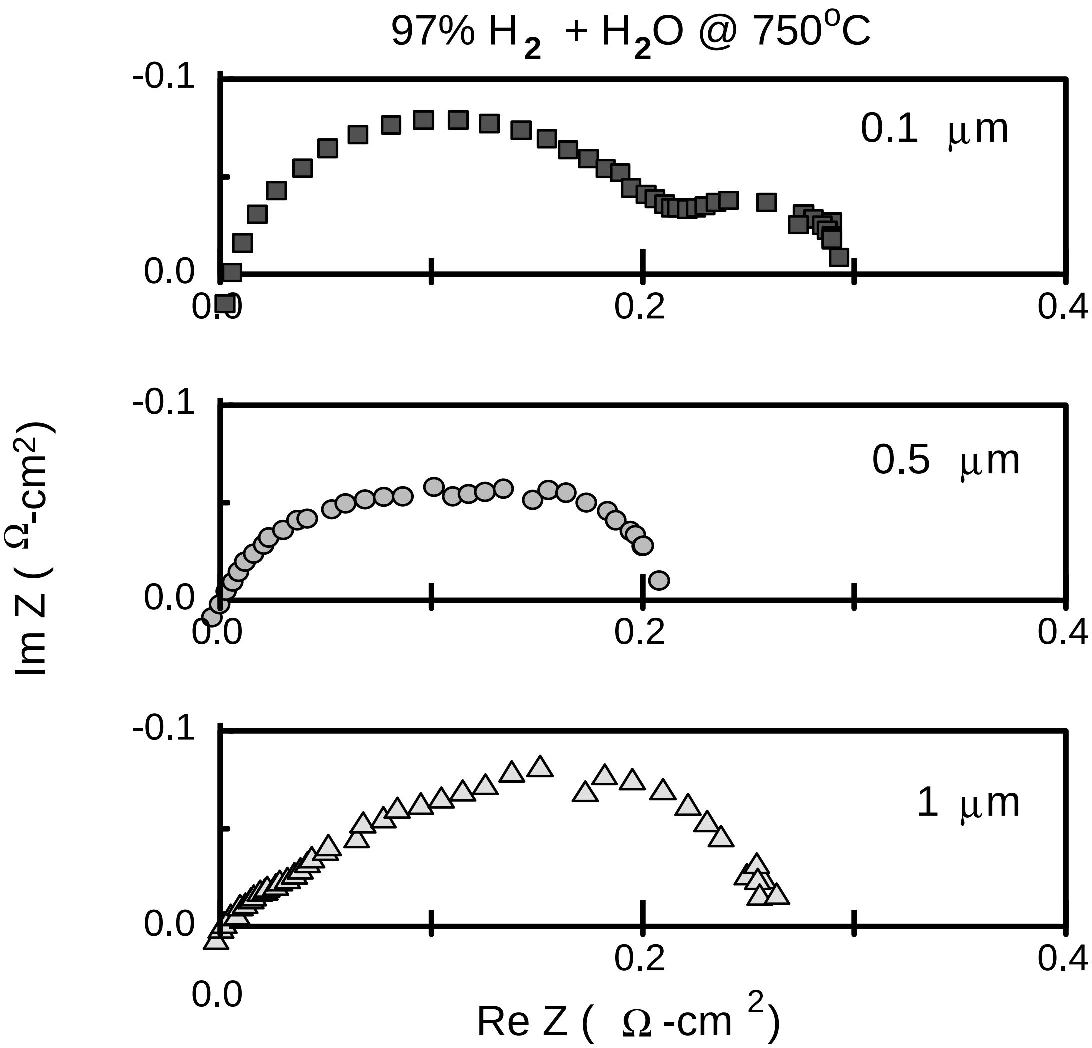
- protects YDC from reduction
- blocks electron current flow



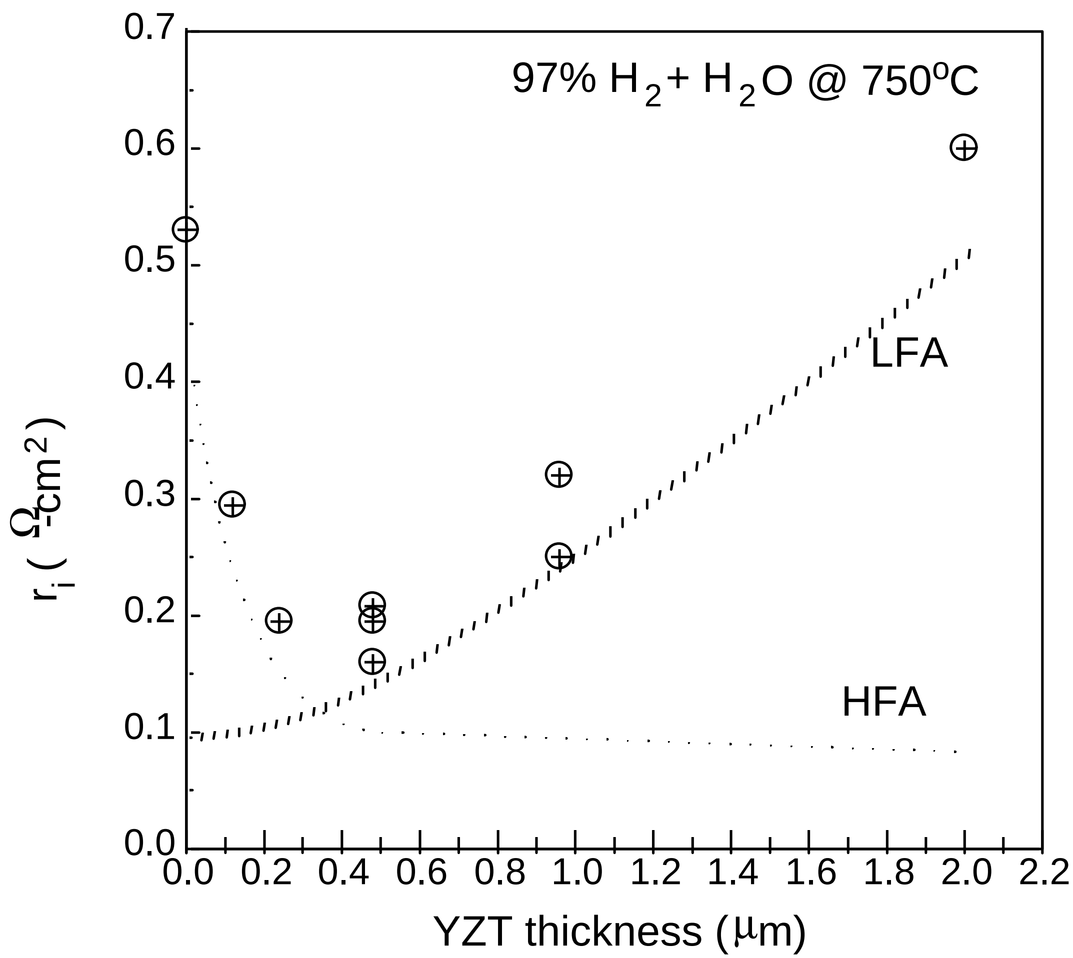
Electrolyte Resistance: Measured vs. Predicted



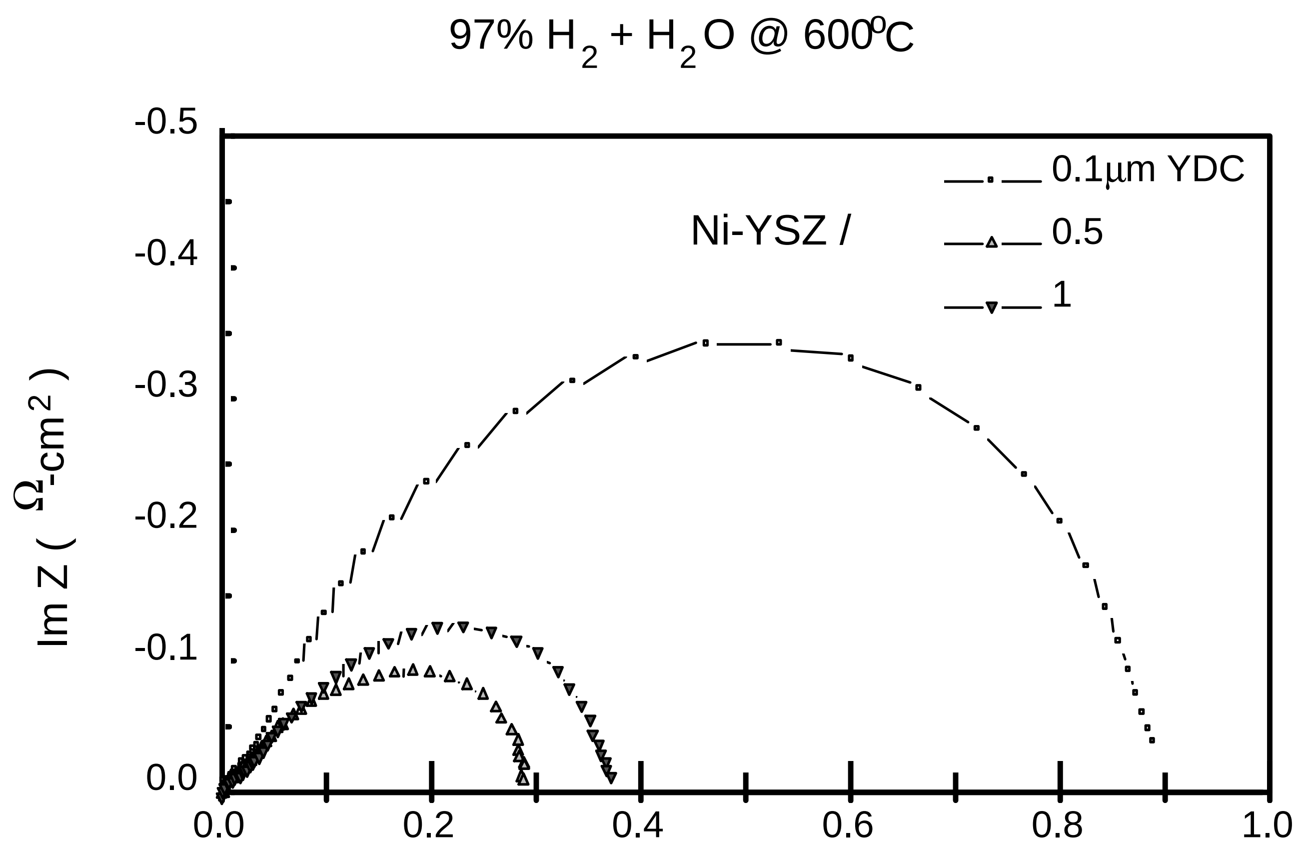
YZT Thickness Effect



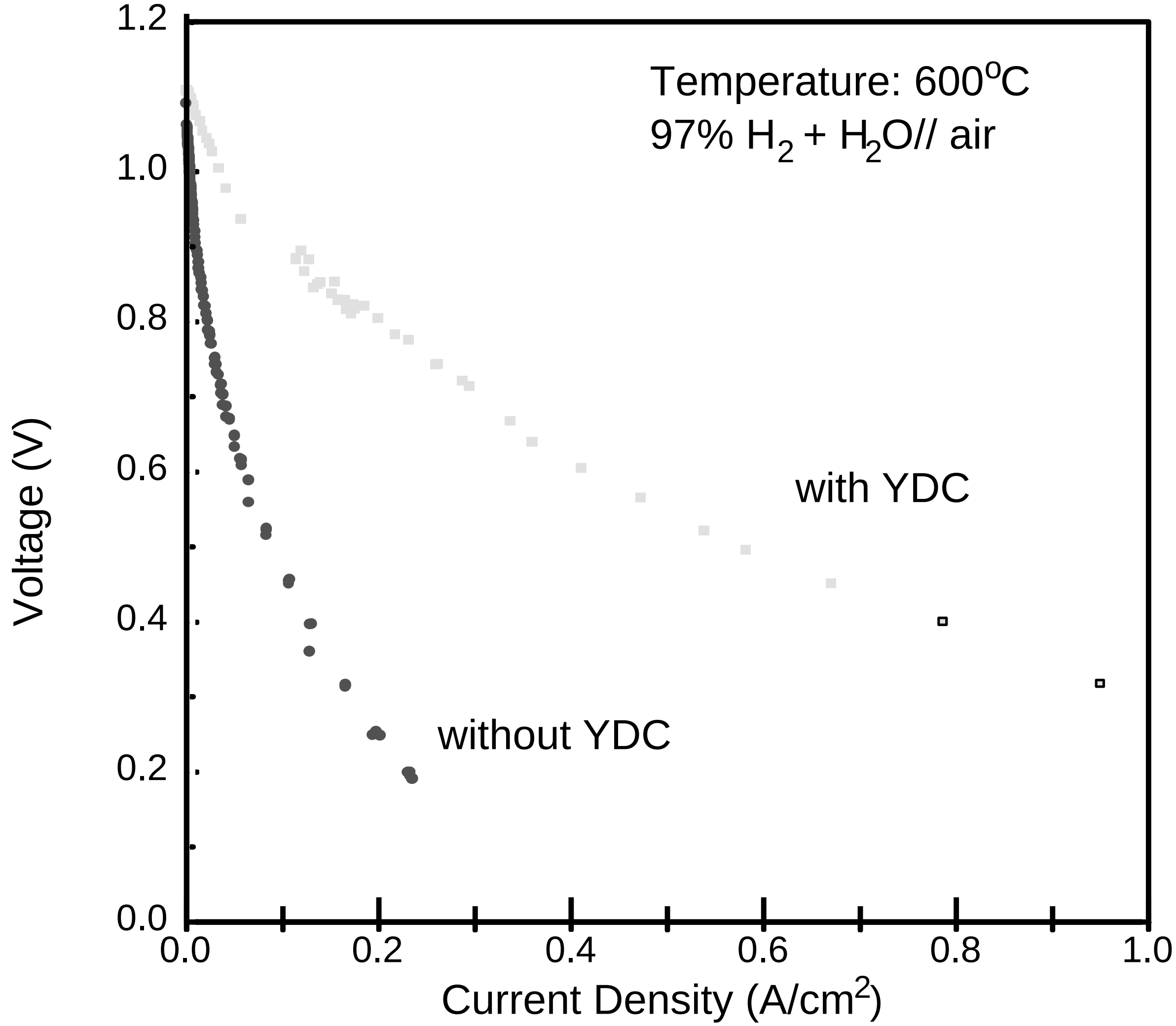
YZT Thickness Effect



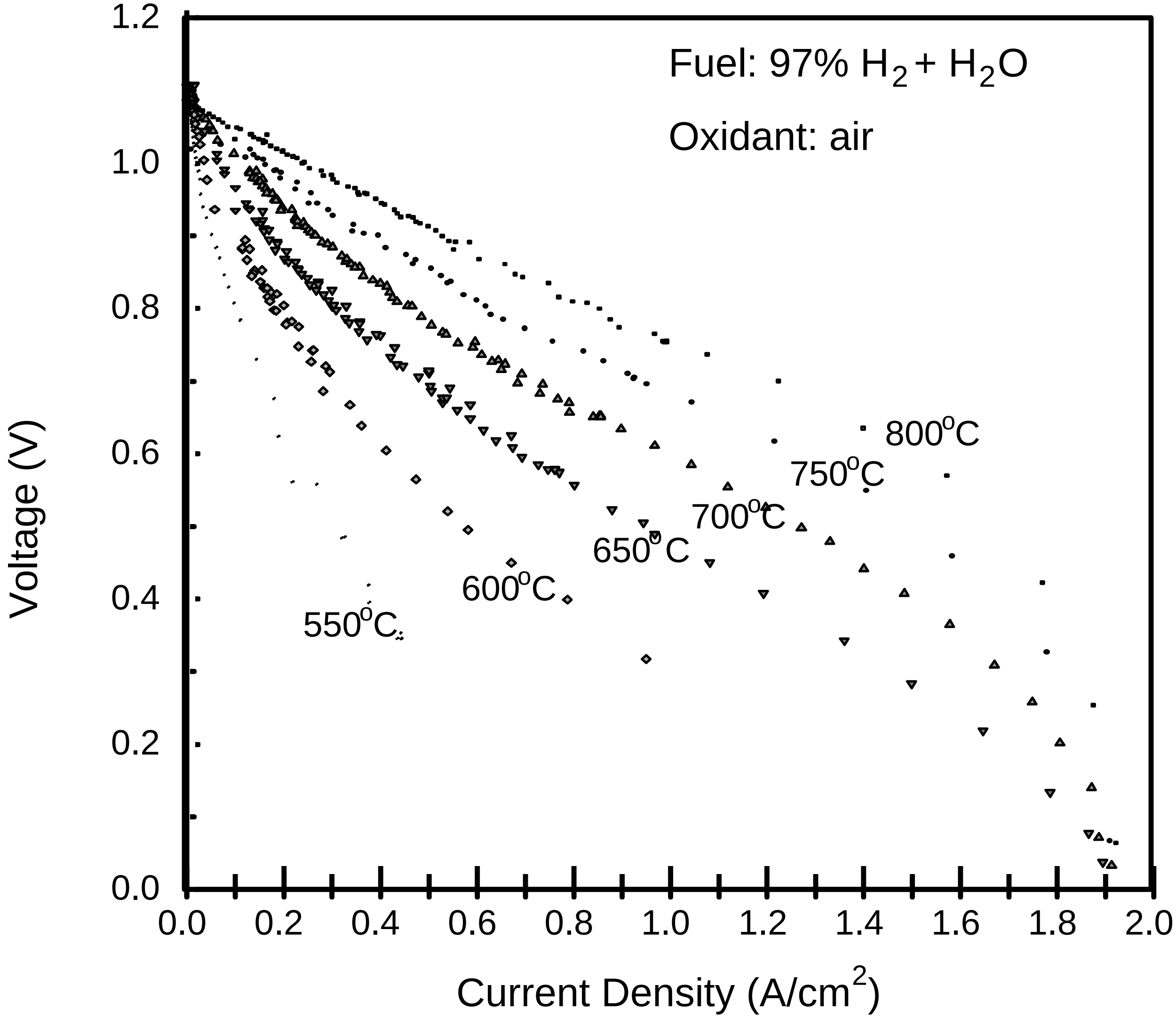
YDC Thickness Effect on Ni-YSZ anode



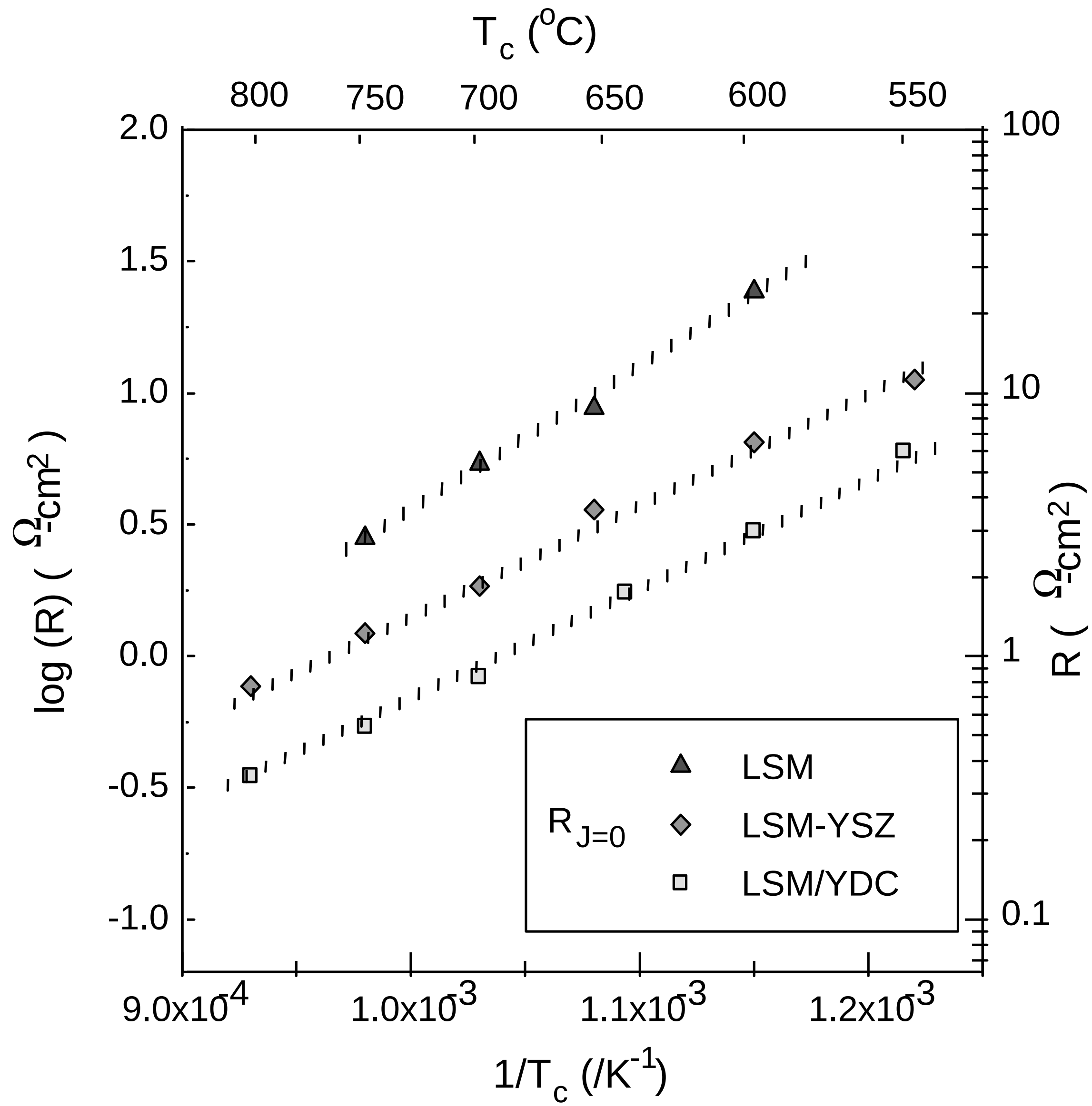
Cell Performance:
LSM/YSZ/YDC/Ni-YSZ
LSM/YDC/YSZ/YDC/Ni-YSZ



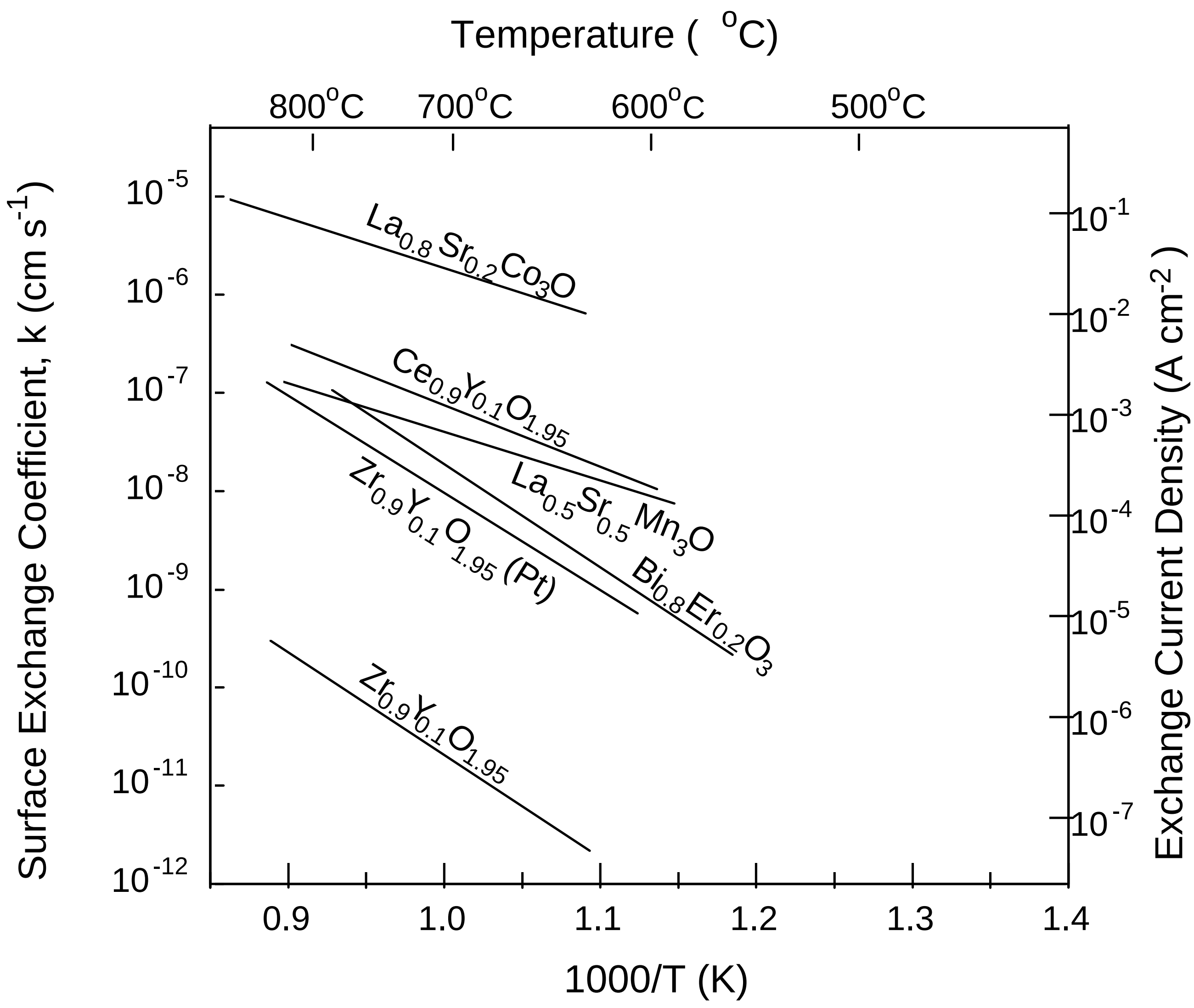
Cell Performance: LSM/YDC/YSZ/YDC/Ni-YSZ



Cell Resistance
Cathode/YSZ/YDC/Ni-YSZ

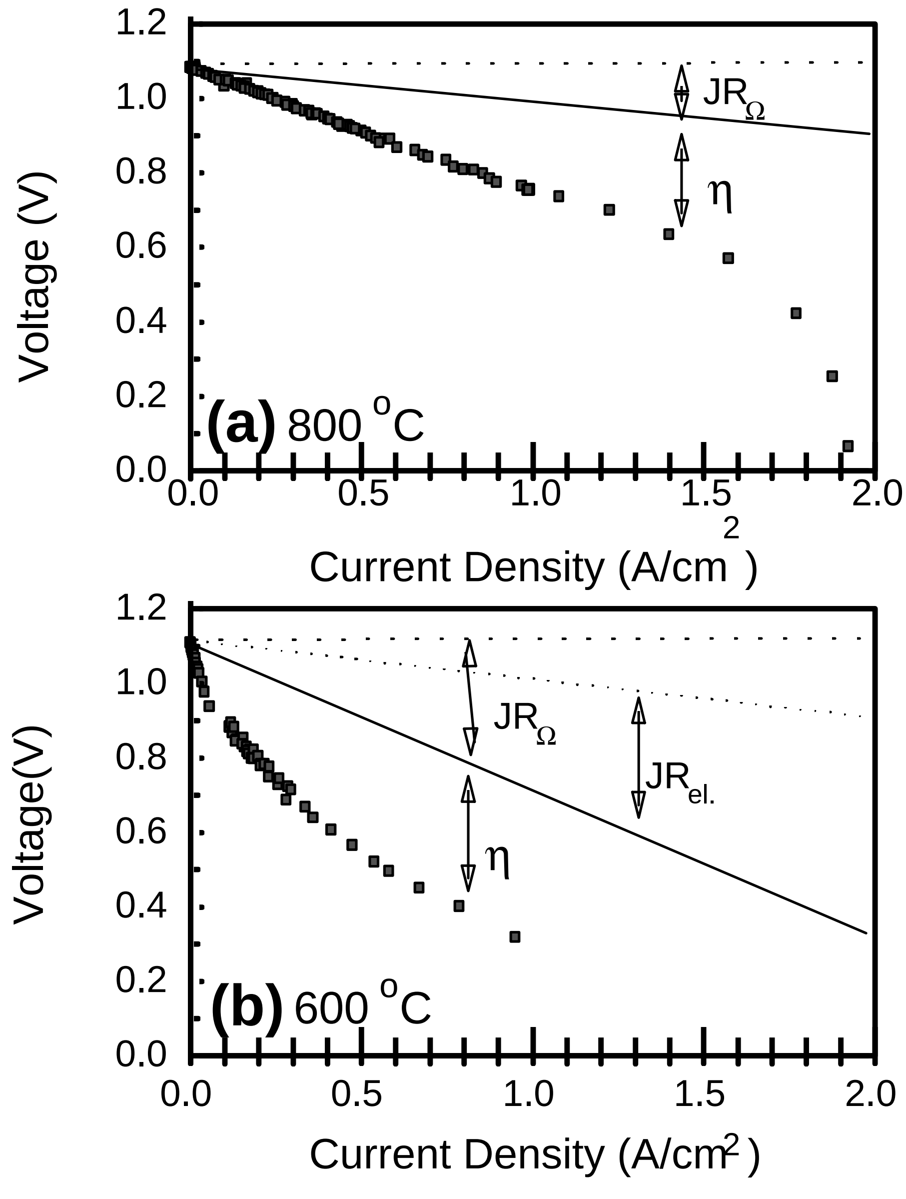


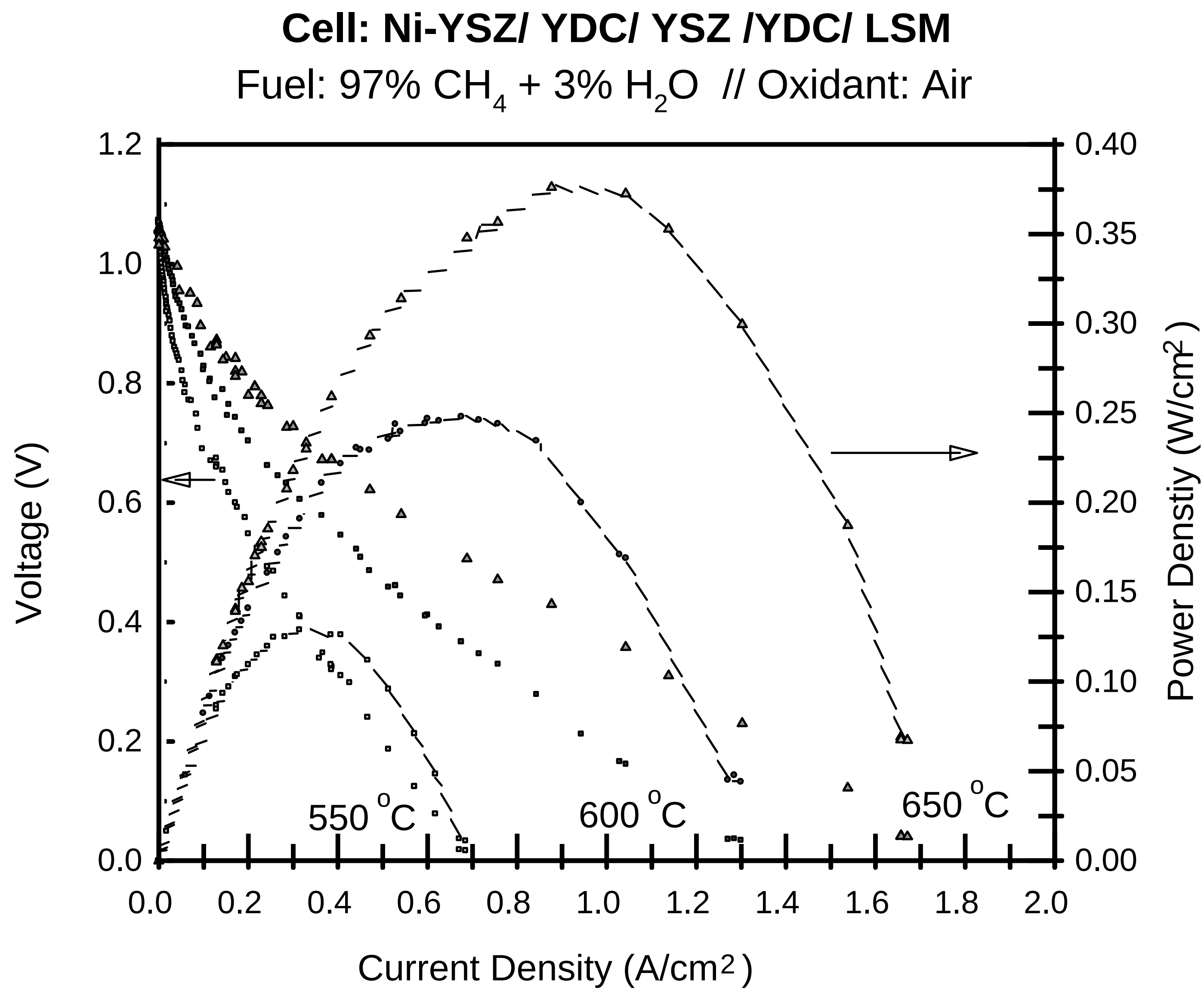
Oxygen Surface Exchange Coefficient



* B.C.H. Steele, *Solid State Ionics*, **75** (1995) 175.

Loss Analysis for 600 and 800ºC



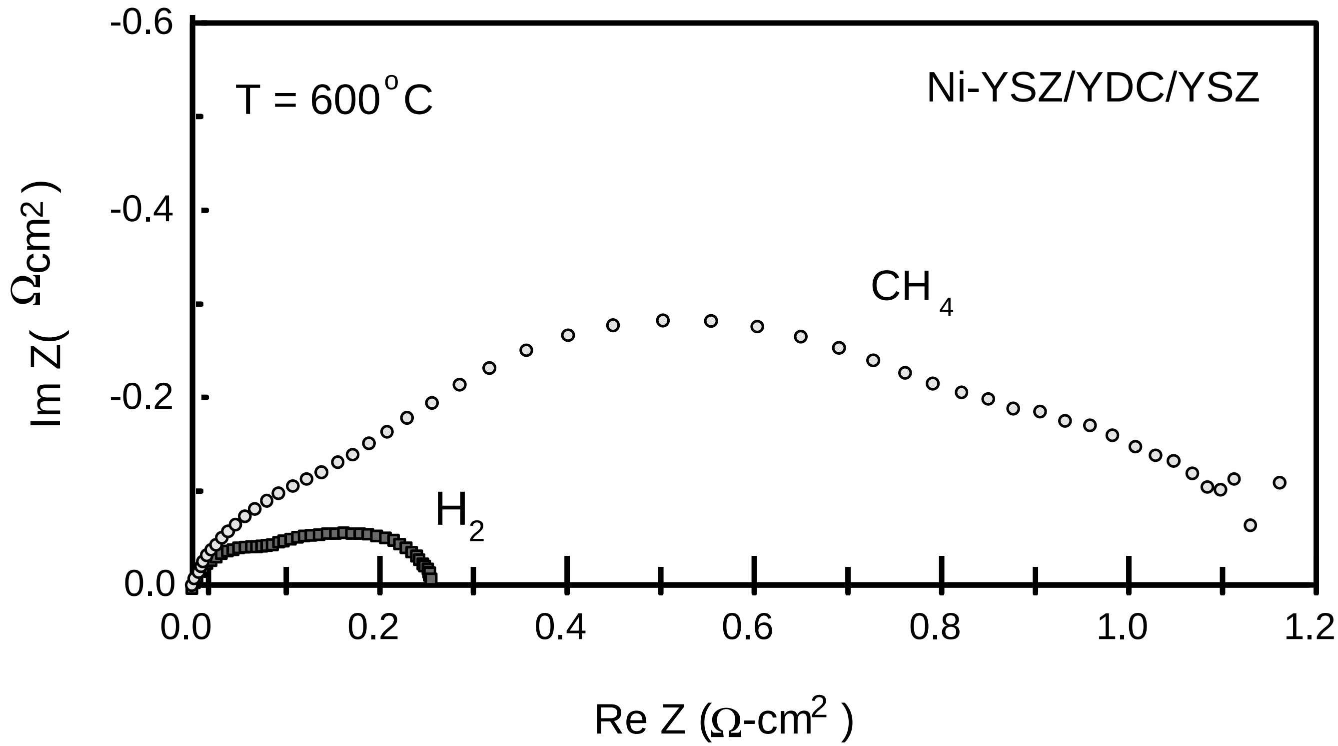


Cell: Ni-YSZ/ YDC/ YSZ /YDC/ LSM
Fuel: 97% CH4 + 3% H2O // Oxidant: Air
Voltage (V)
Power Denstiy (W/cm2)
Current Density (A/cm2)
550 oC
600 oC
650 oC
1.2
1.0
0.8
0.6
0.4
0.2
0.0
0.40
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.00
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0

Anode Performance with Hydrogen and Methane

Gases: 97% Hydrogen + 3% Steam

97% Methane + 3% Steam



Conclusions

- Low-T electrolyte resistance reduced using YSZ-YDC bi-layers.
- On the anode side, YDC enhances anode performance via mixed conductivity and/or increased redox reaction rate.
- On the cathode side, the YDC apparently provided a higher oxygen surface exchange coefficient.
- Low-T SOFC performance:
  - $600^{o}C$ 300 mW/cm^2
  - $700^{o}C$ 630 mW/cm^2
  - $800^{o}C$ 870 mW/cm^2
- Good single-cell performance without carbon deposition with methane fuel