Characterization of SOFC Cathode Impedance under Polarization Using Appropriate Counter Electrode Design

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Abstract:

Electrochemical impedance spectroscopy (EIS) is an important diagnostic tool for evaluation of a solid oxide fuel cell's performance. However, the impedance spectrum of a typical SOFC convolutes anode and cathode behavior, even when using reference electrodes. Symmetrical half cells provide impedance behavior unambiguously assignable to the electrode, but only at open circuit voltage (OCV). The exact assignment of impedance features to the cathode under working condition (cathodic polarization) remains a challenge. This work demonstrates the operation of LSM/YSZ/LSCF cells operated under load in air at 800°C. The LSCF counter electrode, deposited on the YSZ substrate by spray pyrolysis, exhibits a small polarization resistance typically on the order of 0.1 ohm cm^2. By taking advantage of the low polarization resistance and high peak frequency of the counter electrode, the impedance behavior of the LSM working electrode can be characterized over a range of different polarizations. This technique provides a reliable platform to study the activation and degradation mechanisms of SOFC cathode materials in a well-controlled system with minimum interference from the counter electrode.

Characteristics of an ideal CTR electrode

- **Well-defined polarization and EIS behavior independent of current density**
- **Ohmic behavior (polarization resistance independent of current)**
- **Polarization resistance smaller than the WKG electrode**
- **Peak frequencies in EIS data separated from peak frequencies of the WKG electrode**
- **Long term stability**
- **Easily fabricated using low sintering temperatures**

Symmetrical LSCF/YSZ/LSCF cell

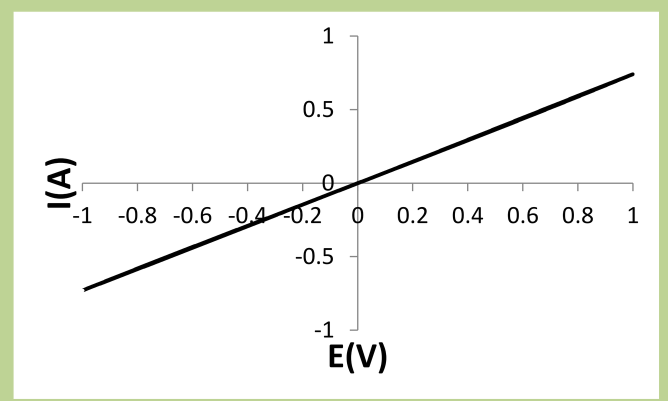

Fig. 1: Cyclic voltammetry at 1000 mV/s illustrating nearly ohmic behavior of a symmetrical cell with LSCF electrodes at 800°C in air.

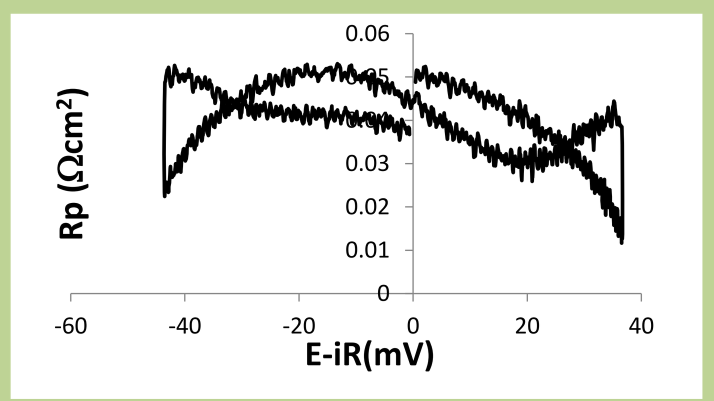

Fig. 2: Polarization resistance per LSCF electrode after correction for ohmic resistance of the YSZ electrolyte.

EIS of LSCF/YSZ/LSCF cell at fixed DC voltages

Positive voltages. Negative voltages.

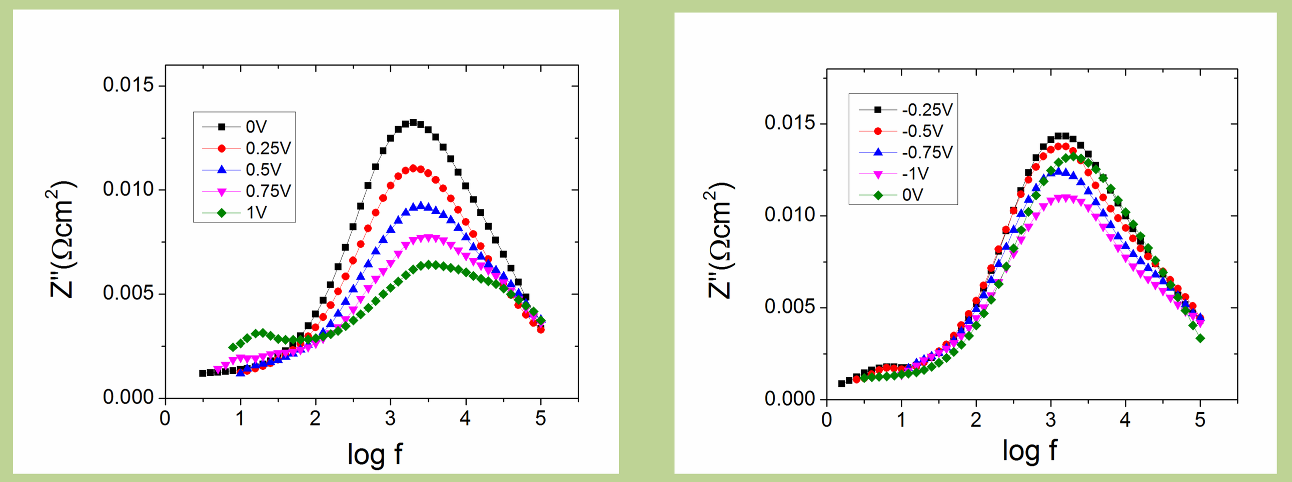

Fig. 3: Bode plots of the LSCF/YSZ/LSCF cell with respect to applied cell voltage after 120 s conditioning at each voltage. The Z" data is corrected for the two electrodes.

Aging effects of LSCF/YSZ/LSCF cell

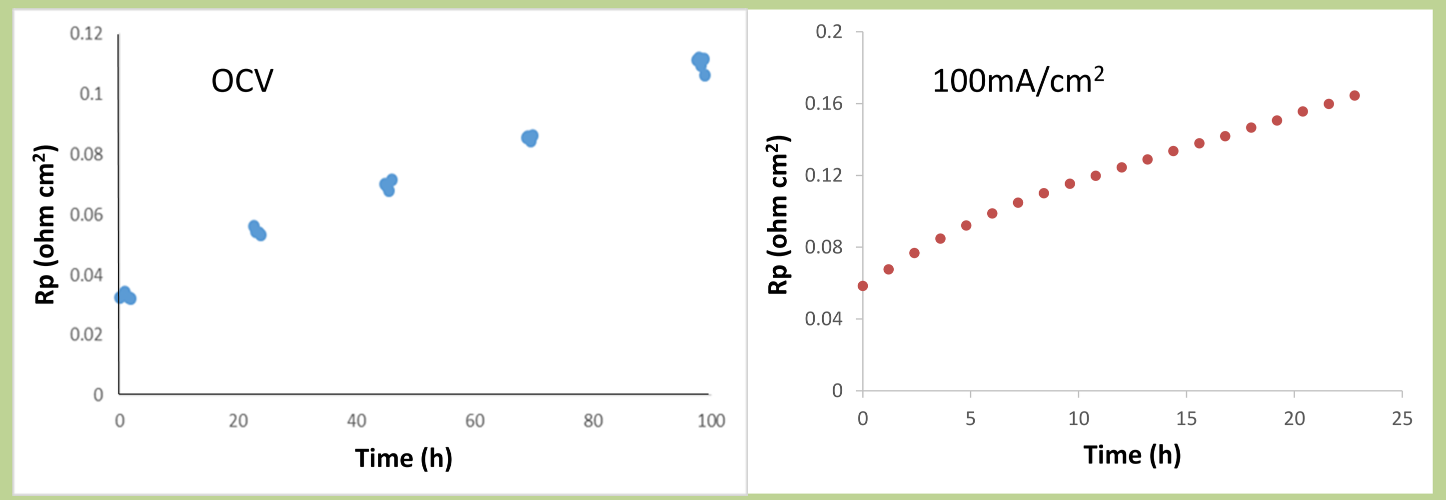

Fig. 4: Over 100 hour in air at 800°C under OCV, the polarization resistance nearly triples. Passing current leads to accelerated degradation.

Applications

1. **Thin, dense sputtered LSM film electrode with LSCF CTR electrode under cathodic polarization**

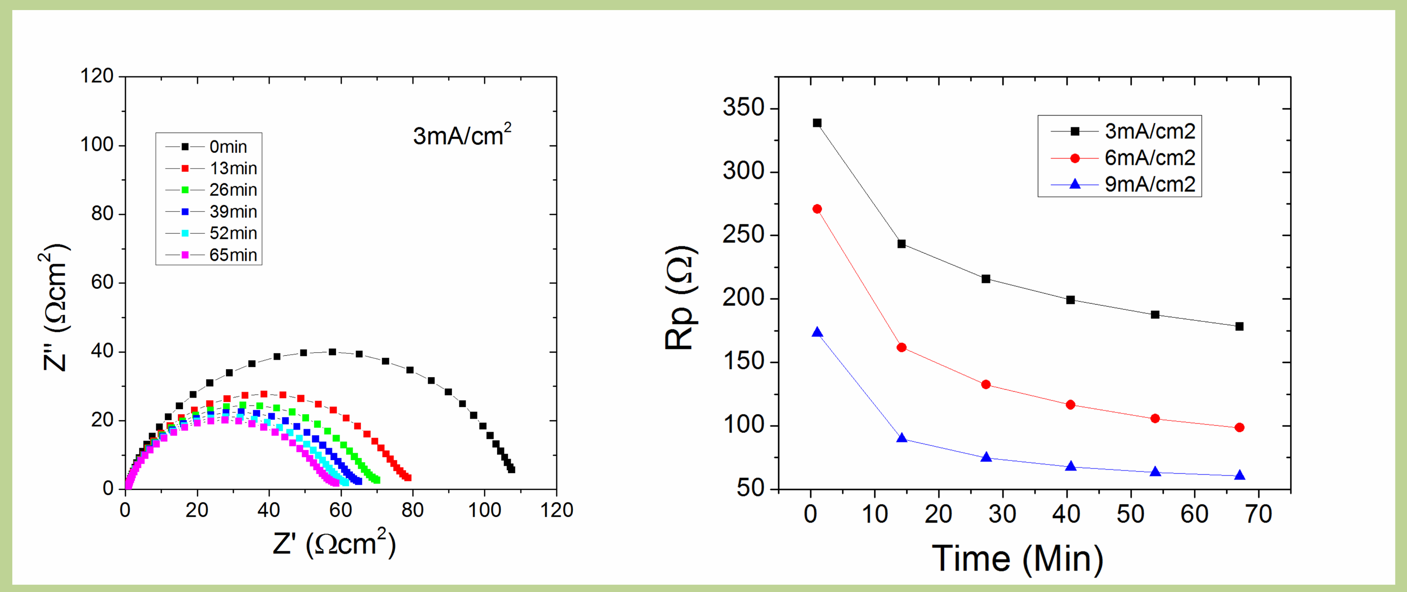

Fig. 5: Under small current (3, 6, 9 mA/cm^2), the polarization resistance of LSM electrode was reduced to 53%, 41% and 36% within 1 hour. The polarization resistance of the LSCF CTR electrode is negligible on this scale.

2. **LSM/YSZ composite electrode with LSCF CTR electrode**

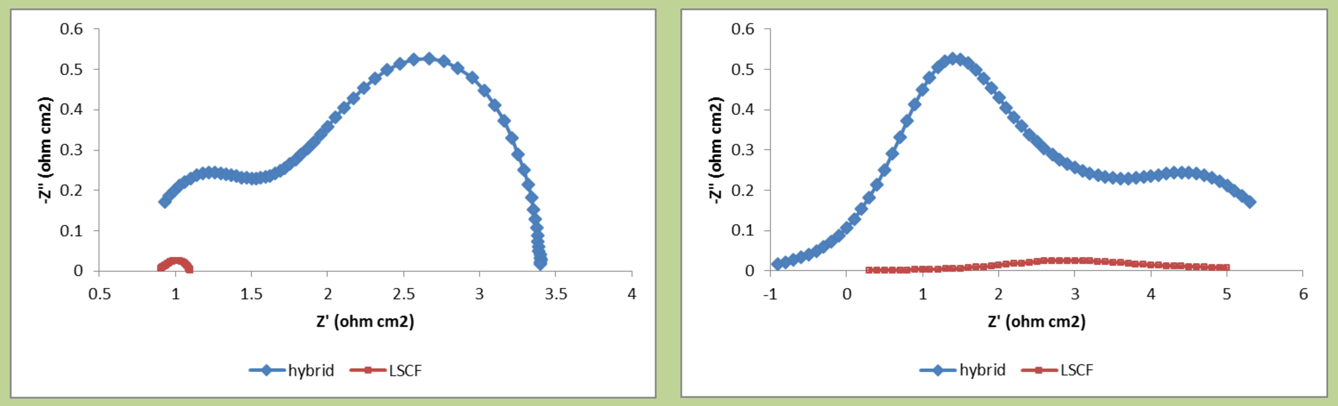

Fig. 6: Nyquist and Bode plots of the cell compared with the LSCF CTR electrode impedance from symmetrical cell studies.

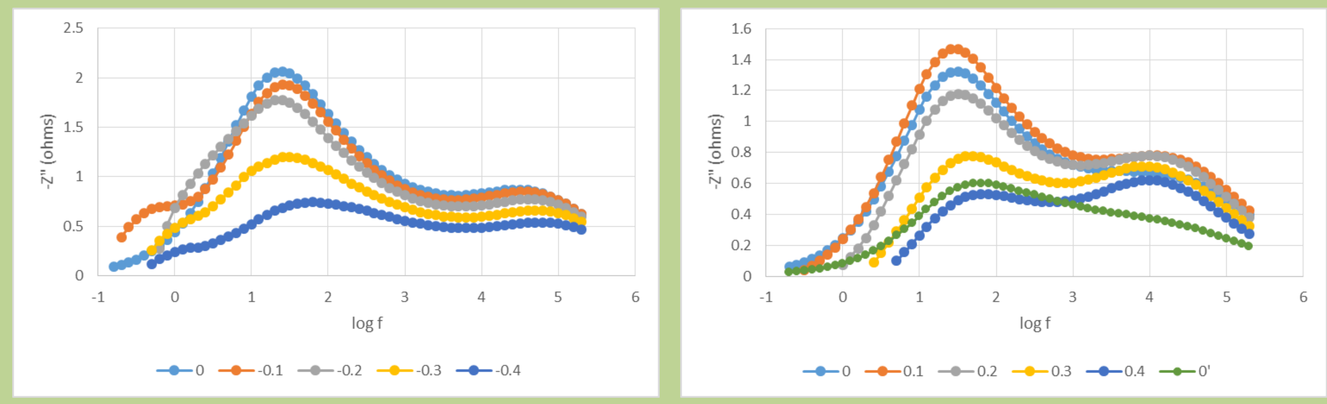

Fig. 7: Bode plots of the cell at constant cell voltage after 120 s at the same cell voltage.

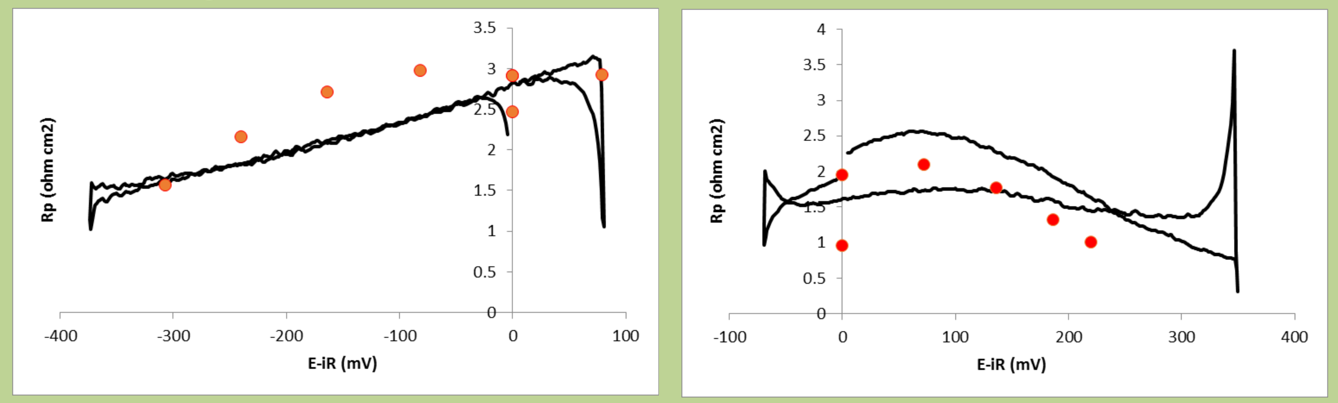

Fig. 8: Comparison of polarization resistances of the LSM/YSZ composite electrode as a function of applied DC voltage. Solid black lines from cyclic voltammograms, red circles from impedance measurements.

Summary:

LSCF counter electrodes formed by spray pyrolysis show many advantages for characterization of polarization behavior of LSM based electrodes:

- **Easily fabricated, low sintering temperature (600°C)**
- **Low polarization resistances at high peak frequencies**
- **Nearly ohmic behavior; R_p nearly constant up to 1 A/cm^2**
- **No current spikes or noise**
- **No evidence for oxygen starvation**
- **Negligible drift in EIS data during data collection**
- **The impedance behavior of two types of LSM electrodes have been mapped as a function of applied voltage.**

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