Improving Durability and Performance of Solid Oxide Electrolyzers by Controlling Surface Composition on Oxygen Electrodes

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Introduction & Goals

Strontium Segregation in SOFC Cathodes:
- Study focuses on state-of-the-art SOEC cathode material (La0.8Sr0.2Co0.8Fe0.2O3−δ (LSCF))
- LSCF exhibits Sr segregation from cathode lattice to cathode surface
- Segregated SrO then reacts with Cr and S to form inactive surface phases

Predominant Sr Segregation Mechanism:
- Under cathodic polarization, an electostatic attraction mechanism drives Sr segregation
- Electrostastic driver: attraction of negatively charged A-site dopant to surface enriched with positively charged oxygen vacancies

Surface Composition Control Methods:

Objective: Minimize Sr segregation and improve cell stability by infiltration of more oxidizable cations on state-of-the-art LSCF electrodes.

Experimental Results

Electrochemical Characterization Suggests Stabilized Cells with Surface Infiltration:
- Infiltrated cells: significant stabilization of Rp. Monotonic increase for as-prepared cells.
- As prepared cells: stable decrease in I vs. t. Infiltrated cells: stable current (Hf infil) or stable initial current that later degrades sharply (Zr infil).

Comparison of surface chemistry and polarization resistance:
- Sr surface/lattice and A/B ratios show no clear trend – results not consistent with infiltration reducing Sr segregation.
- Rp values at end of experiment – again no clear trend.

Temperature dependence of surface infiltrated cations:
- HI 4d signal – evidence of HI dissolusion away from surface at T > 600°C
- Surface Sr 3d – nearly constant up to 700°C, large increase after 800°C cycling.

Computational Results

Grand Canonical Monte Carlo (GCMC) + Density Functional Theory (DFT) to Construct Realistic LSCF Surface Reconstruction

Region 1: Full GCMC on both metal cations and oxygens
Region 2: MC (exchange site position; exchange site cations positions; assume Sr and La still at A site, Co and Fe still at B site) on cations. GCMC on oxygens
Region 3: Fix atoms in positions

CrO3 and SO2 Adsorption Energies on Pristine LSCF slabs
- CrO3 and SO2 adsorption energies show the same trend with configurations. The surface oxygen sublattices are the most preferential deposition sites.
- Adsorption energies decrease as the number of bonds formed from the adsorption increase
- BO6-terminated slabs have higher (yet still negative) adsorption energies.

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