

Understanding Oxygen Surface Exchange Kinetics in Solid Oxide Cell Electrodes via *In Situ* Optical Transmission Relaxation: Sr(Ti,Fe)O₃ Case Study

Ting Chen^{1,2}, George F. Harrington^{1,3,4}, Kazunari Sasaki^{1,2,3} and Nicola H. Perry^{1,4*}

¹I2CNER; ²Department of Hydrogen Energy Systems; ³NEXT-FC; ⁴Massachusetts Institute of Technology

Reversible solid oxide cells (R-SOCs: Solid Oxide Fuel Cells (SOFCs) and Solid Oxide Electrolysis Cells (SOECs)) are promising energy conversion devices for both power generation (direct conversion of chemical fuels to electricity) and energy storage (H₂ or syngas production), but their efficiency and stability need to be further improved by increasing and stabilizing the surface exchange coefficient (k_{chem}) at the oxygen electrode. k_{chem} reflects the ease with which oxygen is exchanged between the gas and solid phase at the electrode surface.

In this talk, I would like to introduce an *in-situ* optical transmission relaxation (OTR) approach enabling non-contact and continuous measurements of the oxygen surface exchange kinetics. We have applied OTR to investigate the relationship between processing, electrode structure (grain size, crystallinity), and k_{chem} . We employed SrTi_{0.65}Fe_{0.35}O_{3- δ} (STF35) thin films as a model system due their rapid oxygen surface exchange kinetics and the ability to systematically vary microstructure in this geometry.

A series of STF35 thin films with varying grain boundary density were grown by pulsed laser deposition under different conditions. The impact of growth temperature on crystalline quality, orientation, grain size, surface roughness and surface chemistry was evaluated by X-ray diffraction, scanning probe microscopy, transmission electron microscopy, and angle-resolved X-ray photoelectron spectroscopy (AR-XPS).

We found that: I) Amorphous thin films did not exhibit visible oxygen exchange behavior, but highly crystalline films showed fast k_{chem} . II) Neither grain boundaries nor crystallographic orientation cause observable changes in surface exchange kinetics in this composition. III) AR-XPS results showed greater surface Sr concentrations in as-deposited films grown at higher temperatures. Combined with optical studies showing optimal k_{chem} for intermediate film growth temperatures, these results suggest that rapid k_{chem} requires both crystallinity and low Sr surface concentration. Such results might guide ways to optimize electrode processing for efficient performance.