Electrochemical Study of (La0.6Sr0.4)0.99CoO3-δ Thin Film Microelectrodes

Solid Oxide Fuel/Electrolysis Cells (SOFC/SOEC, collectively termed SOC) are one of the most promising reversible energy conversion/storage technologies. Long term durability is required for such devices to become economically feasible. One approach to make SOCs more durable and at a lower cost is to decrease the operation temperature. However, lowering the operation temperatures of SOCs has shown to be challenging due to the difficulty in finding suitable oxygen electrodes which have high catalytic activity for oxygen reduction and fast ionic transport. (La,Sr)CoO3-δ (LSC) is one of the promising cathode materials due to its high electronic and ionic conductivity as well as good catalytic activity for oxygen reduction at intermediate temperatures (500-700 °C). However, LSC is characterized by low chemical stability. Multiple degradation mechanisms are reported for LSC such as zirconate formation due to reactivity with YSZ (electrolyte) at operating temperature, decomposition at low pO2, SrO enrichment of and precipitation at the surface and Cr poisoning.

Several studies on the electrochemical properties of oxygen electrode materials are reported in the literature. Most of the studies are performed on porous electrodes with the purpose of having a realistic scenario; however we still lack fundamental understanding of the underlying degradation mechanism. Even though porous electrode studies have provided invaluable information about the degradation of the oxygen electrode, the deconvolution of geometrical effects from the intrinsic properties of the material is very difficult. Therefore, the presented work aims to study the oxygen reduction mechanism using geometrically well-defined dense model electrodes. Gd doped CeO2 (CGO) was deposited on polished single crystals YSZ (100) and on top of the CGO layer was deposited a (La0.6Sr0.4)0.99CoO3-δ (LSC40) using pulsed laser deposition (PLD). The thin CGO film (~100 nm) was deposited to avoid any reaction between the YSZ and LSC40 (250 nm). Subsequently, using photolithography and ion beam etching the microelectrode arrays with varying diameters (from 100 µm to 5 µm) were produced. Each sample has 4 macro-electrodes which were used as counter-electrode while performing electrochemical measurements. To observe the effect of temperature on the film microstructure and chemistry one sample was heat treated for 16 hours. SEM images, AFM and ToF-SIMS reveal similar behavior for both heat treated and as-deposited films. ToF-SIMS depth profiling reveals a Sr and Co rich surface compared to the bulk of the LSC40 for both samples. The difference between the two samples are in the distribution of common impurities, such as silica. After the heat treatment, the Si signal is higher in the LSC/CGO and CGO/YSZ interfaces (See Figure 1 a-b).

The electrochemical measurements were recorded in a Controlled Atmosphere High Temperature Scanning Probe Microscope (CAHT-SPM)5 which can reach temperatures up to 850 °C. The impedance spectra reveal a high frequency intercept which is related to the conductivity of the electrolyte, a small arc at high/middle frequencies originating from the electrode electrolyte interface, and finally a low frequency arc which is related to the air electrode interface. The evolution of the impedance spectra with changing temperature, atmosphere, polarization and microelectrode size combined with surface chemical analysis and imaging techniques reveals information about the underlying degradation mechanism of LSC electrodes.

General information
Publication status: Published
Organisations: Department of Energy Conversion and Storage, Department of Chemistry
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Publication date: 2017
Peer-reviewed: Yes
Event: Abstract from 232nd ECS meeting, National Harbor, Washington, DC, United States.
URLs:
http://ma.ecsdl.org.proxy.findit.dtu.dk/content/MA2017-02/39/1726#aff-2
Source: Findit
Source ID: 2304160853
Research output: Contribution to conference › Conference abstract for conference – Annual report year: 2017 › Research › peer-review