Oxygen reduction at the SrCo_{0.8}Fe_{0.2}O_{3-δ} mixed conductor

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Introduction

The objective of reducing the operating temperature of solid oxide fuel cells (SOFC), while maintaining economically viable power densities, has led SOFC material scientists to the conclusion that mixed electronic - ionic conducting oxides hold the key to meeting these performance targets. In this work, the activity of $SrCo_{0.8}Fe_{0.2}O_{3-\delta}$ (SCF) towards the oxygen reduction reaction (ORR) was measured using a three electrode half cell configuration over the temperature range of 550 to 1000 °C. The SCF mixed conductor has been reported in the literature as having the highest ionic conductivity of the La_{1-x}Sr_xCo_{1-y}Fe_yO_{3- $\delta}$ class [1] Our goal is to use it as a performance baseline for comparison to other mixed conducting materials to be synthesized.}

Previous reported research by other groups using SCF materials as a SOFC cathode have limited their electrochemical analysis to only full cell I-V-P curves [2]. In the present work, we use three electrode techniques to carry out a detailed kinetic analysis of the ORR at SCF materials.

Experimental Methods

After ball milling stoichiometric amounts of SrCO₃, Fe₂O₃, and CoO precursors together for 24 hours, and calcining at 1000 °C for 4 hours, a single phase material was obtained. The working, counter, and reference electrodes were fabricated through screen printing a slurry of these powders onto a dense gadolinium doped ceria (GDC) electrolyte support, followed by sintering at temperatures ranging from 1050 to 1200 °C for four hours. Cyclic voltammetry (CV), and electrochemical impedance spectroscopy (EIS) were utilized to study the low field (EIS, CV), high field (CV, potentiostatic) current-voltage characteristics of this material in air from 550 to 1000 °C. Powder XRD and SEM techniques were also employed to characterize both the crystallographic properties and particle/electrode morphology of SCF materials and cathodes.

Results and Discussion

For a SCF working electrode sintered at 1200 °C for four hours, the charge transfer resistance obtained from EIS curve fitting was found to be 0.068 Ω cm² at 700 °C. Furthermore, using the current interrupt method to overcome the series resistance problem, 2.5 A/cm² was generated at an overpotential of -200 mV. A detailed analysis of the total cathode resistance dependence on temperature will also be provided in terms of the charge transfer and interfacial polarization components as well as the Arrhenius characteristics of the SCF material.

It was found that the use of a 50:50 vol. % GDC/SCF functional layer between the electrolyte and pure SCF current collecting layer led to a significant reduction in the operating performance, generating only 750 mA/cm² at -200 mV and 700 °C. From EIS curve fitting, it was found that the magnitude of the charge

transfer resistance for this composite electrode remained relatively unchanged from a single phase cathode, whereas the interfacial polarization resistance increased substantially, accounting for 60 % of the total open circuit polarization resistance of the SCF/GDC half cell at 700°C, compared to 20 % of the total resistance for the noncomposite SCF cathodes.

The shape of the i - η curve for all SCF cathodes was found to exhibit a strong temperature dependence, showing a consistent transition around 700 °C from Butler-Vulmer exponential behavior to an increasingly linear form, followed by quasi reversible behavior above 800 °C. SEM analysis revealed that the performance of cathodes made from this mixed conductor is largely related to the degree of interfacial intimacy between the electrolyte and electrode materials. Furthermore, high currents can be obtained from mixed conducting cathodes, even when little apparent gas phase surface area of the electrolyte is exposed to air.

References

[1] Y. Teraoka, H. Zhang, K. Okamoto, Mat. Res. Bull. 5. (1998)

[2] Wan J-H, Goodenough JB. 2003. 8th Int. Symp. on Solid Oxide Fuel Cells (SOFC VIII), Paris, France. T1: In press