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Impedance Modeling of Mixed Ionic Electronic Conducting Cathodes for Solid Oxide Fuel Cells

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Solid Oxide Fuel Cell (SOFC) cathodes are under continuous development, and one focus of later year's research has been on lowering the operating temperature of the SOFC. It has been recognized that in order to operate at temperatures below 700°C the cathode should contain a mixed ionic electronic conductor (MIEC) such as strontium doped lanthanum cobaltite as cathode. A significant number of papers has been published on the topic of MIEC cathodes. Where most studies investigate the performance of the developed cathodes versus the microstructure or preparation of the electrodes, only few studies deal with the interpretation of the recorded impedance spectra and even fewer with impedance models of the cathode.

In this study a cathode consisting of the mixed conductor $(La_{0.6}Sr_{0.4})_{0.99}CoO_{3-\delta}$ (LSC40) and the ionic conducting material $Ce_{0.9}Gd_{0.1}O_{1.95}$ (CGO10) has been prepared in the ratio 50/50 wt%. The cathode was prepared using standard ceramic techniques such as planetary ball milling and preparation of an ink for screen printing. The ink was screen printed onto both sides of a CGO10 tape and fired. This symmetrical cell was characterized using impedance spectroscopy in the temperature range 550 – 850°C and in the pO_2-range 1 – 0.028 atm.

To analyze the impedance spectra a modified version of the Adler Lane Steel (ALS)-model [1] was developed. The ALS model is a 1D-continuum model that describes the impedance response of a porous MIEC cathode. The model takes into account the oxygen vacancy transport in the electrode, oxygen exchange at the gas/MIEC interface, gas diffusion in both the electrode and a stagnant gas layer outside the electrode, and the vacancy concentration of the MIEC. This model has been modified as to also include an ionic conductor.

As the oxygen non-stoichiometry of the LSC40 is a well known function of the temperature and pO_2, the fitting of an impedance spectrum of a LSC40:CGO10 composite cathode in principle reduces to a fit of an effective vacancy diffusion coefficient, D_v, and an exchange flux density, r_0.

Figure 1 shows area normalized impedance spectra of the cathode response at 750°C together with the best fit using the modified version of the ALS-model. In general the impedance model can describe the recorded impedance spectra with very high accuracy at all the temperatures and oxygen partial pressures.

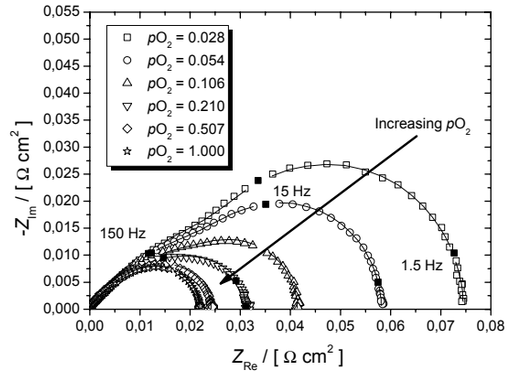

Figure 1. Impedance spectra of a LSC40:CGO10 composite cathode recorded at 750°C under different oxygen partial pressures, pO_2.

Figure 2 shows the fitted values of D_v and r_0 for one set of micro-structural parameters. It is observed that D_v decreases slightly with decreasing pO_2 while r_0 decreases by approximately a factor 3 in the investigated pO_2-range. These tendencies are as expected for a LSC40:CGO10 composite cathode. For the remaining temperatures a similar tendency has been observed and both D_v and r_0 are thermally activated parameters also as expected.

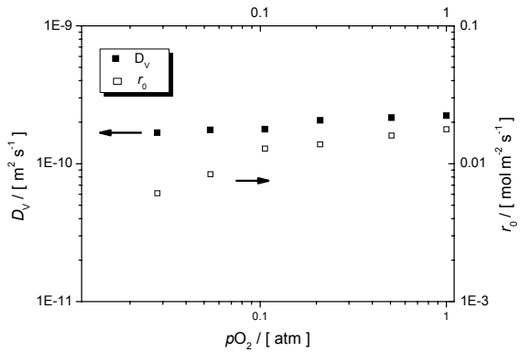

Figure 2. Fitted values of the effective vacancy diffusion coefficient, D_V, and an exchange flux density, r_0 as a function of the oxygen partial pressure at 750°C.

The developed model is also used in a trend study of how the impedance spectra evolve with different parameters such as thickness of the electrode, catalytic active surface area, volume fractions of the constituent phases etc. These results indicate that there exists an optimum value of the cathode thickness. For cathode thicknesses larger than this value, the diffusion of gaseous oxygen through the porous structure limits the performance of the electrode. For thicknesses smaller than the optimum value the performance of the electrode starts to be limited by the available electrochemically active area.

[1] S.B. Adler, J.A. Lane, and B.C.H. Steele, *J. Electrochem. Soc*, **143**, 3554-3564 (1996)