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Proton diffusion in the Ni/YSZ electrode of a Solid Oxide Cell

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A Solid Oxide Fuel Cell (SOFC) can be used as a Solid Oxide Electrolyser Cell (SOEC) for high temperature electrolysis of H_2O and CO_2 into H_2 and CO (syngas). Syngas can be catalyzed into various types of hydrocarbon energy carriers. This is highly interesting for synthetic fuel production due to a possibility of high efficiencies and low costs.

Unfortunately, under certain conditions, the cells show a paprtial passivation during the first 100 hours of operation, see figure 1. In some cases the cell performance subsequently recovers slowly over time. Electrochemical impedance spectroscopy (EIS) combined with gas changes shows that the passivation happens on the Ni/YSZ-electrode.

During this initial period of SOEC operation, the Ni/YSZ-electrode EIS response builds up as shown in figure 2. The 45 degree straight line at high frequency ending in a semicircle at low frequency is the response from diffusion across a finite Nerstian diffusion layer.[1] Comparison of the increase in polarization resistance (R_p) with the corresponding decrease in summit frequency indicates that the increase in R_p is due to an increase in a diffusion length, δ.

Isotope experiments (H → D) after operation in SOEC mode results in a decrease in summit frequency corresponding to 34% increase in diffusion coefficient. The classical diffusion theory predicts an increase of 41%.

By assuming the observed diffusion process is proton diffusion in the bulk of YSZ[2] one finds diffusion lengths of the order of 40 nm before SOEC operation and 400 nm after. Impurity rim ridges at the three phase boundary (TPB) with sizes up to 3 μm has been observed on Ni-point electrodes on YSZ.[3]

The above findings points toward a model for steam reduction as shown in figure 3. Steam is adsorbed on the YSZ surface where it is reduced. Protons migrate to the Ni-surface where they desorb as H_2. The observed increase in R_p is due to a build up of impurities at the TPB.

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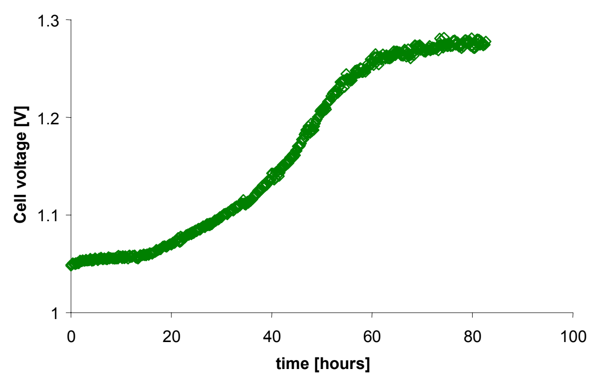

Figure 1. Cell voltage during SOEC operation. Test conditions were 750 °C, -0.25 A/cm^2, 70% H_2O + 30% H_2 to the Ni-electrode and O_2 to the LSM-electrode.

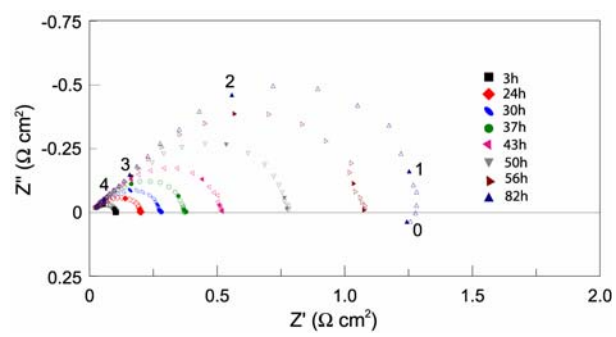

Figure 2. Nyquist plot of the Ni/YSZ-electrode EIS response obtained during SOEC operation. Test conditions are given in figure 1. Time after onset of SOEC operation is given in hours (h) Numbers on figure refer to frequency decade for the closed symbols. Note the decrease in summit frequency.

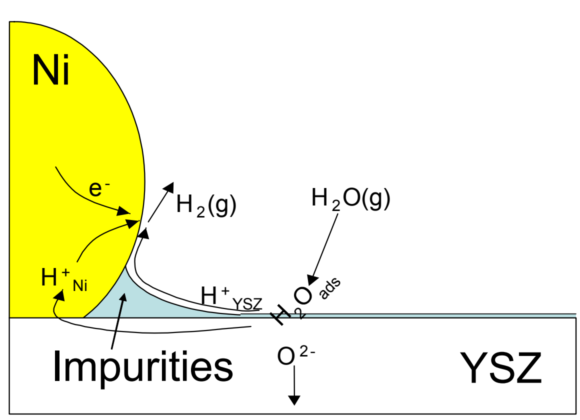

Figure 3. Impurities build up at the TPB and increase the diffusion length for protons migrating from the YSZ to the Ni.