

Kinetic Studies on Ni-YSZ Composite Electrodes - DTU Orbit (08/11/2017)

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Introduction Polarization of the Solid Oxide Cell (SOC) causes current to flow. If the fuel electrode is anodically polarized, the cell operates in fuel cell mode, oxidizing a fuel like hydrogen, carbon monoxide or hydrocarbons. In cathodic polarization the cell operates in electrolysis mode, reducing steam, carbon dioxide or both at the fuel electrode. Independent of polarization direction, the current flowing through the electrodes of an SOC is limited by processes such as adsorption and desorption of reactants or products, diffusion through the porous electrodes, activation or charge transfer at the reaction sites gas conversion at the flow fields, and ohmic drop across the electrolyte. Since these processes occur in both electrodes and some of them with overlapping characteristic frequencies, it is particularly challenging to isolate and characterize a particular mechanism. Furthermore, when polarized, the cell heats up due to joule heating of the electrolyte but also the electrodes either heat or cool due to exothermic oxidation or endothermic reduction of gaseous reactant species. Kinetic investigation of SOC electrodes independent of the above effects thus requires a carefully chosen cell geometry, methodology and operation conditions. Experimental The investigated cells consist of porous Ni/8YSZ composite working-electrodes with an active area between 0.8 and 1 mm² and ~100 mm² counter electrodes of the same material screen-printed on a special shaped 8YSZ electrolyte pellet. The electrodes are sintered in air at 1350 °C. Details of the cell geometry are given elsewhere¹. The cells were characterized by electrochemical impedance spectroscopy using a Gamry Reference 600TM potentiostat. Current/voltage characteristics were recorded at different temperatures and gas compositions using the same instrument. The tests are carried out in a single gas atmosphere with maximum flow rate of 6 L/h. Results and Discussion Current density vs working electrode overpotential curves recorded in the temperature range 800 – 650°C in a 50/50 H₂/H₂O fuel mixture are displayed in figure 1(a). The curve at 700°C shows that for a current density of 100 mA/cm² in cathodic polarization, an overpotential of ca. 150 mV is required, compared with 100 mV in anodic polarization. This reflects asymmetry^{2–6} in the kinetics of hydrogen oxidation and steam reduction. By recording current density vs overpotential curves at H₂/H₂O ratios of 30/70, 50/50 and 70/30 as displayed in figure 1(b) it could be shown that in the potential window investigated herein the dependence of kinetics on H₂/H₂O ratio is not significant. At any given potential in the investigated window, and independent of operation mode, there is a slight increase in current density with increasing steam content consistent. This translates to a decreasing area specific resistance of the fuel electrode electrochemistry with pH₂O. A power law dependency of -0.33 is reported in literature⁷. Outlook In this work experimental results of kinetic investigations on state of the art solid oxide cell electrodes carried out using a novel solid oxide cell geometry, allowing, for the very first time, determination of kinetic parameters void of influences such as temperature or reactant starvation will be presented. The results will provide a basis for discussion of existing analytical descriptions of the current/overpotential relations of SOC electrodes. References 1. C. Graves, T. L. Skafte, B. R. Sudireddy, J. Nielsen, M. Mogensen, in preparation. 2. T. Kawada et al., *J. Electrochem. Soc.*, 137, 3042–3047 (1990). 3. J. Mizusaki et al., *Solid State Ionics*, 70-71, 52–58 (1994). 4. C. R. Graves, S. D. Ebbesen, and M. Mogensen, in *ECS Transactions*, vol. 25, p. 1945–1955, ECS (2009). 5. P. Holtappels, L. G. J. de Haart, and U. Stimming, *J. Electrochem. Soc.*, 146, 1620–1625 (1999). 6. J.-C. Njodzefon, D. Klotz, A. Kromp, A. Weber, and E. Ivers-Tiffée, *J. Electrochem. Soc.*, 160(2013). 7. A. Leonide, Y. Apel, and E. Ivers-Tiffée, in *ECS Transactions*, vol. 19, p. 81–109, ECS (2009). Figures: Figure 1: Current density vs overpotential curves recorded (a) in the temperature range 800- to 650°C in a 50/50 H₂/H₂O ratio and (b) at 800°C in H₂/H₂O ratios 30/70, 50/50 and 70/30. [Figure]

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