

Investigation of oxygen reactions in a screen-printed Pt/YSZ-model electrode system

Y. Zheng^{a,b}, U. Sauter^b, L. Kunz^b, M. Streeb^b, G. Oehler^b,
K. Sahner^b, R. Moos^a

^aUniversity of Bayreuth, Functional Materials, 95440
Bayreuth, Germany

^bRobert Bosch GmbH, Applied Research 1, 70839
Gerlingen, Germany

Oxygen reactions in platinum/yttria stabilized zirconia (Pt/YSZ) systems have been investigated for several decades because of various possible applications like solid oxide fuel cells (SOFC) [1] or oxygen sensors [2]. However, research up to now was mostly either done on sputtered thin film electrodes deposited on single crystal YSZ (e.g. in [3]), which may differ strongly from electrodes in real applications, or on screen-printed porous electrodes (e.g. in [4]), which are difficult to reproduce. Therefore, we have investigated the properties of screen-printed, but dense Pt electrodes (with some ceramics additives to balance mechanical stress) on polycrystalline YSZ.

The microstructure of the electrodes is analyzed with Focused Ion Beam Scanning Electron Microscopy (FIB-SEM) and then reconstructed three-dimensionally. This method is for example described in [5]. For functional characterization of the electrodes, electrochemical impedance spectroscopy (EIS) is used. The impedance spectra are fitted with equivalent circuits for interpretation and for comparison with our physicochemical model. Other measurement techniques like cyclic voltammetry are used for the determination of the double layer capacitance.

Our physicochemical model is based on a model proposed by Mitterdorfer [6] and uses an impedance computation method by Bessler [7]. The model takes into account the following processes (see Fig. 1): adsorption/desorption, surface diffusion and charge transfer. A grain boundary diffusion path is also considered, as we have observed a dependence of the charge transfer resistance on the Pt/YSZ interface area size, although the electrodes are dense and have no continuous pores (see Fig. 2). Until now grain boundary diffusion in Pt has only been hypothesized for thin film Pt electrodes in literature [8-10]. In order to identify transport parameters through the dense electrode layer, we have conducted experiments with electrodes of different thicknesses and varying Pt grain sizes.

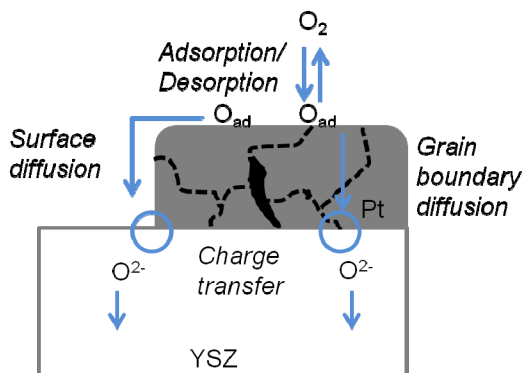


Fig. 1: Assumed Processes at Pt/YSZ-electrode system for electrode model.

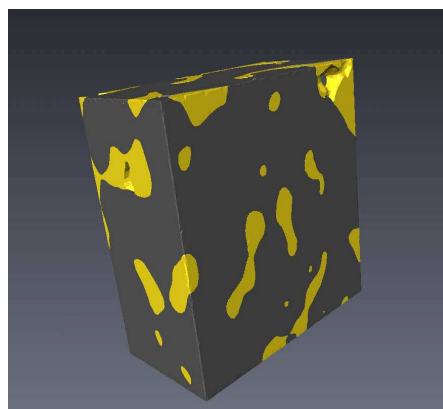


Fig. 2: Reconstructed structure from FIB-SEM of investigated electrode; grey – Pt, yellow – ceramics.

References

- [1] S. B. Adler, "Factors Governing Oxygen Reduction in Solid Oxide Fuel Cell Cathodes", *Chem. Rev.*, **104**, 4791-4843(2004).
- [2] C. López-Gándara, F. M. Ramos, A. Cirera, "YSZ-Based Oxygen Sensors and the Use of Nanomaterials: A Review from Classical Models to Current Trends", *Journal of Sensors*, **2009**, 15 (2009).
- [3] A. K. Opitz, J. Fleig, "Investigation of O₂ reduction on Pt/YSZ by means of thin film microelectrodes: The geometry dependence of the electrode impedance", *Solid State Ionics*, **181**, 684-692 (2010).
- [4] A. Mitterdorfer, L. Gauckler, "Reaction Kinetics of the Pt, O₂(g)|c-ZrO₂ System: Precursor-Mediated Adsorption", *Solid State Ionics*, **120**, 211-225 (1999).
- [5] J. R. Wilson, W. Kobsiriphat, R. Mendoza, H.-Y. Chen, J. M. Hiller, D. J. Miller, K. Thornton, P. W. Voorhees, S. B. Adler, S. A. Barnett, "Three-dimensional reconstruction of a solid-oxide fuel-cell anode", *Nature Materials*, **5**, 541-544 (2006).
- [6] A. Mitterdorfer, L. J. Gauckler, "Identification of the Reaction Mechanism of the Pt, O₂(g)|Yttria-Stabilized Zirconia System, Part II: Model Implementation, Parameter Estimation and Validation", *Solid State Ionics*, **117**, 203-217 (1999).
- [7] W.G. Bessler, "Rapid Impedance Modeling via Potential Step and Current Relaxation Simulations", *Journal of The Electrochemical Society*, **154**, B1186-B1191 (2007).
- [8] A. K. Opitz, A. Lutz, M. Kubicek, F. Kubel, H. Hutter, J. Fleig, "Investigation of the oxygen exchange mechanism on Pt|yttria stabilized zirconia at intermediate temperatures: Surface path versus bulk path", *Electrochimica Acta*, **56**, 9727-9740 (2011).
- [9] T. Ryll, H. Galinski, L. Schlagenhauf, P. Elser, J. L. M. Rupp, A. Bieberle-Hutter, L. J. Gauckler, "Microscopic and Nanoscopic Three-Phase-Boundaries of Platinum Thin-Film Electrodes on YSZ Electrolyte", *Advanced Functional Materials*, **21**, 565-572 (2011).
- [10] R. Schmiedl, V. Demuth, P. Lahnor, H. Godehardt, Y. Bodschwinna, C. Harder, L. Hammer, H. P. Strunk, M. Schulz, K. Heinz, "Oxygen diffusion through thin Pt films on Si(100)", *Applied Physics A*, **62**, 223-230 (1996).