QUANTITATIVE PHYSICOCHEMICAL MODELING OF SURFACE AND BULK TRANSPORT IN NI/YSZ PATTERNED ANODES

Marcel Vogler[1,2], Wolfgang G. Bessler[2]
[1]Universität Heidelberg, Interdisciplinary Center for Scientific Computing (IWR), Heidelberg, Germany
[2]German Aerospace Center (DLR), Institute of Technical Thermodynamics, Stuttgart, Germany
marcel.vogler@iwr.uni-heidelberg.de

Keywords: SOFC, Ni/YSZ, model anode, surface diffusion, elementary kinetics, modeling, simulation, validation

Introduction and approach

The traditional nickel/yttria-stabilised zirconia (Ni/YSZ) materials system is widely used today as anode for solid oxide fuel cells (SOFCs). However, the mechanistic details of the hydrogen oxidation reaction are still unclear. Previous studies [1-3] focused on a general qualitative characterization of the Ni/YSZ anode.

The aim of this study is to clarify the mechanistic details and identify processes governing anode performance. The model is used to interpret experimental results and to aid designing key experiments. In collaboration with Utz et al. [4] this was done using electrochemical characterization of Ni/YSZ model anodes.

In our model we resolve in detail the heterogeneous chemistry on the Ni and YSZ surfaces, we account for surface and bulk diffusion processes and use an elementary kinetic description for the charge transfer. We are able to comparatively study different electro-chemical pathways (i.e. charge transfer via surface spillover processes across the three phase boundary or bulk charge transfer across the electrode-electrolyte interface; see Fig. 1).

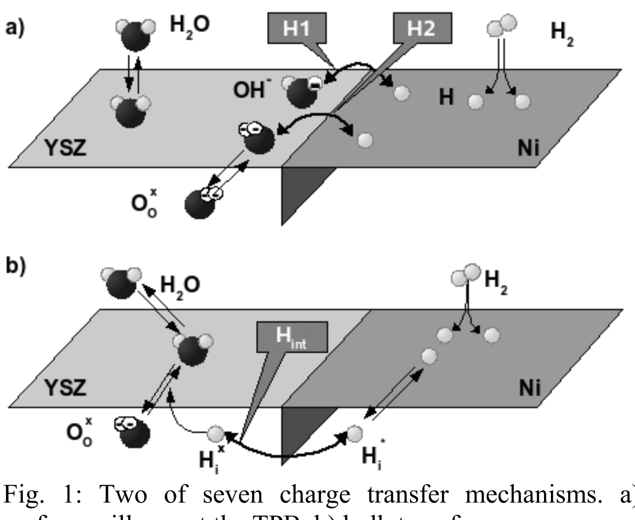

Fig. 1: Two of seven charge transfer mechanisms. a) surface spillover at the TPB, b) bulk transfer.

Model validation

Using published experimental [2] we were able to validate our model in good agreement with experimental data (Fig. 2) and determine unknown model parameters.

Model-based experiment design

Various numerical studies have been performed with different surface-spillover and bulk-bulk charge transfer pathways allowing for the identification of crucial experimental conditions needed for model discrimination. The dependence of electrode resistance on hydrogen concentration was found to be characteristic for the different charge transfer mechanisms.

Model-based interpretation of experiments

Current-voltage characteristics were compared to experimental data from Utz et al. [4] in a wide range of gas composition ($8{\cdot}10^2$ Pa < pH$_2$ < $9{\cdot}10^4$ Pa and $2{\cdot}10^1$ Pa < pH$_2$O < $6{\cdot}10^4$ Pa). Results show a cathodic limiting current indicating transport limitation by surface diffusion. Performing a sensitivity analysis reveals the importance of the electrolyte surface chemistry.

Conclusions

Model-based experiment design and interpretation is of key importance for clarification of mechanistic details in Ni/YSZ model anodes. Apparently inconsistent results from previous studies may be explained within one self-consistent picture.

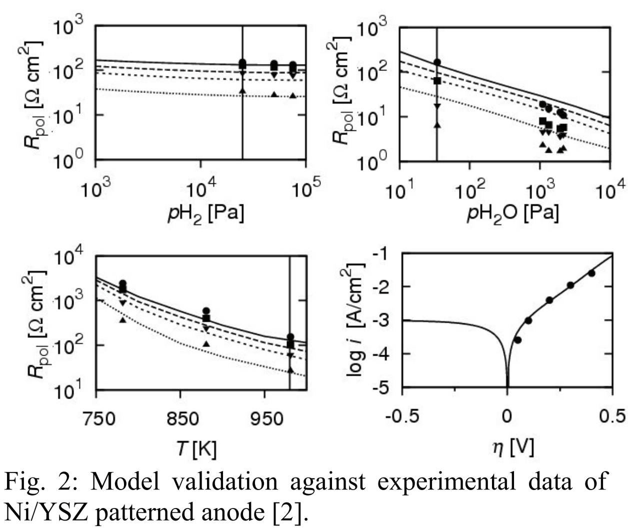

Fig. 2: Model validation against experimental data of a Ni/YSZ patterned anode [2].

References

[1] J. Mizusaki, et al. *J. Electrochem. Soc.*, 141,:2129-2134, 1994.

[2] A. Bieberle et al., *Electrochem. Soc.*, 148, A646-A656, 2001.

[3] B. deBoer, *PhD thesis*, 1998.

[4] A.Utz et al, SSI-17, 2009 (abstract submitted).