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Diffusion and Segregation of Iron in the Electrode System Pt(O_2)/YSZ

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The system Pt(O_2)/YSZ (YSZ = yttrium stabilized zirconia) is important for the general microscopic understanding of electrode processes in solid state electrochemistry. In spite of its simplicity many questions are unanswered still today after several decades of research. It is now generally accepted that the oxygen exchange takes place at the tpb (tpb = three phase boundary) gas/platinum/YSZ or in an active region in its close vicinity (Fig. 1).

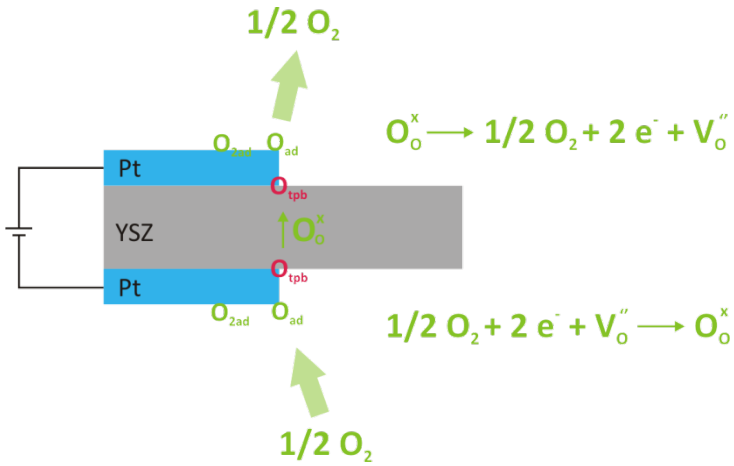

Fig. 1 The oxygen exchange processes at the tpb during electrochemical polarization.

It is also known that impurities can strongly influence the electrode kinetics. For example, silicon can form a blocking layer of SiO_2 which degrades the electrode performance [1].

One main impurity of commercial platinum catalysts is iron (Fe). To analyse the influence of iron in the electrode system, PLD (PLD = pulsed laser deposition) was used to prepare thin, well defined multilayers of platinum and iron on YSZ(111) single crystals with an overall thickness of about 1 µm. With those samples, diffusion und polarization experiments were made in air and vacuum.

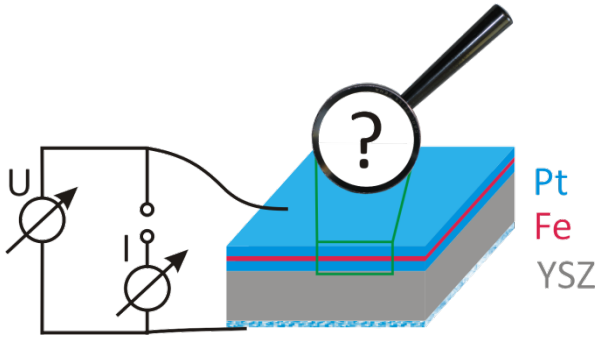

Fig. 2 Scheme of the experimental setup.

For the electrode characterization before and after the experiments we used ToF-SIMS, HREM, EDX and XRD. A main result is that the driving force for the diffusion and segregation of iron is the presence of oxygen. Annealing in air always caused the formation of iron oxide islands on the surface (Fig. 3, left). Electrochemically pumping of oxygen to the interface YSZ/Pt resulted in accumulation of iron there, while pumping of oxygen away from the interface led to no segregation, as shown by ToF-SIMS (Fig. 3, right). The fast diffusion of iron through the platinum layers can be explained by grain boundary diffusion.

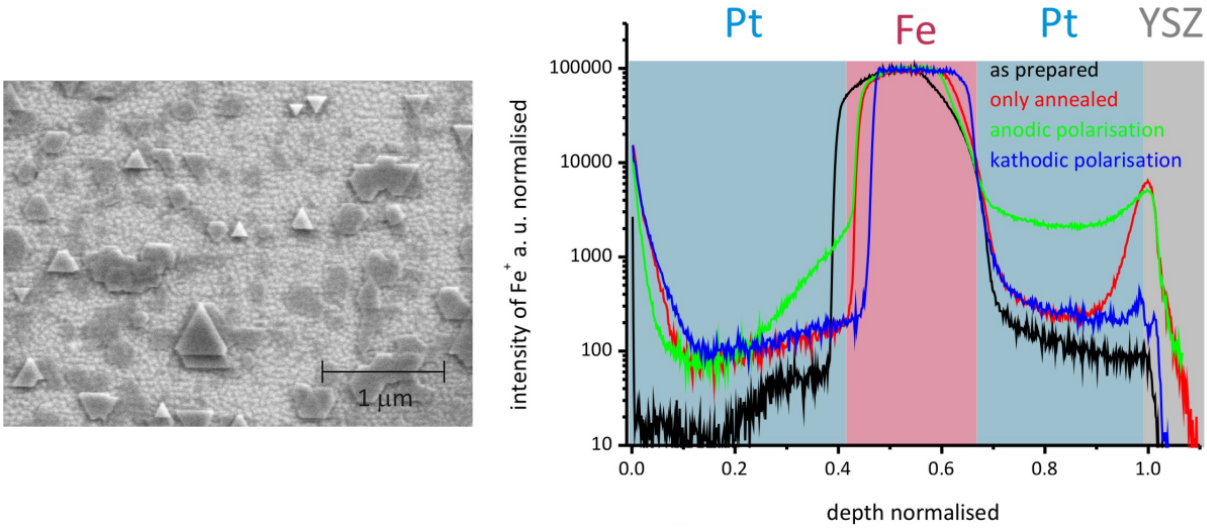

Fig. 3 Left: SEM picture of a sample annealed in air showing iron oxide islands. Right: SIMS depth profiles of different pieces of a sample exposed to different conditions.

References

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