

Chemical Potential Diagrams for polarized (La,Sr)MnO_{3+δ}-Cathode/8YSZ-Electrolyte-Interfaces

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The chemical stability of electrode/electrolyte-interfaces is correlated with the performance and the long term stability of a Solid Oxide Fuel Cell. The chemical stability is affected by the change in oxygen activity at the interfaces due to polarization of the electrode [1]. Chemical Potential Diagrams (CPD) based on the thermodynamic properties of the applied materials provide decisive information about the stability of the interface and the formation of secondary phases [2],[3].

In this work the stability of the LSM-cathode/8YSZ-electrolyte interface (LSM: La_{1-x}Sr_xMnO_{3+δ}, 8YSZ: 8 mol% Y₂O₃ doped ZrO₂) was analyzed using the software MALT [4]. CPD's of the cathode/electrolyte-interfaces have been calculated considering the electrode polarization. In case of an LSM-cathode, the electrode polarization is related to a gradient in oxygen activity. Fig. 1 shows a scheme of the oxygen transport in the cathode. The oxygen activity at the interface is linked to the overvoltage U_{pol} by the Nernst equation. To analyze the stability of a polarized cathode/electrolyte-interface by chemical potential diagrams, the interfacial oxygen partial pressure p(O₂)_{interface} < p(O₂)_{oxidant gas} has to be considered. For example at 1000 °C a voltage loss of 200 mV at the cathode corresponds to a p(O₂)_{interface} of 1.4·10⁻⁴ atm. In addition the defect chemistry of the materials, the related p(O₂)-dependency of the defect concentrations and the interdiffusion between LSM and 8YSZ has to be taken into account. A mixture phase [5] consisting of virtual compounds with the relevant defects was applied to regard the defect chemistry of LSM. The thermodynamic data of these compounds was adapted to achieve an agreement in oxygen nonstoichiometry with experimental results. In Fig. 2 the measured oxygen nonstoichiometry of LaMnO₃ [6] and the calculated values of the mixture phase consisting of the adapted compounds LaMnO_{2.5}, LaMnO₃, LaMn_{0.75}O₃, La_{0.667}MnO_{2.5} and La_{0.667}MnO₃. are displayed.

Fig. 3 shows the LSM mixture phase in the chemical potential diagram of the LSM/8YSZ system. The number of variable chemical potentials in a 3-dimensional chemical potential diagram is limited to 4. In the La-Sr-Mn-Zr-Y-O-system two compounds (SrO in 8YSZ and YMnO₃) had to be fixed. In the diagram the LSM/8YSZ-intersection represents a stable cathode/electrolyte-interface whereas the LSM/La₂Zr₂O₇-intersections correspond to the area where a lanthanum zirconate layer is formed. The stability of the LSM/8YSZ-interface is restricted to p(O₂)-values > 10⁻⁹ atm. Lanthanum zirconate formation takes place if the voltage losses exceed 0.53 V. If the interfacial chemical potential of Mn is reduced due to the diffusion of Mn into 8YSZ, lanthanum

zirconate formation can take place at higher p(O₂) resp. lower voltage losses as well. This kind of CPD provides valuable information about the stability of a cathode/electrolyte-interface under operating conditions. Further results will be presented in this contribution.

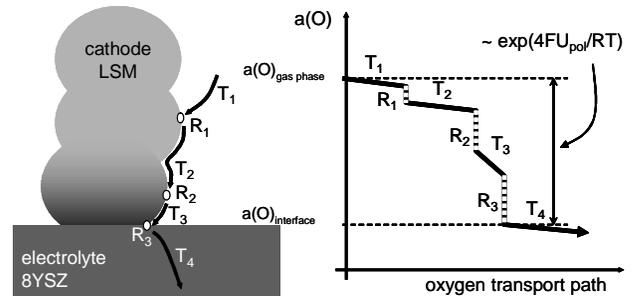


Fig. 1: Gradients in oxygen activity a(O) at a polarized cathode/electrolyte-interface.

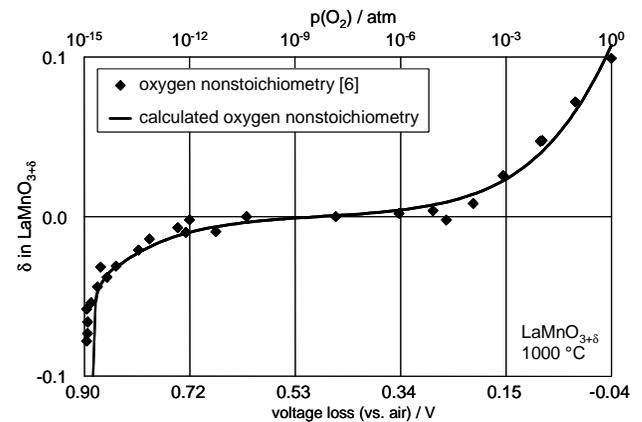


Fig. 2: Calculated and measured oxygen nonstoichiometry of LaMnO_{3+δ}.

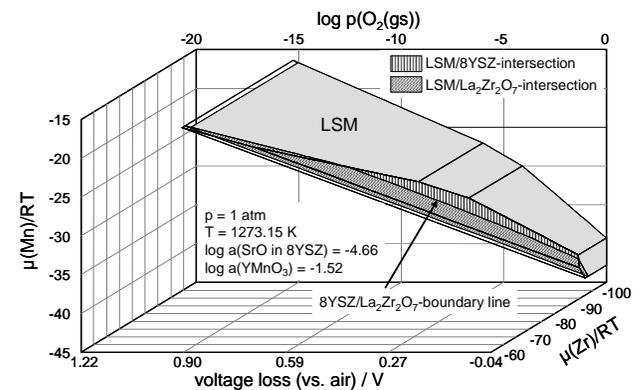


Fig. 3: Chemical Potential Diagram for the polarized LSM/8YSZ-interface.

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