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[1] M. T. Johnson, C B. Carter and H. Schmalzried, J. Am. Ceram. Soc 83, 1768-1772 (2000) [2] C. Korte, N. Ravishankar, C. B. Carter and H. Schmalz¬ried, Solid State Ionics 148(1-2), 111-121 (2002) [3] C. Korte, N.D. Zakharov and D. Hesse, Phys. Chem. Chem. Phys 5(25), 5530-5535 (2003) [4] C. Korte, and B. Franz and D. Hesse, Phys. Chem. Chem. Phys. 7(2), 413-420 (2005)

EFFECT OF (EXTERNAL) ELECTRIC FIELDS ON HETEROGENEOUS SOLID STATE REACTIONS WITH ONE OR MULTIPLE PRODUCTS – SPECIAL ROLE OF GRAIN BOUNDARY DIFFUSION

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Key Words: Solid state reactions, (external) electric fields, grain boundary transport, materials degradation.

During operation ceramic materials are often exposed to high electrical fields, which create a second driving force for mobile components in addition to the chemical potential gradient. Due to the ongoing miniaturisation in modern applications, interfaces gain in importance for the materials properties. Solid state reactions, negligible on macroscopic length scales, become more important on the nanoscale and thus become a frequent source of materials degradation

In this contribution the influence of an electric field on the kinetics and the morphological evolution of (heterogeneous) solid state model reactions will be highlighted [1-4]. Experiments on the reaction couples $MgO + In_2O_3$ (forming one product: $MgIn_2O_4$) and $Al_2O_3 + Y_2O_3$ (forming three products: YAG, YAM and YAP) were performed in thin film technique. Using linear transport theory, a time independent growth rate for the product layer(s) is expected, depending on the magnitude and the direction of the ionic current through the electrochemical cell and the difference of the ionic transference numbers in the product phase. This is generally different compared to solely diffusion controlled reactions without electric fields, where the reaction rate decreases with increasing product layer thickness. In a spinel forming reaction an enhanced growth rate for the product layer is predicted, when the divalent cations are more mobile and the trivalent oxide is attached to the cathode side (Fig. 1).
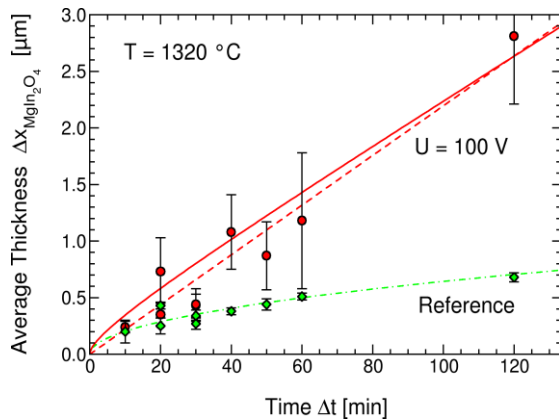

Fig. 1: Thickness of the $MgIn_2O_4$ layer vs. time for the reaction $MgO + In_2O_3$. Reference without field (diamonds) and with an applied external electric field (circles).

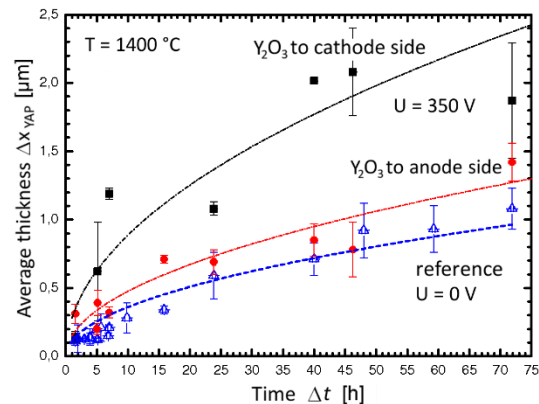Fig. 2: Thickness of the YAP layer vs. time for the reaction $Al_2O_3 + Y_2O_3$. Reference without field (triangles) and with electric field, Y_2O_3 layer to the cathode side (squares) and to the anode side (circles)

The role of grain boundaries as fast diffusion paths is highly emphasised. The morphology of the product layer is significantly different compared to a non-field-driven reaction. An analysis shows that the ionic transference numbers in (large angle) grain boundaries differs significantly from the bulk phase, causing locally different growth rates of the product layer. In case of a spinel forming reaction the divalent oxide tends to grow along grain boundaries through the complete product layer, reaching the trivalent oxide.

In solid states reactions forming more than one product layer, applying an additional electric field opens a possibility to control the product formation. Driven by an electric field, the growth kinetics of the product layer depends on the difference of the ionic transference numbers, without field on the Nernst-Planck coupled conductivities. In case of the reaction between $Al_2O_3 + Y_2O_3$ (three products: YAG, YAM and YAP) the formation of the perovskite phase (YAP) can be selectively enhanced when connecting the Y_2O_3 layer to the cathode side (Fig. 2).

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