Modeling of Binder Burnout and Sintering of Solid Oxide Electrolyte Tapes by Thermokinetic Analysis

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For optimal properties of ceramic electrolytes low heating rates and high temperatures are preferable, but these conditions contradict industrial applications. In this work the burnout of the organic binder which precedes the actual sintering as well as the sintering behaviour of thin 8YSZ electrolyte green tapes were investigated by thermokinetic analysis. With the help of the advanced thermokinetic software tool *Kinetics* (1) a comprehensive model based on the formalism of homogeneous chemical kinetics was built up without a knowledge of the physical nature of the underlying processes. This kinetic model reduced the measured data to a few formal parameters. Therefore, it was possible to calculate and optimise mass loss and shrinkage curves for an arbitrary temperature profile by computer without resorting to further experiments.

For the determination of the binder burnout kinetics, samples were heated up to 873 K in a thermogravity balance at different heating rates. The mass loss of a 8YSZ electrolyte tape doped with 1 mol% alumina (8Y1ASZ) due to the burnout of the organic binder is shown in fig.1. Simulation curves based on a kinetic model with four steps in a consecutive order indicated good accordance with the measured data. Verification of the applied model and its parameters was done by calculating a temperature profile for a constant mass loss rate (RCM) and by comparing it with an actual measurement based on this temperature profile (fig.2). Further experiments revealed that using a RCM temperature profile positive influence was taken on the sintering behaviour, because sintering onset was decreased and shrinkage increased in comparison with a conventional constant heating rate profile.

For the sintering experiments the temperature profiles were divided into three segments. For the first segment – the binder burnout – an optimised RCM temperature profile was used. Afterwards the samples were fired to 1673 K at different heating rates and then they were being held at this temperature during 2 -5 h. The actual shrinkage curves are presented in fig.3. For the better illustration only the sintering regime is shown and sintering onset of the different curves was set to time null. Sintering process was formally described by two consecutive reactions which were followed by two competitive reactions (fig.3), because total shrinkage depended on the heating rate. Based on this model a *rate controlled sintering* (RCS) temperature profile was calculated and comparison with actual measurement indicated a good agreement (fig.4).

SEM analysis as well as electrical conductivity measurements revealed, that by using rate controlled binder burnout and rate controlled sintering, optimisation of electrolyte properties were achieved. Time and energy saving was possible due to a decrease of total sintering time and maximum sintering temperature.

References

1. J. Opfermann, J. Therm. Anal. Cal, **60**, p641-658, (2000)

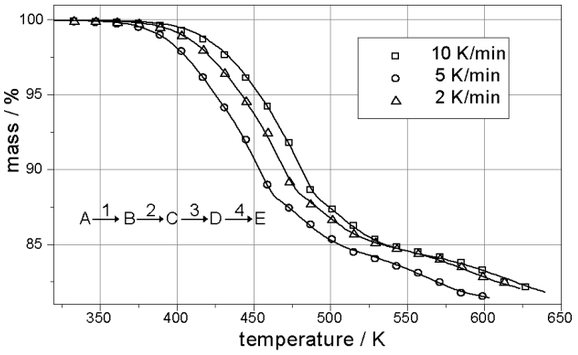

Fig. 1: *Mass loss of a 8Y1ASZ electrolyte tape due to binder burnout for different heating rate. A four-step model was used for the simulation (symbols: measurement, lines: simulation).*

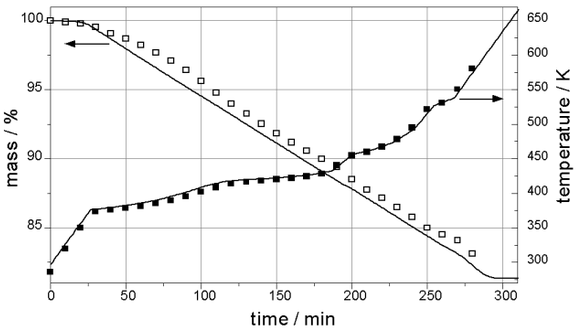

Fig. 2: *Comparison between prediction and measurement of binder burnout for a RCM temperature profile (0.07 %/min).*

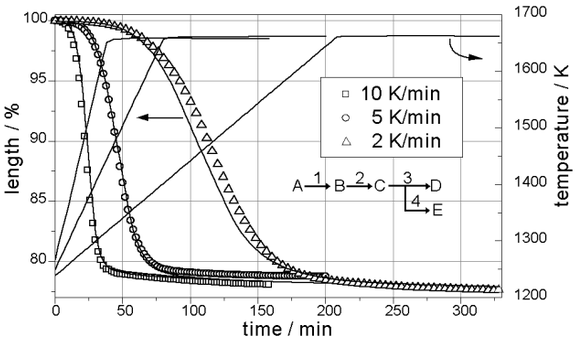

Fig. 3: *Sintering behaviour of 8Y1ASZ electrolyte tapesfor different heating rates as a function of time. A four-step model was used for the simulation (symbols: measurement, lines: simulation).*

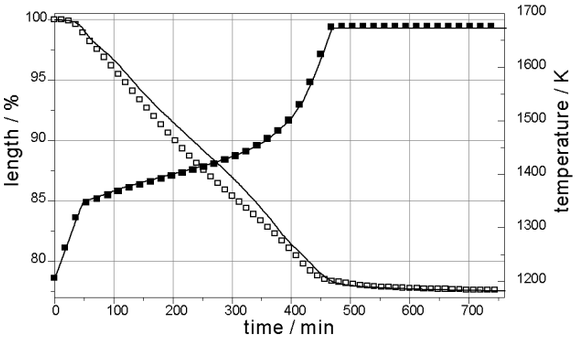

Fig. 4: *Comparison between prediction and measurement of sintering for a RCS temperature profile (0.05 %/min).*