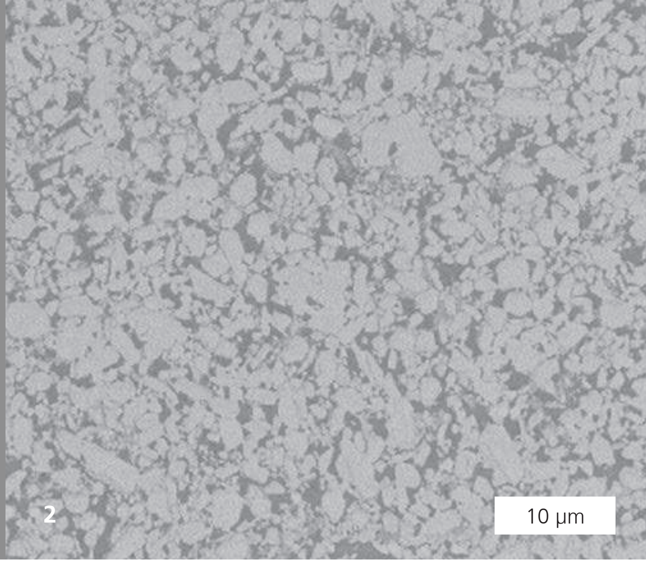
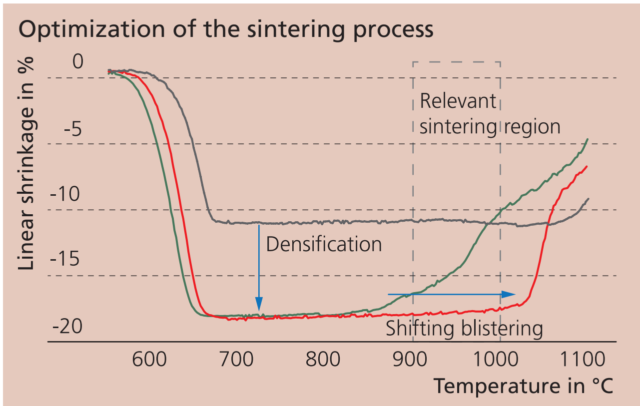


OPTIMIZATION OF SODIUM ION CONDUCTING GLASS-CERAMICS FOR SOLID ELECTROLYTES

Dr. Jochen Schilm, Dr. Axel Rost, Dipl.-Ing. Dörte Wagner, Dr. Katja Wätzig, Dr. Marco Fritsch

The evaporation of sodium at high temperatures (> 1600 °C) and the formation of a multiphase microstructure make the sintering of typical sodium-conducting solid-state electrolytes (i.e., NASICON and Na-ß"-Al_2O_3) an arduous task. Glass-ceramic materials in the system Na_2O-Y_2O_3-SiO_2 present an alternative that allows sintering below 1000 °C while achieving comparable conductivities. The aim of the present work was to produce dense monolithic and planar membranes by tape casting and pressureless sintering in air. The development of sintering-active glass-ceramic materials with ionic conductivities comparable to those of NASICON and commercial Na-ß-Al_2O_3 ceramics was the starting point of this work. The sintering process was optimized in order to achieve suitable process control for tape casting as an established ceramic shaping technology. A major challenge was the formation of porous microstructures during sintering at temperatures of above 800 °C due to evaporation of gaseous substances (H_2O and CO_2) in the highly viscous glass melt and the resultant volume expansion (foaming) of the components (see diagram). This hindered the formation of the conductive crystalline phase $Na_5YSi_4O_{12}$ with a dense microstructure and promoted the formation of the less conductive phases $Na_3YSi_3O_9$ and $Na_9YSi_3O_{18}$. Optimization of the glass synthesis process through combination of an adjusted grinding and precrystallization step shifted this behavior to higher temperatures. Furthermore, the average particle size of the prepared powders was reduced to less than 2 µm, which can be taken as a requirement for preparation of substrates with a thickness of less than 100 µm (Figure 1). Glass-ceramic materials with an ionic conductivity of $1.4 \cdot 10^{-3}$ S cm^{-1} at 25 °C in conjunction with an increase of the sintered density from 85–90 % to 97 % of the theoretical value were realized with the optimized process



(Figure 2). The doctor blade process was used for the fabrication of planar glass-ceramic substrates based on glass powders. Adhesion of the glass was avoided through modification of the sintering substrates and the heat treatment step; freestanding substrates with a thickness of 90 µm at the highest density were achieved. The dimensions of the sintered substrates were in the range of 50 x 50 mm.

Services offered

- Development/optimization of ion-conducting glass and glass-ceramic materials
- Sintering/shaping technologies for solid-state electrolytes
- Characterization of physical and electrochemical material properties
- Manufacturing and testing of cells

1 *Cross section.*
2 *Na^+-conducting glass-ceramic substrates.*

Contact Jochen Schilm • Phone +49 351 2553-7824 • jochen.schilm@ikts.fraunhofer.de