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The role of lamination conditions on electrochemical and mechanical performance of ceramic electrolytes for solid oxide fuel cells

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Abstract

The effects of the lamination pressure, temperature and duration on the mechanical, the electrochemical, the microstructural and the dimensional properties of the tape cast yttria stabilized zirconia (YSZ) electrolyte are experimentally investigated. The mechanical performance of the electrolyte is measured via tensile tests while the electrochemical performance is obtained by measuring the performance of the electrolyte supported cells fabricated in this study. The microstructural properties of the electrolytes laminated under different conditions, on the other hand, are evaluated via a scanning electron microscope. The dimensional change of the electrolytes depending on the lamination conditions after the sintering process at 1450 °C for 5 h is also considered in the study. The results show that YSZ electrolyte with high mechanical and electrochemical performance can be fabricated by low cost tape casting and isostatic pressing techniques after a careful decision of the lamination conditions. Within the parameter ranges considered, the optimum lamination pressure and temperature is decided to be 50 MPa and 60 °C, respectively, whereas the optimum lamination duration is found to be 10 min according to the experimental investigations.

Keywords: Solid oxide fuel cell; Electrolyte; Yttria stabilized zirconia; Lamination; Isostatic pressing

1. Introduction

Yttria stabilized zirconia (YSZ: 8 mol% Y_2O_3–ZrO_2) is the most common electrolyte material used in the fabrication of solid oxide fuel cells (SOFCs). Although it exhibits good ionic conductivity [1–3], mechanical properties [4–6] and chemical stability [7–9] under SOFC operation conditions, a conventional SOFC structured on YSZ electrolyte requires high operation temperatures around 1000 °C for an acceptable cell performance [10–12].

There can be found other electrolyte materials in the literature alternative to YSZ showing higher ionic conductivity thus higher performance at relatively lower operation temperatures. However, they all have some certain stability problems reported. Gadolinium or samarium doped ceria (GDC/SDC), for example, shows an electronic conductivity [13–15] which is a disadvantage for an electrolyte leading to both electrochemical and mechanical degradation. Similarly, strontium and magnesium doped lanthanum gallate (LSGM) reacts with extensively used Ni-based anode materials forming La–Ni phase resulting in a significant performance loss due to the de-activation of the anode catalyst [16–18]. Scandia stabilized zirconia (ScSZ), on the other hand, is found to have the highest ionic conductivity among the zirconia based electrolytes [19–21]. However, it is reported to suffer from the phase change causing a substantial conductivity drop [22–24]. Therefore, YSZ is chosen as the electrolyte material in this study.

Tape casting technique is generally employed to fabricate various SOFC components depending on the cell design with the desired properties including the thickness and the structure such as dense and thick electrolyte layer as the electrolyte support [25–29], porous and thick anode support layer [30–34] or dense and relatively thin electrolyte layer for the anode supported cell [35–39]. Furthermore, the single step lamination of the dense electrolyte layer sandwiched between two porous

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electrolyte scaffolds for the infiltration of the electrodes can be possible through a tape casting route [40–44]. In addition to being well known and low cost technique, the tape casting method is also considered to be the most suitable method for the mass production of SOFCs [35,37,45–47].

The tape casting method is based on the preparation of aqueous or non-aqueous slurries depending on the starting powder. The quality of the final product is mainly determined by the rheological properties which are affected by the powder type and size as well as the ratio of the additives such as solvent, binder, plasticizer, dispersant etc. Thus, there exist numerous studies in the literature presenting the rheological characterizations of various tape casting slurries [48–53]. Mukherjee et al. [48], for instance, investigated the effects of the dispersant type and the powder size on the green and the sintered densities of the tape cast YSZ. Phosphate ester was found to be much more effective dispersant than menhaden fish oil in the view points of the viscosity and the pseudoplasticity of the non-aqueous slurry. The best dispersion was achieved with finer powders and phosphate ester as a dispersant. Similarly, Bitterlich et al. [49] presented a detailed study focusing on the characterization of the aqueous slurry of YSZ by measuring the viscosity and its time dependent behavior as well as the strength of the internal structure of the slurry. In this aspect, four tape casting slurries having different compositions were examined. The slurry with the lowest binder to powder ratio provided the lowest sintering shrinkage due to having the highest green density and the thinnest green sheet behavior because of the low viscosity. However, this slurry showed the weakest internal structure which was found to be dominated mainly by the binder content. Lewis et al. [50], on the other hand, focused on the stress development during drying process. Increasing the plasticizer content, improving the colloidal stability by modifying the slurry rheology and controlling the solvent evaporation by altering the drying conditions were proposed in order to minimize the stress in the tape casting layer.

As a solid oxide fuel cell electrolyte, YSZ should serve highly dense structure in order to avoid internal combustion of the gases. In order to achieve this, the lamination method and parameters are also as significant as the rheological behavior of the tape casting suspension, since they determine the packing of the particles in the green tapes so the green density and the final dimensions, and provide an uniform density leading to the reduction of the internal stresses, the elimination of the cracks and the delamination between layers. Although there are numerous studies on the rheology of the tape casting slurries for SOFCs, there is very little done for the lamination process of the green tapes. Therefore, this paper focuses on the effects of the isostatic pressing conditions on the electrochemical, the microstructural and the mechanical properties of the tape cast YSZ electrolyte.

2. Experimental

2.1. Slurry preparation and tape casting

YSZ powders ($(Y_2O_3)_{0.08}(ZrO_2)_{0.92}$) with 6–9 m^2/g surface area and 0.5–0.7 μm particle size obtained from NexTech Materials (OH, USA) were selected as the starting powders. A non-aqueous tape casting slurries of YSZ were obtained by employing an alcohol based solvent composed of a mixture of methyl ethyl ketone and ethanol (both from Sigma-Aldrich, Munich, Germany) at a weight ratio of 2:3, respectively. 1 g organic dispersant (fish oil, Sigma-Aldrich) and 20 g solvent were added to 20 g of YSZ powders. The mixture was then ball milled for 24 h. After the addition of 5 g plasticizer (polyethylene glycol, Sigma-Aldrich) and 5 g binder (butvar, Sigma-Aldrich), the mixture was again subjected to another 24 h ball milling. Due to having laboratory scale devices and equipment, a required number of tape casting slurries with the same composition were prepared. The slurries were then tape cast by a laboratory scale tape casting equipment on Teflon coated Mylar strip with a blade gap of 200 μm. The final thickness of the electrolyte tapes was measured around 40 μm after drying in air atmosphere for 30 min.

2.2. Lamination process and sintering

The lamination of the YSZ tapes was achieved in two steps. In the first step, six YSZ tapes were stacked together and pre-laminated via laboratory scale uniaxial press under 5 MPa pressure for 4 min. The pre-laminated electrolytes were then moved to warm isostatic press in the second step. In order to investigate the effects of lamination conditions on the mechanical and the electrochemical performance of the YSZ electrolyte, the pressure and the temperature during the isostatic pressing process as well as the pressing time were altered within the limits of the isostatic pressing device. All tensile samples were cut into rectangular shape with 25 mm × 90 mm outside dimensions after the isostatic pressing and sintered at a temperature of 1450 °C for 5 h in a high temperature laboratory scale furnace with a temperature control unit. The thicknesses of the samples, on the other hand, were allowed to change without restrictions depending on the lamination conditions. The thicknesses of the samples just after the isostatic press could not be measured, since it is not possible to obtain accurate data due to the deformable nature of the green laminates which may give inconsistent results. However, the thicknesses and the outside dimensions of the electrolytes after the sintering were measured by a digital micrometer with a sensitivity of 0.001 mm and a digital compass with a sensitivity of 0.01 mm, respectively. These measurements were performed on samples belonging to each case considered. However, they were prepared by using the same tape casting slurry and casting conditions in order to obtain reasonable conclusions about the green and final density as well as the mechanical/electrochemical properties of the final products depending on the lamination conditions. The thickness and the width data are also needed to calculate the fracture strength by simple dividing the fracture force to the cross-sectional (thickness x width) area. After determining the final shrinkage of all samples, electrolyte supports for the cell tests were cut similarly by a laser cutter again according to the previous dimensional measurements such that all have the same outer dimensions of 60 mm × 60 mm after the sintering step at 1450 °C for 5 h. Thus, the change in the electrolyte resistance or the electrochemical performance of the electrolyte can be due to the variation of the