

DEVELOPMENT OF CERAMIC MULTILAYER DEVICES FOR CLEAN AND EFFICIENT ENERGY CONVERSION

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ABSTRACT

Recently, ceramic multilayer devices such as solid oxide fuel cells (SOFC), solid electrolysis cells (SOEC), oxygen transport membranes (OTM) and electrochemical flue gas purification devices have attracted attention for highly efficient energy conversion and reduction of air pollution. DTU Energy is developing these devices on a proof of concept level and fabricating the required ceramic multi-layer structures, using state of the art ceramic processing techniques. Selection of stable and high performing materials and reliable and cost efficient ceramic fabrication processes are important for the commercialization of these technologies. The success of such devices is often limited by materials and their microstructure (high temperature stability, performance) or by reliability issues (e.g. mechanical robustness, integrity of gas sealings). This study tries to show exemplarily for the development of oxygen transport membranes (OTM) some of the common challenges in materials selection, fabrication and testing of such ceramic multilayer devices. Oxygen transport membranes (OTMs) are dense ceramic membranes that can supply oxygen to high temperature combustion processes, for example for oxy-fired biomass gasification or in cement industry [1].

Membrane material development

An OTM material should ideally have high ionic and electronic conductivity as well as good chemical stability under both oxidizing and reducing conditions. For reactions under strongly reducing atmospheres and at high temperatures, such as for syngas production, single phase doped ceria materials, such as $Ce_{0.1}Gd_{0.2}O_{2-\delta}$ (CGO), were evaluated as thin film oxygen membranes [2, 3]. For less reducing atmospheres (e.g. oxygen separation from air), CGO has too low electronic conductivity. To overcome this problem, the strategy of preparing dual phase membranes seems promising [4], and several different candidate composites were investigated. Dual phase membranes including $La_{0.6}Sr_{0.4}FeO_{3-\delta}$ (LSF-CGO) and ZnO-CGO were selected for further studies, fabricated as thin film membranes and tested.

Design and fabrication of asymmetric oxygen transport membranes

Planar and tubular thin film membranes of single phase CGO, dual phase LSF-CGO and ZnO-CGO have been prepared on porous MgO, YSZ and on Ni-YSZ cermets. A planar membrane of LSF-CGO was also prepared by using phase inversion tape casting resulting in a support structure with an highly oriented porous structure to reduce gas transport limitations. The tubular asymmetric membranes were prepared on MgO and YSZ porous supports. The design and fabrication of planar and tubular asymmetric membranes will be explained by highlighting specific challenges, for example the simultaneous co-firing of the porous support structure and the dense membrane layer.

Membrane testing

Depending on membrane architecture and test conditions, CGO based single phase or dual phase membranes demonstrated high oxygen fluxes of 2 to 10 Nml min⁻¹ cm⁻² at 850°C when subjected to air on the feed side and strongly reducing conditions on the permeate side (CO, H_2).

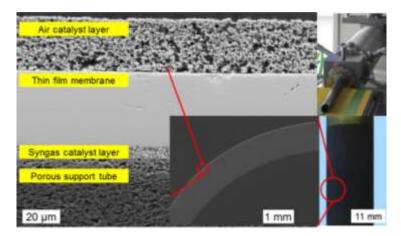


Figure 1: Tubular asymmetric oxygen transport membranes [3] showing a microscope picture of the microstructure (left), a photo of top part of a membrane tube (right, bottom) and a picture of the extrusion process for the porous support (right, top).

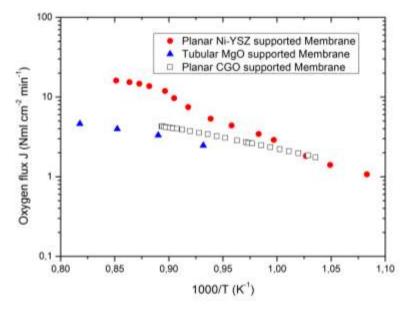


Figure 2: Oxygen permeation through single phase asymmetric CGO oxygen transport membranes with different architectures, measured in air vs humidified hydrogen: a) planar Ni-YSZ supported CGO membrane (red circles), b) planar CGO supported CGO membrane (squares) and c) tubular MgO supported CGO membrane (blue triangles).

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