A solar-based membrane reactor for hydrogen production

S. Baumann[a,*], F. Zeng[a], S.M. Gross-Barsnick[b], K. Bittner[b], F. Schulze-Küppers[b], J.P. Rincon Duarte[c], S. Ackermann[d], N. Margaritis[b] and N. C. Neumann[c]

[a] Institute of Energy and Climate Research IEK-1, Forschungszentrum Jülich GmbH, Jülich, Germany

[b] Central Institute of Engineering, Electronics and Analytics ZEA-1, Forschungszentrum Jülich GmbH, Jülich, Germany

[c] Institute of Future Fuels, German Aerospace Center DLR, Cologne, Germany

[d] Synhelion SA, Lugano, Switzerland

[*] s.baumann@fz-juelich.de

Abstract

Green hydrogen is a prerequisite for a carbon-neutral industry with an expected global demand of over 180 Mt per year [1]. To cover this drastic demand increase, existing technologies, such as electrolyzers, need to be quickly enhanced and promising novel concepts pursued. Solar thermal-powered membrane reactors can produce green hydrogen based on steam thermolysis at below 1000 °C. This technology has the advantage of being powered purely by thermal energy, which omits the requirement of expensive large-scale electrical energy conversion and storages. In contrast, concentrated solar energy has the potential of a cost-efficient 24-7 operation when a thermal energy storage is included [2]. Project MESOWAS aims to develop and test a solar-based membrane reactor for hydrogen production based on a solid oxide cell stack design [3]. In the membrane stack reactor, steam is introduced as feed gas in countercurrent to biomethane on the permeate side of the oxygen permeable membrane. Thus, hydrogen is produced on the retentate side and syngas (CO/H_2) on the permeate side by partial oxidation of biomethane. The stack will consist of up to 4 membrane layers in the size of max. 100 cm² each. Solar thermal energy will cover the energy demand of the reactor for the H_2O splitting reaction. The solar heat integration is simulated to guide the reactor design towards achieving a homogenous solar flux distribution.

The membrane material was selected on theoretical basis with regard to chemical stability, chemical expansion, and ambipolar conductivity in operation conditions considering doped ceria as well as doped strontium titanate. $Sr_{0.97}Ti_{0.75}Fe_{0.25}O_{3-\delta}$ (S97TF25) was chosen for the Proof-of-Concept (PoC) in particular due to its high stability in syngas conditions as well as significantly lower chemical expansion compared to doped ceria. The major downside of this material is a minimum in ambipolar conductivity at medium p_{O2} calculated from literature data (Fig. 1). Therefore, additional doping strategies will be followed in parallel to the component development in order to assess the general potential of doped $SrTiO_3$ for variable targeted application after PoC-module operation. Disc-shaped membranes are fabricated and sealed in metal frames, which are stacked to form this PoC-module. Both housing metal selection and joining technique are determined by the membrane material and its properties. In particular precise match in thermal expansion and chemical compatibility of all reactor components is required. Based on S97TF25 thermal/chemical expansion behavior ferritic chromium containing steels were selected for the frames. For operation up to 900 °C gastight S97TF25-metal joints are realized by means of glass solders based on calcium silicate glasses as well as reactive air brazing.

The temperature distribution of the membrane stack reactor is simulated with Ansys based on different solar flux distribution profiles. The flux distribution from the High-Flux solar simulator in Cologne [4] is obtained with the FEMRAY (Finite Element Mesh Ray Tracing) approach and analyzed with SPRAY (Solar Power Raytracing Tool). A setup where the membrane stack reactor is located at the focal point of the flux distribution was assessed as unfavorable, as the temperature distribution at the stack surface of > 220 K is too inhomogeneous. Thus, different options to homogenize the solar flux distribution were investigated, e.g. setting multiple focal points, moving the reactor behind the focal point, including an absorber plate and a cavity design. Simulations have shown that positioning the reactor 30 cm behind the focal point will lead to a more homogeneous temperature distribution on the membrane reactor with a max. ΔT of about 60 K in the center of the reactor area, which is the region where the membrane is placed.

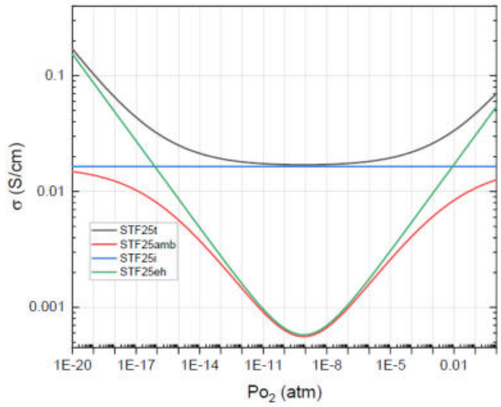

Fig. 1: Estimation of relevant conductivities for STF materials as a function of pO2.

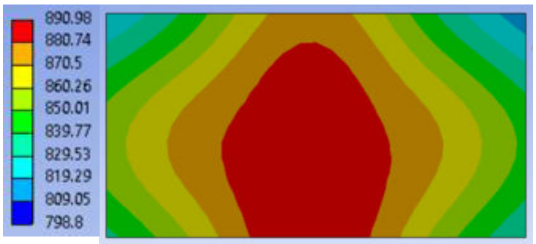

Fig. 2: Simulated surface temperatures of a membrane stack reactor with 4 lamps based on the DLR high flux simulator in Cologne. Position of reactor is 30 cm behind the focal point. Scale gives temperature in °C.

The solar thermal-powered membrane reactor is designed for hydrogen production with biomethane as reducing agent. The initial simulations yield promising results for an operation at 900 °C. The solar-powered operation was optimized towards a homogenous temperature distribution in the stack reactor to avoid thermally-induced instability of the membranes. A Proof-of-Concept module will be installed and operated in a solar furnace available at DLR.

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