

# ADIABATIC REACTOR SIMULATIONS OF THE REVERSE FLOW CATALYTIC MEMBRANE REACTOR CONCEPT WITH PEROVSKITE MEMBRANES

J. Smit, W. Zhang, M. van Sint Annaland and J.A.M. Kuipers,

University of Twente, Enschede, The Netherlands

In Gas-To-Liquid processes natural gas is converted to syngas via partial oxidation with pure oxygen, which is subsequently converted to liquid fuels via the Fischer-Tropsch reaction. Mainly due to the large costs associated with cryogenic air separation and complex heat integration GTL processes have not found widespread application yet, although some large plants are currently being commercialised in Qatar. In these GTL processes excess oxygen (20-40%) needs to be used for the partial oxidation unit together with preheating of the feed streams (up to 400 °C) to achieve high syngas yields.

The air separation costs can be substantially reduced by integrating the air separation inside the partial oxidation reactor using perovskite membranes as was shown by a number of authors (e.g. Balachandran, 1995). In order to integrate the recuperative heat exchange inside the membrane reactor as well to reduce costs even further the reverse flow concept can be used. Therefore, we recently proposed the Reverse Flow Catalytic Membrane Reactor (RFCMR) concept with perovskite membranes (Smit et al., 2004), see Figure 1 for a schematic overview.

With a simulation study in which a constant oxygen flux all along the membrane was assumed the conceptual feasibility of the RFCMR concept with perovskite membranes was already demonstrated (Smit et al., 2003). However, it is well known that the oxygen permeation rate through a perovskite membrane strongly depends on the temperature and the local syngas composition. Therefore, the oxygen permeation flux through a perovskite membrane ( $\text{LaCaCoFeO}_{3-\delta}$ ) was measured experimentally with different inert and reducing sweeping gasses at different temperatures. From the experimental results an expression for the permeation rate was derived.

A simulation study has been carried out on the conceptual feasibility of the RFCMR concept with perovskite membranes, using a detailed reactor model where the influences of the temperature and the local gas composition at the permeate side on the oxygen permeation rate are fully accounted for. The simulation results clearly show that stable reactor operation without the occurrence of temperature runaways can only be achieved when the oxygen permeation rate is restricted. Therefore, precautions are required to limit the oxygen permeation flux, for example by the use of porous support layer that acts as an additional mass transfer resistance (while simultaneously the mechanical strength of the membrane can be enhanced).

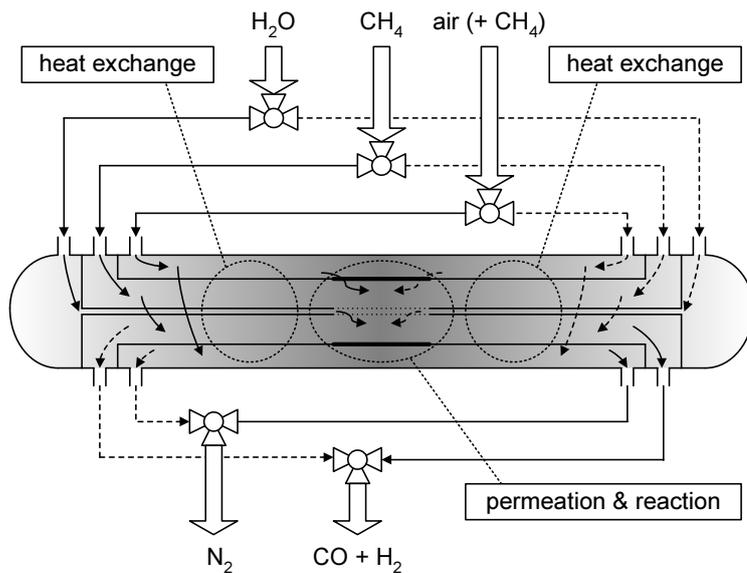


Figure 1. Schematic overview of the RFCMR concept with perovskite membranes.

Balachandran, U., Dusek, J.T., Mieville, R.L., Poeppel, R.B., Kleefisch, M.S., Pei, S., Kobylinski, T.P., Udovich, C.A., and Bose, A.C., *Appl. Cat. A-Gen.*, 133:1 (1995), 19–29

Smit, J., van Sint Annaland, M., and Kuipers, J.A.M., *Int. J. Chem. React. Eng.*, 1 (2003), A54

Smit, J., van Sint Annaland, M., and Kuipers, J.A.M., *Chem. Eng. Res. Des.*, 82:2 (2004), 245-251