Testing and analysis of a membrane reactor utilizing LTA membrane for intensification of CO_2 conversion routes

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Highlights

- Detailed mathematical model for water permeable membrane reactors developed.
- Validation of membrane reactor performance for CO_2 utilization in lab-scale.
- Conversions up to 2-3 times higher compared to the traditional reactor equilibrium can achieved.

1. Introduction

Process intensification by steam separation is a promising concept enabling the development of novel CO_2 utilization routes [1,2]. Many types of hydrophilic zeolite materials have been reported in literature to be used in membrane reactors (MRs) for in-situ H_2O removal, among which LTA membranes show great potential [3]. The permeation of water and other species through LTA membranes is a complex phenomenon. The apparent flux of water vapor and the selectivity to the transport of permanent gases are controlled by a number of (often competing) mechanisms. The understanding of these mechanisms is crucial to the development and optimal design of novel membrane reactors for a wide variety of applications, including the most important carbon utilization processes. In this work we study the impact of water concentration on the permeation through LTA membranes in the presence of other molecules, at reaction conditions, develop appropriate balance equations to model membrane reactors. We will also present experimental results obtained from a lab-scale membrane reactor system.

2. Methods

We have developed mathematical models for the permeance of water vapor and other species which includes consideration of both micropores transport as well as transport through defects (macropores). We have derived balance equations models for such membrane reactors which incorporate the permeance model, i.e., the permeance is treated as a variable which depends on local conditions (composition, temperature etc.), not as an empirical parameter, as has been done in most of the literature. These models are used to study optimal system configurations. Heat balance equations are coupled, enabling to gauge a number of system configurations of heat supply, calculate the heat required and study the implications of temperature gradients (including radial gradients) in the system on its performance, given the complex temperature dependency of the controlling phenomena.

We have also designed and constructed a lab-scale experimental system to test and study membrane reactor configurations utilizing LTA membranes. The design of the membrane-reactor assembly is depicted in Figure 1. Membrane tubes have been obtained from Fraunhofer Institute, Germany, see a photo in Figure 1.-left. We use commercial catalytic pellets to investigate the reverse-water-gas-shift (RWGS) and methanol synthesis reactions, feeding pure CO_2 and H_2.

3. Results and discussion

Some model predictions of the CO2 conversion in the RWGS reaction with respect to membrane area (represented by the Pem number) at various values of selectivities (permeance of water vs permeance of H2) are given in Figure 2, for two cases: Left – no sweep gas is used, right – sweep gas is used in the permeate with a flow rate 10 times higher than the feed flow rate. It is apparent that improving the selectivity above a certain value does not lead to significantly higher conversion if no sweep is used.

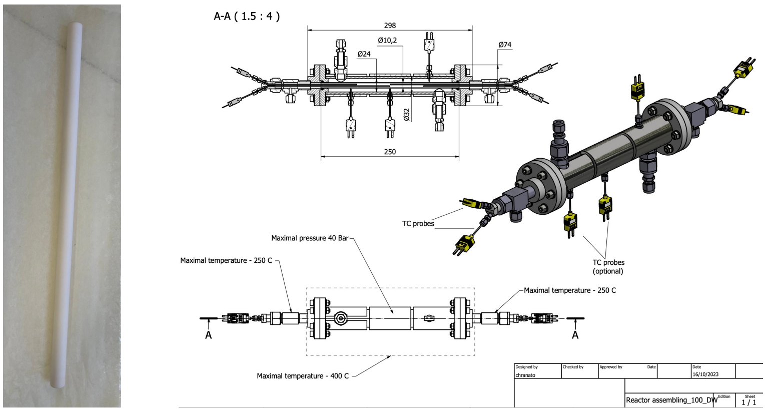

Figure 1. Drawing of the recent version of the design of a lab-scale membrane reactor assembly. A photo of one of the membrane tubes which will be used is on the left.

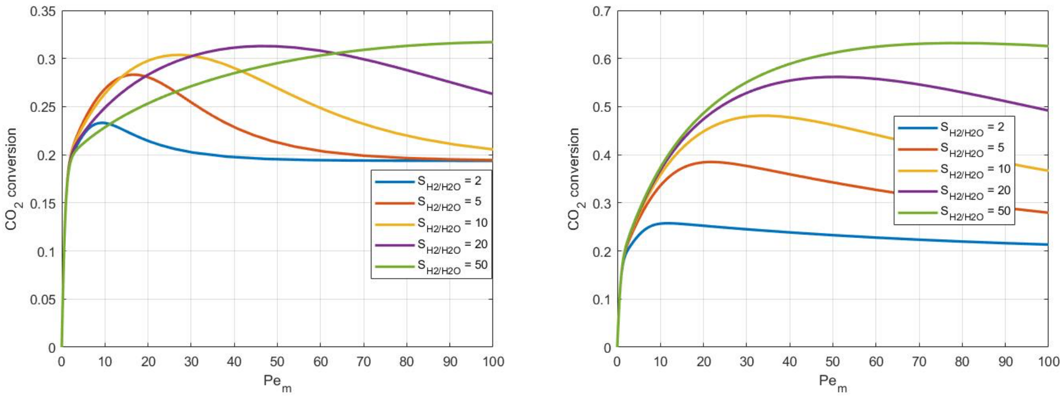

Figure 2. CO_2 conversion vs the dimensionless Pe_m number for various selectivity values ($S_{H2O/H2}$). Temperature is 573 K, reaction pressure is 10 bar and permeate is at 1 bar. Co-current configuration. Left: No sweep gas. Right: Sweep gas at a flow rate 10 times higher than feed rate.

4. Conclusions

Here, we analyze and demonstrate the intensification of CO_2 conversion in membrane reactor employing hydrophilic LTA membrane. We have developed a mathematical model to predict the reactor performance and analyzed the impart of operating conditions on the CO_2 conversion in the RWGS reaction. Significant intensification can be achieved under optimal conditions, reaching conversions 2-3 times higher compared to the traditional reactor equilibrium conversion. Improving the membrane selectivity to water may not have a high impact when operating without sweep gas.

References

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Keywords

CO2 utilization; Membrane reactors; LTA membranes; Modelling.