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Scalability of multitube membrane modules for hydrogen separation: Technical considerations, issues and solutions

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ABSTRACT

Palladium membrane technology has shown promising features for the development of a sustainable hydrogen economy. Nonetheless, the contribution of a palladium membrane technology to economic and societal development requires its commercialization, diffusion and utilization. To generate enough incentives for commercialization, it is necessary to demonstrate the scalability and robustness of the membranes in industrial settings. Consequently, this work utilizes pilot-scale experimental data generated under industrial conditions to validate a Computational Fluid Dynamics (CFD) model, which was up-scaled and utilized to determine the intrinsic phenomena of palladium membrane scale up. This study reveals the technical/engineering requirements for the effective design of large scale multitube membrane modules. Mass transfer limitations and concentration polarization effects were studied quantitatively with the developed model using the defined parameters Concentration Polarization Coefficient (CPC) and Effective Average CPC (EAC). Methods for diminishing the concentration polarization effect were proposed and tested through the simulations such as i) increasing convective forces and ii) designing baffles to create gas recirculation. For scaled-up membrane modules, mass transfer limitation is an important parameter to consider as large modules showed severe concentration polarization effects. Certainly, this work shows for the first time the main features required when designing large scale membrane reactor modules.

1. Introduction

Hydrogen as an important industrial raw material and clean energy carrier is attracting attention since traditional energy carriers (fossil fuels) are facing potential depletion and environmental problems caused by CO_2 emissions [1]. Ultrapure H_2 is required for most industrial applications, which leads to the necessary study of H_2 separation processes. Palladium membrane technology is an efficient and economically preferred method for H_2 purification and a promising method for carbon capture while coupled with the integrated gasification combined cycle (IGCC) systems [2–4]. Integrating palladium membranes with IGCC systems allows the co-production of electric power and pure H_2. Different membrane modules were studied in previous lab-scale studies and have shown promising results [5,6]; however, for evaluating the commercial potential of the technology, data from larger scale studies are needed. Previously, a pilot scale palladium membrane module for H_2 separation from coal-derived syngas was assembled at Worcester Polytechnic Institute (WPI) and tested at NCCC (National Carbon Capture Center). In order to achieve a larger scale module with larger membrane surface area compared with single-tube lab scale modules, a multi-tube configuration (7 membrane tubes) was chosen for its high membrane surface area to module volume ratio [7]. The test provided data for membrane permeance, H_2 product purity and H_2 recovery during the 900 h run under a single specified operating condition. The optimization of the membrane module requires a thorough understanding of the H_2 separation process such as mass transfer limitations, tube-to-tube H_2 flux variation and flow pattern.

The mass transfer limitation of the retentate side, concentration polarization, is caused by the rapid depletion of H_2 molecules at the surface of the membrane [8]. This depletion is caused by the difference in membrane permeance (high) and H_2 diffusion (low). The driving force in palladium membranes is the H_2 partial pressure difference across the membrane; consequently, as the concentration of H_2 reduces at the membrane surface in the retentate, the performance of the membrane declines downstream. The concentration polarization phenomenon is often neglected; however, it is very significant for membranes with high permeance and selectivity. As the theoretical

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Nomenclature

A	Membrane surface area m
D_i	Binary diffusion coefficient of species *i* in mixture $m^2\ s^{-1}$
F	External body forces (gravity, electric, etc.) N
I	Identity matrix –
j_i	Mass flux of component *i* mol s^{-1}
L	Membrane length m
M_i	Molar mass of component *i* g mol^{-1}
M_n	Average molar mass of mixture g mol^{-1}
p_i	Partial pressure of component *i* in the reaction zone bar
p	Total pressure bar
Q_{in}	Volumetric inlet flow rate $m^3\ s^{-1}$
R_g	Ideal gas constant J K mol^{-1}
R_i	Production rate of species *i* mol m^3
$\bar{P}_{H2}$	Hydrogen permeance of the membrane $Nm^3\ m^{-2}\ h^{-1}\ bar^{-0.5}$
T	Temperature K
t	Membrane thickness μm
u	Velocity field m/s
x_i	Mole fraction of species *i* –
x	Coordinate perpendicular to module axis m
y	Coordinate perpendicular to module axis m
z	Coordinate along module axis m
Δ	Delta index (see Eq. 11) –
ρ	Density of the gas mixture kg m^{-3}
σ_{AB}	Lennard–Jones potential between molecules A and B –
$\varphi_{D,AB}$	Collision integral for diffusion Å
μ	Viscosity of the fluid Pa·s
ϕ_{ij}	Binary factor in mixture viscosity correlation –
ω_i	Mass fraction of species i

selectivity of palladium membrane is infinity, it is necessary to consider the mass transfer limitation caused by this effect.

Some methods of improving the mass transfer limitations have been previously studied; changing the operating conditions can effectively reduce the concentration polarization effect and improve the reactor performance [9,10]. Higher gas hourly space velocity (GHSV) favors the mass transfer in the reactor. Changing the reactor configuration can also improve the reactor performance. For instance, Mori et al. [8] concluded that increasing the linear flow rate by reducing the reactor radius can significantly reduce the concentration polarization effect in steam reforming of methane and improve the H_2 recovery and methane conversion. In addition, by adding baffles to the reactor configuration, the concentration polarization effect can also be reduced [6,8].

In this work, the mass transfer limitations of the pilot-scale palladium membrane module designed and assembled at WPI were examined [3]. The module has a shell and tube configuration (Fig. 1.), which contains 7 membrane tubes with 1050 cm^2 of membrane surface area. Hydrogen-enriched coal derived syngas (H_2, N_2, CO, CO_2, CH_4, H_2O) was introduced to the shell side and pure H_2 was permeated through the membrane tubes and collected from the tube side. Under the experimental conditions (450 °C, 12.6×10^5 Pa, and desulfurized syngas inflow rate per unit membrane area of 10.25 $m^3\ h^{-1}\ m_{mem}{}^{-2}$), the unit was able to produce 2.7 kg/day high purity (99.87–98%) H_2.

CFD simulation analysis is a powerful tool for the study of mass transfer phenomena [11]. Furthermore, simulation analysis provides important information for the module behavior under different operating conditions and different module configurations [12–15]. In a previous study, a simplified symmetrical model was developed and verified with the experimental data (Fig. 2.) in order to predict the H_2 recovery and yield under different operating conditions [3].

In the current study, in order to study the tube-to-tube difference for H_2 removal rate, an asymmetrical model, which is much closer to the reality, was developed.

In order to generate incentives for the future commercialization of this technology, further scale up of the module is required [16–18]. During the scaling up process, it is important to maintain the operating conditions unchanged [19]. The two main strategies for module scaling up are theoretical and empirical scale-up, which use simulation methods and lab-scale experimental parameters respectively. In this paper, simulation methods were used to scale-up the 7-tube module to a 19-tube module. Previously, Yang et al. reported a numerical method for membrane module scale-up, the module sensitivity of different design variables were studied and the difference between the scaled-up module and the lab scale module was addressed [20]. In the current work, different gas flow rates inside the module and mass transfer limitations under different flow rates were studied and compared with the original scaled module. The performance of each membrane tube at different locations was also studied.

2. Simulation methodology

2.1. Seven-tube module

The actual geometry of the membrane module was developed as shown in Fig. 3. The module shell has an inner diameter of 10.23 cm and a length of 96.52 cm excluding the inlet and outlets; the membrane tubes have an outer diameter of 1.27 cm and length of 38.1 cm. Each membrane tube has a surface area of 150 cm^2. The distance between the central tube and each outer layer tube is 3.33 cm. The design of the unit was based on technical constraints on the construction of the module. For instance, the diameters were determined based on the porous stainless steel (PSS) support configurations available for purchase, while the pitch was fixed in such a way that fittings and connections were feasible. The tube diameter and configuration here was a result of our previous studies [3]. Compared with the simplified geometry, the new geometry includes the second half of the membrane support, the manifold that collects H_2 from the seven tubes, and both retentate outlet and permeate outlet.

Only the shell side flow pattern was studied as no sweep gas was introduced to the tube side and the pressure was controlled at 101.3 kPa. The operating conditions in the simulation were the same as in the NCCC testing rig. The inlet mole percentages were H_2: 34%, N_2: 55%, CO: < 1%, CO_2: 10% and H_2O: ~0. The feed flow rate per unit membrane area was varied over the range 1.35 – 13.48 $m^3\ h^{-1}\ m_{mem}{}^{-2}$, the temperature was T = 450 °C, the shell side inlet pressure was 12.8 bar and the tube side pressure was fixed at 1.01325 bar.

Assumptions of the simulation model include:

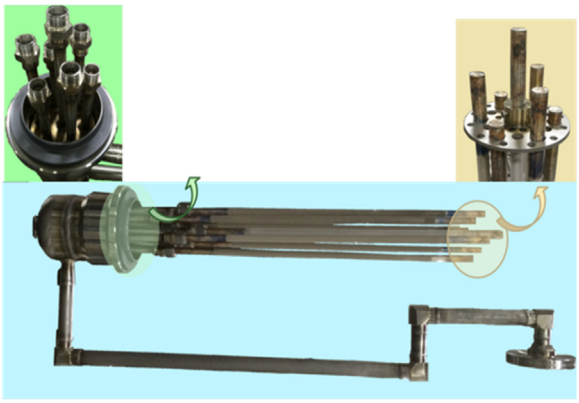

Fig. 1. Membrane module configuration.