ISCRE25
FLORENCE 2018
BRIDGING
SCIENCE & TECHNOLOGY
The 25th International Symposium
on Chemical Reaction Engineering
20-23 May 2018
Florence, Italy
www.aidic.it/iscre25

Hydrogen production in a packed bed membrane reactor: a preliminary analysis of mass dispersion in solid foams

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Highlights

- A model was developed for a membrane reactor packed with a solid foam.
- Mass dispersion in solid foams was described by fitting H_2 separation experiments.
- The results were used to predict the behavior of a membrane reactor.

1. Introduction

Membrane reactors for hydrogen production are receiving significant attention. Three components of the membrane reactor deserve particular attention: the hydrogen permeable membrane, the catalyst, and the catalystõs mechanical support. With regards to the membrane, it is now well known that Pd-based membranes present a high permeability to hydrogen along with an infinite selectivity. Ongoing research is focused particularly on the choice of alloy that could reduce inhibition by competitive adsorption without significant reductions in permeability and selectivity [1-2]. The issue of selecting an appropriate catalyst arises because one of the main advantages of membrane reactors is the possibility of operating at lower temperatures while reaching comparable or higher methane conversion values compared to the traditional hydrogen-producing processes. In the case of steam reforming of light hydrocarbons, research efforts have successfully identified catalysts that allow high reaction rates [3-4]. Finally, the mechanical support of the catalyst must be chosen in order to reduce pressure losses along the reactor and enhance heat and mass transfer. To attain this, the use of solid foams has been proposed [4-5]; however, information regarding mass dispersion within these foams is still lacking.

In recent works by some of the authors of this study, an isothermal membrane reactor has been thoroughly modeled and analyzed. It has been shown that there exists an optimal operating pressure, above which only a negligible increase in hydrogen recovery can be obtained [6]. The main assumption that has been made so far is that of a negligible hydrogen pressure in the permeate side. A preliminary analysis has shown that the results obtained are qualitatively valid even when removing this assumption, but a quantitative analysis and an assessment of the influence of a mass balance in the empty permeate side on the optimal pressure is still lacking. The present work is an extension of the membrane reactor model developed and presented in [6-8], with the objective of characterizing mass dispersion in solid foams. To this end, hydrogen separation experiments have been carried out in a membrane module packed with solid foams and fed with nitrogen and hydrogen mixtures. Results have been analyzed through the transport-permeation model developed in [7], here solved numerically using a finite element commercial software.

2. Methods

A 2D isothermal model was developed and solved through the finite element method using COMSOL Multiphysics. A complete description and explanation of the equations used to describe both a packed separator [7] and a membrane steam reforming reactor [6], under the assumption of negligible hydrogen pressure in the permeate, is reported in previous works. In both cases the packing is included in the volume

between two co-axial cylinders and the hydrogen-permeable membrane is placed on the outer wall of the innermost tube. The dimensionless form of the model equations allow the identification of three main parameters that determine the performance of the system: the Damkholer number, which represents the ratio between the rate of reaction and the rate of convection; the Peclet number (Pe), representing the ratio between the rates of convection and radial diffusion; and a parameter ɔ representing the ratio between the rates of permeation across the membrane and convection. It has been found that when the rate of reaction is not limiting, it is possible to determine the optimal ratio between inner and outer diameters of the reactor based on the extent of a reaction boundary layer, whose thickness results from the relative magnitude of the resistance to mass transport offered by dispersion in the packed bed and permeation across the membrane [8]. The permeance of the membrane depends on its thickness and composition and may be determined from permeation tests; on the other hand mass dispersion depends on the geometric characteristics of the packing, on the molecular diffusion coefficient of the different species, and on the velocity and pressure profiles in the packed bed. In order to evaluate mass dispersion in the solid foams, permeation tests were carried out in a membrane module packed with SiC foams. The main characteristics of the setup are described in Table 1 and a summary of the operating conditions is reported in Table 2. The model has been used to fit an effective Pe number in each condition. These values were then compared to the molecular Pe number. The results were used to predict the behavior of a membrane reactor for the production of hydrogen from methane steam reforming.

Table 1. Summary of the main characteristics of the experimental setup

Foam		**Reactor Characteristics**			
Material	Void fraction	Inner diameter [m]	Outer diameter [m]	Length [m]	Sweep configuration
SiC	0.85	0.014	0.0413	0.5	Counter-current

Table 2. Summary of experiments

T [°C]	**P retentate [bar]**	**P permeate [bar]**	**Total inlet flow rate [cc/min]**	**Inlet composition [mol fraction] H_2/N_2**	**Sweep flow rate [cc/min]**	**H_2 permeate outlet flow rate [cc/min]**
~450	4-9	1.5	3000-7500	0.34/0.66-0.86/0.14	0-2000	230-5200

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Keywords
Membrane reactor; modeling; solid foam; mass dispersion