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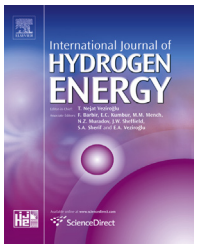

Modelling and simulation in conventional fixed-bed and fixed-bed membrane reactors for the steam reforming of methane

Jornandes Dias Silva [a,*], *Cesar Augusto Moraes de Abreu* [b,1]

[a] *Polytechnic School – UPE, Laboratory of Environmental and Energetic Technology, Rua Benfica - 455, Madalena, Cep: 50750-470, Recife, PE, Brazil*

[b] *Department of Chemical Engineering, Federal University of Pernambuco (UFPE), R. Prof. Artur de Sá, 50740-521, Recife, PE, Brazil*

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ABSTRACT

This paper presents a dynamics mathematical model to simulate the steam reforming of methane that take place in conventional fixed bed reactor (FBR) as well in fixed bed membrane reactor (FBMR) with steam added both with co-current mode. The model covers all aspects of main chemical kinetics, heat and mass phenomena in the membrane reactor with hydrogen permeation in radial direction across a Pd-based membrane. Firstly, a dynamics study was made for describing that temperatures of gaseous and solid phases reach to steady-state as well as molar flow rates. The effect several parameters including the axial position (z) divided by the reactor length L_z, reaction temperature and hydrogen partial pressure ($P_{H_2} = P_{pz}$) in permeation side were investigated. The conversion of methane is significantly enhanced by the partial removal of hydrogen from the reaction zone as a result of diffusion through the Pd-based membrane. Simulation results showed that a conversion from 99.85% could be achieved in a FBMR at reaction temperature of 600 °C relative to a conversion from 88.87% to 950 °C in a FBR. Besides, results showed that the yield of H_2 reached to level from 1.548 (dynamics-state) and 1.626 (steady-state) in a FBMR at reaction temperature of 550 °C while the yield of H_2 achieved to level from 1.261 (dynamics-state) and 1.445 (steady-state) in a FBR at reaction temperature of 725 °C.

Introduction

The process intensification strategy using membrane reactors can lead to the development and the re-design of more compact and efficient new processes that allow better exploitation of raw materials, lower energy consumption and plant volume reduction. Catalytic steam reforming of methane (CSRM) can be carried out in a fixed-bed membrane reactor (FBMR) as a way to intensify the hydrogen production [1].

* *Corresponding author.*
E-mail addresses: jornandesdias@poli.br (J.D. Silva), cesar@ufpe.br (C.A.M. de Abreu).
[1] Tel.: +55 81 2126 8901.

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The world consumption of hydrogen per year (137 million kg of hydrogen per day) is about 50 million ton and its request is rapidly increasing [2]. CSRM is currently a well-established technology and has been the most important industrial process for the production of hydrogen and/or synthesis gas (syngas) [3]. As the main production route of hydrogen on the industrial scale, their different aspects have been investigated by many authors [4–6].

In the CRSM process, methane reacts with steam to produce a mixture of hydrogen, carbon monoxide and carbon dioxide [7,8]. The production of hydrogen in a FBMR is highlighted by changing the reaction equilibrium toward the product side through effective pressure difference induced by the continuous removal of hydrogen over the membrane [4,6,9]. Hydrogen was produced in a FBMR from biogas based on the evidences of the dry methane reforming process conducted with a nickel catalyst at 700 K under atmospheric pressure [10]. The complexity of optimizing the operating conditions of a FBMR is significant due to the subjacent phenomena occurring within this type of reactor [11,12]. Reforming reactions are highly endothermic and can be performed with active catalysts (Ni, Rh) at high temperature, high pressure and steam-carbon ratio varying between 1.4 and 4.

Several studies have been carried out with CSRM process to separate H_2 from a gaseous mixture by using a Pd-based FBMR [13]. The commercial separation of hydrogen through metallic membranes is mainly focused on palladium alloys membrane. The production of hydrogen in the reaction zone of the FBMR permits the hydrogen permeation through hydrogen-selective membrane [14–16].

The purpose of this paper is to evaluate the performance of the conventional fixed-bed reactor (FBR) and fixed-bed membrane reactor (FBMR) used to simulate the CSRM. Based on the energy and mass balances of chemical species, a system of partial differential equation (PDE) was formulated for describing governing equations of the energy and mass balances. Simulations presented from the model equations provide the evaluation of the CSRM process in FBR and FBMR reactors with a nickel catalyst. The performance of above reactors was studied in terms of temperature profiles, methane conversion at different temperatures, hydrogen production and the selectivity of H_2, CO, CO_2 and CH_4.

Physical model

A schematic configuration of the proposed system used in the development of the mathematical model is shown in Fig. 1. The system consists of two concentric tubes (including the conventional FBR and FBMR reactor), where the external tube is constructed in steel and the internal tube constitutes the thin Pd-based membrane. The catalyst is placed in the annular space between the two tubes, forming the fixed bed (external reaction zone). During the operation, the reaction zone (fixed bed) is continuously fed with a gas mixture ($H_2O/CH_4 = 2.13$) inlet, while a pressure reduction is applied inside the internal tube (internal permeation zone) through which an inert carrier gas flow to promote the transport of hydrogen flux as from the membrane until at the permeation zone outlet.

The base data used for the Geometric conditions, Catalyst properties and Inlet operating conditions are illustrated in Table 1 in relation to the FBR and FBMR reactors. The temperature range (500–950 °C) of the numerical experiments was chosen to cover all possible conditions that may occur in this study in the FBR and FBMR reactors.

Chemical components of the reforming process

The catalytic steam reforming process includes as reaction PSRM, WGS and OSRM [3], equating as follow:

$$CH_4 + H_2O \leftrightarrow CO + 3H_2; \quad \Delta H^0_{298K} = 205.8 \text{ kJ mol}^{-1} \text{ (PSRM)} \qquad (1)$$

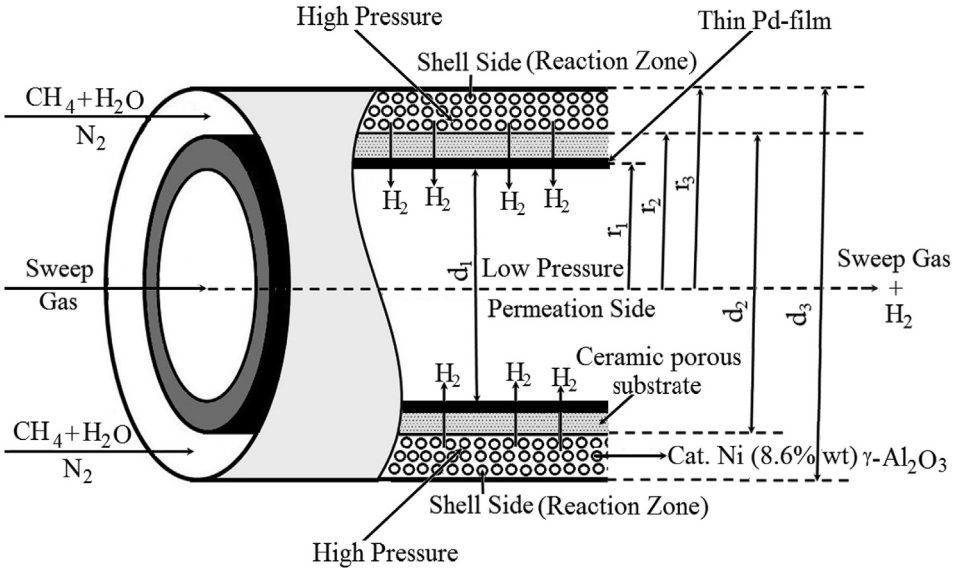

Fig. 1 – Schematic diagram of the FBR and FBMR reactors.