MODELING OF METHANE STEAM REFORMING IN A PALLADIUM MEMBRANE REACTOR

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***Abstract*— A mathematical model of a membrane reactor used for methane steam reforming was developed to simulate and compare the maximum yields and operating conditions in the reactor with that in a conventional fixed bed reactor. The methane steam reforming reaction has been investigated from a modeling viewpoint, considering the effect of different parameters on methane conversion and H_2/CO ratio. Results show that the membrane reactor presents higher methane conversion yield and can be operated at milder conditions than the fixed bed reactor. Control of the H_2/CO ratio is possible in the membrane reactor making the technology more suitable to be used in connection to gas-to-liquid processes (GTL).**

***Keywords*— Steam Reforming, Methane, Membrane Reactors**

I. INTRODUCTION

Methane steam reforming is one of the most important chemical processes for the production of syngas and hydrogen. In recent years, the abundant availability of natural gas and the increasing demand of hydrogen have led to high interest to further develop this process increasing the yield of syngas (Shu *et al.*, 1994; Rostrup-Nielsen, 1984).

Methane syngas reforming involves two reversible reactions: reforming and water gas shift. The first is endothermic and limited by thermodynamic equilibrium. Therefore, the development of a membrane-based separation process can open-up the possibility of increasing the conversion of the reforming process. As hydrogen is selectively removed from the reactor, the chemical equilibrium of the reactions is shifted towards the products, resulting in an increase in the conversion of methane to hydrogen and carbon monoxide. As an additional advantage, the membrane reactor offers the possibility of supplying hydrogen with the same conversion degree in higher purity, than that supplied by the conventional reactor, under less severe operational conditions. Methane stream reforming in a membrane reactor becomes a transfer-limited reaction related with membrane porosity and diffusivity, rather than an equilibrium-limited reaction (Assaf *et al.*, 1998).

The development of palladium-based membrane separation process has opened up a new possibility to enhance membrane steam reforming conversion. Some researchers have reported methane conversions as high as 96% with Pd-membranes in isothermal operation (Shu *et al.*, 1994; Oertel *et al.*, 1987; Adris *et al.*, 1991; Lin *et al.*, 2003). New improvements are been done in membrane materials and structures, which supports the selective metal in porous glass and porous alumina ceramic substrate, and in nanostructured carbides.

This work presents the mathematical modeling of a one-dimensional, non-isothermal membrane reactor operating at steady-state, comparing and discussing reactor and yield improvements with results from a conventional fixed bed reactor.

II. MEMBRANE REACTOR

In methane steam reforming, the catalytic fixed-bed reactor is fed with a gas mixture of CH_4 and H_2O in a molar ratio from 1:3 to 1:4. Commercial catalyst is composed of nickel supported in alumina and the reactor is composed of vertical tubes (between 10 and 900 tubes inside the reactor) with internal diameters from 7 to 16 cm and lengths from 6 to 12 m, inserted in a radiant furnace chamber. The feed conditions are about 600°C and from 1.5 to 3.0 MPa. The maximum temperature that the reactor can support is limited by the metallurgical limitations of the tubes, since at higher temperatures the metal tubes can creep under stress.

The membrane reactor configuration is quite simple and consists of an external steel tube (shell) with an inner membrane wall tube were the sweep gas flows (permeation zone). Methane and steam are continuously fed into the catalytic zone, and a sweep gas, usually nitrogen is introduced on the permeate zone to drag the permeated hydrogen. A scheme of the reactor is shown in Fig. 1.

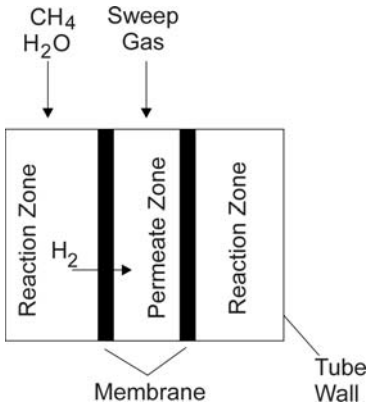

Fig. 1. Scheme of the membrane reactor.