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Demonstration of a scaled-down autothermal membrane methane reformer for hydrogen generation

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ABSTRACT

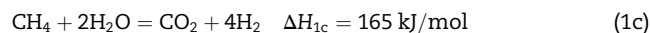
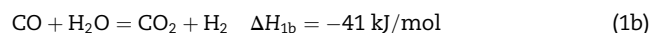
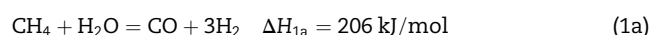
A novel concept for hydrogen generation by methane steam reforming in a thermally coupled catalytic fixed bed membrane reformer is experimentally demonstrated. The reactor, built from three concentric compartments, indirectly couples the endothermic methane steam reforming with the exothermic methane oxidation, while hydrogen is separated by a permselective Pd membrane. The study focuses on the determination of the key operation parameters and understanding their influence on the reactor performance. It has been shown that the reactor performance is mainly defined by the dimensionless ratio of the methane steam reforming feed flow rate to the hydrogen maximal membrane flow rate and by the ratio of the oxidation and steam reforming methane feed flow rates.

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1. Introduction

Hydrogen as an energy carrier alternative to fossil fuels has attracted much attention during last decades, especially as a fuel for energy efficient and environmentally friendly polymer electrolyte fuel cells. Yet, the implementation of hydrogen-based energy generation technologies is hindered by the drawbacks in its purification and transportation. On-site small-to-medium scale hydrogen generation using a membrane reformer has the potential to overcome the purification and transportation drawbacks. However, while Pd-based membranes allow for highly selective hydrogen separation suitable for fuel cell applications, the endothermicity of the reforming process still remains one of the major factors significantly lowering the efficiency of the membrane reformer. Steam reforming of methane (Eqs. (1a)–

(1c)) is of notable interest, since there are large resources of natural gas in the world.



Hydrogen generation by steam reforming of methane using membrane reactors has been extensively investigated numerically [1–7] and experimentally [3,8–14]. Most experimental studies have used external heat sources and very few works have addressed more energetically efficient coupling of exothermic and endothermic reactions, which is required for mobile or stationary units that are independent of the electricity grid. In reviewing the subject the aspects of heat

Abbreviations: FBMR, fluidized bed membrane reactor; MOx, methane oxidation; MSR, methane steam reforming; PBMR, packed bed membrane reactor; Ox, oxidation; SR, steam reforming.

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Nomenclature	
A_{H_2}	pre-exponent, mol/(m ² s bar ^{0.5})
CR	coupling ratio
d	wall thickness, mm
D	diameter, mm
E_a	activation energy, kJ/mol
f	conversion
F	molar flow rate, mol/s
HR	hydrogen recovery
ΔH	heat of reaction, kJ/mol
J_{H_2}	hydrogen outlet flux, Ncm ³ /(cm ² min)
L	reactor length, m
L_{mf}^{SR}	methane steam reforming loading
p	partial pressure, bar
P	total pressure, bar
Q	volumetric flow rate, Ncm ³ /min
r	radius
R_g	gas constant
S^M	membrane area, m ²
SV	space velocity, h ⁻¹
t	time, h
T	temperature, °C
V	reactor volume, m ³
y	molar fraction
Y_{CO}	CO selectivity
Y_{CO_2}	CO ₂ selectivity
Y_{H_2}	hydrogen-separation efficiency
z	axial reactor coordinate, m
<i>Greek symbols</i>	
ε	catalytic bed porosity, $\varepsilon = 0.5$
θ	dimensionless temperature
<i>Subscripts</i>	
eff	effluent
f	feed
m	methane
t	total
w	wall
<i>Superscripts</i>	
E	equilibrium
M	membrane compartment
ME	membrane equilibrium
Ox	oxidation compartment
SR	steam reforming compartment

integration are emphasized, in addition to discussing the aspects of reactor and membrane type and geometry.

Reactor selection: A packed bed membrane reactor (PBMR) is commonly employed [1,2,5–7,8,10,12,14]; a fluidized bed membrane reactor (FBMR) has been studied in several works [3,4,9,11,13]. Though the use of the FBMR provides good mass and heat transfer characteristics, the PBMR is more suitable for mobile hydrogen generation applications due to its compactness.

Membrane selection: Since the membrane reformer performance is limited by separation capability, maximizing the membrane permeability is one of the important issues in the development of membrane reactor technologies. Membranes composed of a Pd–Ag thin layer on a ceramic or stainless steel porous supports [10,12,14] are very promising, since they exhibit higher permeabilities than that of Pd–Ag foil membranes. Both membrane types show almost complete selectivity to hydrogen separation and are available commercially. Since the Pd cost is prohibitive, other substitutes with good selectivity for hydrogen separation, like carbon [15,16], silica [17,18], and zeolitic [19,20] hydrogen-separation membranes are extensively investigated. However, their separation selectivities and permeabilities are still not satisfactory.

Externally heated systems: Given that steam reforming is highly endothermic, the membrane reformer should be coupled to a heat source, which in experimental works is commonly conducted by electrical heaters [3,8–10,12,14]. Several such works, of hydrogen production by methane steam reforming (MSR) are described below, before addressing the coupling endothermic and exothermic reactions. Patil et al. [9] have reported the experimental demonstration of an oven-heated FBMR for hydrogen production by MSR incorporating one Pd-based membrane, operating at 550–650 °C, and

generating hydrogen flow equivalent to a fuel cell power output of 10 W. In more recent work [3], Patil et al. have experimentally demonstrated an FBMR equipped with 10 Pd-based membranes that showed almost complete methane conversion at 650 °C and hydrogen flow, equivalent to a fuel cell power of 50–100 W. However, the required heat was still supplied by electrical furnaces. In an oven-heated PBMR equipped with one Pd–Ag membrane Gallucci et al. [8] achieved 70% conversion at 450 °C; while other groups [10,12,14] demonstrated complete methane conversion at 550 °C in a PBMR with a high-permeable membranes; the required temperatures were set up using electric furnaces.

Autothermal reforming: Coupling MSR to an exothermic reaction (e.g. methane oxidation) is a prerequisite if it is aimed to design an independent system. This can be performed directly, by performing endothermic and exothermic reactions over the same catalyst bed (e.g. autothermal steam reforming (ATR) [11,13]), or indirectly, using heat exchange reactor [7]. In ATR oxygen (air) is added to the catalytic bed to oxidize part of methane and to provide the heat required for the endothermic MSR. Catalyst selection is one of the drawbacks of the direct coupling approach, since the catalyst should favor both exothermic and endothermic reactions, as well as withstand the hotspots emerging due to the exothermic reaction. Another problem is temperature distribution along the reactor (see below). Also, when air is used as an oxygen source for ATR in a membrane reactor, nitrogen dilutes the reactive stream, suppressing the hydrogen separation due to diminished hydrogen partial pressure gradient. Since using pure oxygen is not a practical approach, this appears to be an inherent disadvantage of the direct coupling approach. In addition, using air for supplying oxygen will increase the reactor dimensions; air also has to be compressed to the pressures required to drive hydrogen separation.

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