



Experimental and exergy evaluation of ethanol catalytic steam reforming in a membrane reactor



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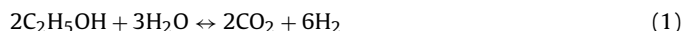
ABSTRACT

The application of exergy analysis in the evaluation of the ethanol steam reforming (ESR) process in a catalytic membrane reactor (CMR) was presented. ESR experiments were performed at $T=873\text{--}923\text{ K}$ and $P=4\text{--}12\text{ bar}$ in a CMR containing Pd–Ag membranes sandwiched by Pd–Rh/CeO₂ catalyst, aiming to produce fuel cell grade pure hydrogen. The effect of the operating conditions on the pure hydrogen production rate, hydrogen yield and recovery, exergy efficiency, and thermodynamic losses was investigated. Total hydrogen yield of 3.5 mol H₂ permeated per mol ethanol in feed with maximum hydrogen recuperation of 90% was measured at 923 K and 12 bar. The highest amount of exergy was destructed via heat losses and the retentate gas stream. Exergy efficiency up to around 50% was reached in the case of the insulated reactor at 12 bar and 923 K. Exergy efficiency placed between 70–90% in the case of recovery of the retentate gas in an insulated reactor. It was concluded that operating at the highest pressure, the lowest S/C ratio, and 923 K gives the best exergy efficiency.

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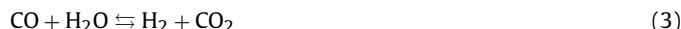
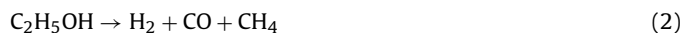
1. Introduction

As an alternative to fossil fuels, hydrogen is considered as a clean energy carrier that can be combusted similar to the conventional carbonaceous fuels or be converted to electricity by fuel cells [1]. The use of renewable biofuels such as bio-ethanol as a source of hydrogen is highly beneficial due to the higher H/C ratio, lower volatility and toxicity, and higher safety of storage that distinguishes ethanol over other substrates. Bio-ethanol is cheaply and easily obtained from biomass and organic waste and can be used directly in catalytic steam reforming processes to produce hydrogen since it contains large amounts of water [2]. Among the reforming processes, steam reforming of bio-ethanol (Eq. (1)) delivers the highest amount of hydrogen per mole of converted bio-ethanol [3].



Huge amount of works has been reported in the literature on catalytic ethanol steam reforming (ESR) specially on the experimental

investigations aiming for hydrogen generation using a variety of catalysts in different reactor configurations [4–8]. The distinctive properties of noble metals such as high activity, hindering carbon from depositing on the catalyst active sites, and durability and robustness during the ESR process have attracted the attention of a lot of research groups towards such catalysts [9,10]. Further, the formation of undesired chemical species is minor or zero when noble metal based catalysts are used for the ESR process [6,9]. The main products of ESR over the Pd–Rh/CeO₂ catalyst are CH₄, CO₂, CO and H₂, which are obtained via following reaction pathways [10–12]:



Eqs. (2), (3) and (4) represent ethanol decomposition, water gas shift reaction (WGS), and methane steam reforming (MSR) reactions, respectively. According to Idriss et al. [10], at $T > 800\text{ K}$, the only present non-condensable products of the ethanol steam reforming over the 0.5 wt % Pd–0.5 wt % Rh/CeO₂ are CO, CO₂, CH₄, and H₂. The experiments were performed in a membrane reactor with selective Pd-based metallic membranes in which the production and separation of hydrogen took place simultaneously. The benefits of catalytic membrane reactors (CMRs) such as simultaneous generation and separation of hydrogen, cost reduction,

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Nomenclature

EX	rate of exergy, W
ex	specific exergy, J mol ⁻¹
h	specific enthalpy, J mol ⁻¹ K ⁻¹
s	specific entropy, J mol ⁻¹
x	molar fraction in gas phase
T	temperature, K
R	universal gas constant, J mol ⁻¹ K ⁻¹
S/C	steam to carbon ratio
FF	flow rate of fuel, mol s ⁻¹
F	molar flow rate, mol s ⁻¹
\dot{m}	mass flow rate, kg s ⁻¹
LHV	lower heating value, J kg ⁻¹
\dot{Q}	heat loss rate, W
\dot{W}_{pump}	power of the pump
W_{el}	electrical power, W
Y_{H_2}	hydrogen yield
R_{H_2}	hydrogen recovery

Subscript

ex	exergy
in	inlet stream
out	outlet stream
fuel	ethanol and water mixture
H2.perm	permeated hydrogen
EtOH	ethanol
H2.total	total hydrogen production

Greek letters

η	efficiency
ε	standard chemical exergy, J mol ⁻¹

simplicity of the design, and reforming reactions promotion beyond the equilibrium limits (the shift effect) are well known and repeatedly reported in the literature [13–16].

According to the open literature, there are a few reported studies on exergy efficiency evaluation of ethanol steam reforming systems for hydrogen production. The term exergy is defined as the maximum work that can be obtained theoretically from a system interacting with the source environment to equilibrium [17].

The main difference between energy (thermal) efficiency and exergy efficiency lies in the consideration of the thermodynamic state of every single component, which results in an exact understanding of the available amount of work, together with the unavoidable irreversibility during a process [18]. Considering the conservation of mass and energy together, exergy analysis is a powerful tool to investigate the imperfections of single components of a system, to obtain a clearer understanding of the local irreversibility and the effect of thermodynamic factors on the performance of an energy system [19,20].

As reported in the literature, Kalinci et al. [21] studied the production of hydrogen via a gasification-boiler system based on experimental data taken from the literature using different types of biomass. They found the maximum exergy efficiency to be about 12%. An exergy analysis of the biological hydrogen production from biomass was done by Modarresi et al. [22] based on experimental results. They reported exergy efficiencies of 36–45%, depending on the process configuration. For reforming processes, Simpson et al. [1] modelled the methane steam reforming process and both irreversible chemical reactions and heat losses were identified as the main source of exergy destruction, whereas exhaust gases contained large amounts of chemical exergy. Casas-Ledón et al. [23] studied hydrogen production from ESR based on the first and

second laws of thermodynamics. They evaluated the exergy efficiency of the system experimentally at different operating conditions (pressure, temperature, and S/C ratio) considering the unused and destructed exergy during the ESR process. They concluded that the exergy efficiency of the ESR system was a function of temperature and S/C ratio, while no effect of pressure on exergy efficiency was observed. A comprehensive exergy analysis of the different types of ethanol reforming processes (ESR, POX and ATR) based on a model in Aspen Plus was performed by Khila et al. [19]. The same formulation as Casas-Ledón et al. [23] was used by Khila et al., and they used Aspen Plus software to calculate the exergy of the inlet and outlet streams at selected operational conditions, according to hydrogen production per mole of inlet ethanol. An exergy efficiency of 70% was claimed for the ESR process, considering total hydrogen production via ESR as the main product. In another study, Tippawan et al. [24] employed the first and second law of thermodynamics to evaluate a modelled ethanol reforming system in connection with a solid oxide fuel cell (SOFC) with a similar formulation as Casas-Ledón et al. [23] and Khila et al. [19]. They studied ESR, partial oxidation (POX), and autothermal reforming (ATR) processes as the reforming sections for hydrogen production, and the best efficiency of the system (reforming + SOFC) was stated equal to 60% when ESR was used as the reformer unit. Finally, Hedayati et al. [25] reported exergy evaluation of the ESR process in a staged membrane reactor based on experimental results. They considered only pure hydrogen as the desired product. It was reported that a big share of exergy is destroyed due to the irreversibility of reforming reactions and heat losses.

In this work, we present energy and exergy analysis of the ESR process in a catalytic membrane reactor (CMR) containing Pd–Ag membranes sandwiched by Pd–Rh/CeO₂ catalyst to produce pure hydrogen (no sweep gas). The exergy evaluation of the system is based on the experimental results. The novelty of this work lies in the application of exergy analysis to evaluate the ESR process in a packed bed CMR configuration based on experimental results and observations.

2. Material and methods

2.1. Experimental

The Pd–Rh/CeO₂ catalyst (0.5% Pd–0.5% Rh) was deposited over cordierite pellets of about 1–3 mm following the procedure described by López et al. [26]. The laboratory setup used for the ESR experiments (fuel reformer) consisted essentially of a fuel tank, a liquid pump, a CMR, a pressure transducer and a condenser. A detailed description of the reformer setup can be found in [25]. The scheme of the experimental setup and the CMR is presented in Fig. 1.

The heating plate was controlled by an electronic controller (Fuji PXR4), provided the temperature measurement registered by a k-type thermocouple which was in close contact with the reactor wall. A HPLC pump (Knauer) was used to pump the water–ethanol mixture (fuel) and to keep the pressure. A backpressure regulator (Swagelok) adjusted the retentate pressure. No pressure regulation was implemented on the permeate side (pure hydrogen outlet), so the permeate side pressure was kept automatically at ambient pressure. Besides, no sweep gas was used so pure hydrogen was obtained at atmospheric pressure.

A commercial membrane reactor provided by REB Research & Consulting [27] was used. The reactor was 10 in. tall and 1 in. in diameter. There were four dead-end Pd–Ag membrane tubes inside the reactor; each one 3 in. tall and 1/8 in. diameter in order to separate hydrogen. The membrane tubes consisted of Pd–Ag (30 μm layer) supported on porous stainless steel (PSS) layer,

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