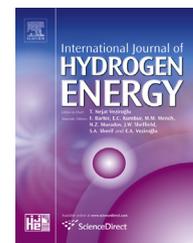


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Hydrogen production from methane and solar energy – Process evaluations and comparison studies

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ABSTRACT

Three conventional and novel hydrogen and liquid fuel production schemes, i.e. steam methane reforming (SMR), solar SMR, and hybrid solar-redox processes are investigated in the current study. H₂ (and liquid fuel) productivity, energy conversion efficiency, and associated CO₂ emissions are evaluated based on a consistent set of process conditions and assumptions. The conventional SMR is estimated to be 68.7% efficient (HHV) with 90% CO₂ capture. Integration of solar energy with methane in solar SMR and hybrid solar-redox processes is estimated to result in up to 85% reduction in life-cycle CO₂ emission for hydrogen production as well as 99–122% methane to fuel conversion efficiency. Compared to the reforming-based schemes, the hybrid solar-redox process offers flexibility and 6.5–8% higher equivalent efficiency for liquid fuel and hydrogen co-production. While a number of operational parameters such as solar absorption efficiency, steam to methane ratio, operating pressure, and steam conversion can affect the process performances, solar energy integrated methane conversion processes have the potential to be efficient and environmentally friendly for hydrogen (and liquid fuel) production.

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Introduction

Hydrogen is an intrinsically clean energy carrier and the enabling factor for the “hydrogen economy” [1]. Besides being combusted for heat generation, hydrogen can be converted into electricity in fuel cells at high efficiencies or used as a building-block chemical for oil refining, Fischer–Tropsch (F–T) synthesis, and ammonia and methanol productions [2]. As a secondary energy source, hydrogen is produced from hydrocarbons and/or water via reforming, electrolysis, or

thermolysis [3]. These process schemes tend to be energy and/or capital intensive. In addition, H₂ production from fossil fuels can lead to significant CO₂ emission in absence of energy-intensive carbon capture and sequestration steps. Currently, over 90% of the world’s total hydrogen production is derived from methane, a primary component of natural gas, via the well-known steam methane reforming (SMR) process [4,5]. In the foreseeable future, natural gas will continue to be the major feedstock for hydrogen production [6].

A typical SMR process is composed of a series of process steps including pretreatment, reforming, water–gas-shift

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(WGS), and product purifications. In the pretreatment step, impurities in natural gas such as sulfur compounds are removed to avoid catalyst poisoning. Subsequently, methane reforming is carried out by catalytically reacting methane with steam at high temperatures to produce syngas:



The overall reaction is endothermic. Over 75% methane conversion is typically achieved under temperatures ranging between 800 and 900 °C and pressures up to 35 atm [7].

The product gas from the reformer, a mixture of CO, CO₂, CH₄, H₂, and H₂O, is then introduced into a WGS reactor system. In the WGS system, CO is reacted with H₂O in syngas to produce additional H₂ and CO₂ by the WGS reaction (Reaction 2). The WGS reaction is moderately exothermic. As a result, the equilibrium favors H₂ production at low reaction temperatures [8]. Kinetically, however, a high reaction temperature is desired. To address this challenge, typical WGS system is comprised of a high temperature shift reactor for high reaction rates and a low temperature shift reactor for high CO conversions. After condensing out steam, the exit gas from the WGS system is predominantly hydrogen and CO₂ (~20%) as well as some residual CO and methane. To produce a high quality hydrogen product, the gas stream is further purified by absorption based acid gas removal (AGR) and pressure swing adsorption (PSA) operations. The purity of final H₂ product is typically 99.9% or higher [7,9].

As a benchmark process for hydrogen production from natural gas, the SMR process generates a significant amount of CO₂ by-product [10–12]. With the growing concerns over global warming, CO₂ capture and storage have been recognized as a necessity for SMR operations [13]. In commercial SMR processes, CO₂ removal is usually achieved by absorption based AGR processes using chemical or physical solvents such as monoethanolamine (MEA), methyldiethanolamine (MDEA), and dimethyl ethers of polyethylene glycol [7,14]. A large amount of steam is required for CO₂ desorption in these absorption based processes, lowering the overall process efficiency [6,14,15]. In addition, around 20–30% methane is typically combusted as a fuel in the reformer furnace to provide the heat for the endothermic reforming step, resulting in additional energy penalty. According to the studies performed by Simpson and Lutz [4], the majority (~30%) of the exergy destruction in the SMR process is attributable to the high irreversibility of combustion and heat transfer between reformer and furnace. Another challenge associated with methane combustion resides in the associated CO₂ emissions. The combustion flue gas contains CO₂ diluted by nitrogen in the air. As a result, CO₂ capture from the combustion exhaust gas, which has a low CO₂ partial pressure, can be particularly energy consuming [13,16]. Feng et al. investigated CO₂ capture on SMR via a number of configurations including burning H₂ instead of CH₄ in the furnace, using pure oxygen instead of air for fuel gas combustion, applying a pre-reformer to reduce the usage of pure oxygen, employing a H₂-membrane in the reformer to separate H₂ [13]. With a base case efficiency of 70% (exergy) in a traditional SMR system, the efficiency drop from

these CO₂ capture steps are reported to range from 2% to 13%. To summarize, a hydrogen generation scheme with higher efficiency and lower emission is highly desired.

An alternative approach to reduce CO₂ emissions from methane reforming is to couple SMR with concentrated solar energy [17], since the latter is a clean energy source capable of providing heat directly to the reaction sites [3,18]. Becker and Funken investigated various cases for operating a solar reforming plant [19]. Although they share many similarities, the key differences between the solar SMR and conventional SMR are (i) use of solar heat for endothermic reforming reactor instead of CH₄ combustion; (ii) recycle of the off-gas from the PSA to the reformer. Möller et al. [8] performed techno-economic studies on the solar steam methane reforming process. In their model, natural gas consumption is reduced by up to 40% and H₂ product is 20% more expensive compared to the conventional SMR process. Besides theoretical studies, up to 500 kW large scale demonstrations of solar reforming have also been performed [17,20–22]. CO₂ emission is reduced in the solar SMR process by avoiding CH₄ combustion, which accounts for ~20% total CO₂ emissions from conventional SMR. The challenges of solar SMR reside in reactor design, solar facility cost and stability of the solar source. In addition, the complexity of WGS operations and acid gas removal steps impose energy penalties to the overall energy conversion scheme.

In addition to steam methane reforming, a so called two-step water-splitting or hybrid solar-redox process has been proposed to produce hydrogen with redox reaction of metal oxides. In such a process, methane and steam are introduced as the feedstock in two redox steps [18,23]:



In the first step, methane is oxidized by the metal oxides to generate syngas, which can be used for methanol or F–T synthesis. The reduced metal oxides are then reacted with steam in the second step to generate H₂. Compared to SMR, the above mentioned hybrid solar-redox process produces F–T ready syngas and pure H₂ with lower CO₂ emission and simpler product separation steps. Such a scheme may offer a competitive alternative to conventional technologies.

A number of research efforts have been directed towards finding effective metal oxides for the two-step water-splitting process. Among the various options, iron oxide is abundant and relatively cheap [24–26]. However, limited experimental data is available with syngas yield and steam conversions generally limited to 25% and 30%, respectively [26–28]. Besides pure iron oxides, iron based mixed metal oxides such as ZrO₂-supported Ni(II)-ferrite [29], Cu–Cr–FeO_x [30], Ni–Cr–FeO_x [31], ZrO₂-supported Co(II)-ferrites [32], have also been tested. Up to 90% methane conversions have been reported for some of these mixed oxides. However, steam to hydrogen conversions are generally limited to about 25%. Kodama et al. [33] reported WO₃/W redox system to be a potential oxygen carrier for two-step methane reforming. While a relatively high methane conversion of 70% and a CO selectivity of 86% are reported, steam to H₂ conversion is limited to 30%. He et al.

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