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## Development of a stand-alone steam methane reformer for on-site hydrogen production



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### ABSTRACT

A small, stationary reformer designed as a stand-alone and self-sustaining type was developed for on-site hydrogen ( $H_2$ ) production. We created a compact reformer to produce  $H_2$  at a rate of  $1 \text{ Nm}^3/\text{h}$  using the previously reported reaction kinetics of steam methane reforming (SMR). Both catalysts for the compact reformer - i.e., 15 wt% and 20 wt% Ni/ $\gamma$ - $Al_2O_3$  - showed good activity, with  $CH_4$  conversion exceeding 90% at  $655 \text{ }^\circ\text{C}$  and a contact time of  $3.0 \text{ g}_{\text{cat}}/\text{h}/\text{mol}$ , which were considered critical thresholds in the development of a small, compact stationary reformer. At an  $H_2$  production rate of  $1 \text{ Nm}^3/\text{h}$ , the catalyst amount was calculated to be 167.8 g and the reformer length required to charge the catalyst was 613 mm, with a diameter of 1 inch. The  $CH_4$  conversion and  $H_2$  production rates achieved with the compact reformer using the 20 wt% Ni/ $\gamma$ - $Al_2O_3$  catalyst at  $738 \text{ }^\circ\text{C}$  were 97.9% and  $1.22 \text{ Nm}^3/\text{h}$ , respectively. Furthermore, a heat-exchanger type reformer was developed to efficiently carry out the highly endothermic SMR reaction for on-site  $H_2$  production. This reformer comprised a tube side (in which the catalysts were charged and the SMR reaction took place by feeding the reactants) and a shell side (in which the heat for the endothermic reaction was supplied by  $CH_4$  combustion). Reforming activities were evaluated using the active 20 wt% Ni/ $\gamma$ - $Al_2O_3$  catalyst, depending on the reactants' gas hourly space velocity (GHSV). The  $H_2$  production rate increased as the GHSV increased. Finally, the reformer produced a  $CH_4$  conversion of 98.0% and an  $H_2$  production rate of  $1.97 \text{ Nm}^3/\text{h}$  at  $745 \text{ }^\circ\text{C}$ , as well as a high reactants' GHSV of  $10,000 \text{ h}^{-1}$ . Therefore, the heat-exchanger type reformer proved to be an effective system for conducting the highly endothermic SMR reaction with a high reactants' GHSV to yield a high rate of  $H_2$  production.

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## Introduction

In areas where natural gas was available in large quantities, interest centered on the steam reforming of methane as a source of hydrogen (H<sub>2</sub>) [1]. Hydrogen is produced by the steam methane reforming (SMR) process in large, centralized industrial plants for use in numerous applications, including chemical manufacturing and petroleum refining [2].

Today, H<sub>2</sub> production can be classified into three types: thermal processes, electrolytic processes, and photolytic processes [3]. Thermal processes use the energy contained in resources, such as natural gas, coal, or biomass, to release the H<sub>2</sub>; in “thermochemical” processes in particular, heat is used in combination with closed chemical cycles to produce H<sub>2</sub> from water. Electrolytic processes use electricity to split water into H<sub>2</sub> and oxygen (O<sub>2</sub>), and the two pathways are electrolysis using zero-carbon sources of electricity (wind or solar) and high-temperature nuclear electrolysis. Photolytic processes use light energy to split water into H<sub>2</sub> and O<sub>2</sub>; its two main categories are photobiological and photoelectrochemical. In addition, there are seven major technology pathways based on the above types of technologies, including distributed natural gas reforming, bio-derived liquids reforming, coal and biomass gasification, water electrolysis, thermochemical production, photoelectrochemical production, and biological production. Table 1 shows the feedstock, energy source, production scale, and types of H<sub>2</sub> production for these seven pathways.

Recently, Dincer and Acar reviewed various potential methods of H<sub>2</sub> production using renewable and non-renewable sources and evaluated these technologies for environmental impact, cost, energy efficiency, and exergy efficiency [4]. The overview of H<sub>2</sub> production methods according to primary energy and material source was comprehensively explained with brief introductions to each method. In particular, biochemical energy was definitely classified as the primary energy source for generating H<sub>2</sub> in dark fermentation. These authors showed that not only photonic energy-

based H<sub>2</sub> production, but also thermochemical water splitting and hybrid thermochemical cycles were the most environmentally benign in terms of carbon dioxide (CO<sub>2</sub>) emissions and acidification potential. Therefore, solar-based H<sub>2</sub> production was regarded as a potential option that deserved greater attention because it would increase both energy and exergy efficiencies and decrease the costs of H<sub>2</sub> production. Thermochemical H<sub>2</sub> production offers a potential route to clean, sustainable H<sub>2</sub> production because it uses only water, heat, and chemicals that are recycled. Furthermore, hybrid thermochemical cycles were found to be promising candidates for the production of H<sub>2</sub> in an environmentally benign and cost-effective way.

Herron et al. also reviewed various methods of producing H<sub>2</sub> specifically from water using solar energy; these methods included electrolysis, photo-electrochemical water splitting, photo-catalytic water splitting, and thermochemical water splitting [5]. According to their study, water electrolysis has reasonably high system efficiencies, and coupling with solar-photovoltaic technology can be a viable way to produce clean H<sub>2</sub> in the near future. Photo-electrochemical and thermochemical methods are capable of achieving solar-to-hydrogen efficiencies similar to those of the electrolysis systems. The photo-catalytic system is considered attractive because of its simplicity, but the low solar-to-hydrogen efficiencies remain critical challenges.

As described above, there are various potential methods of H<sub>2</sub> production using renewable and non-renewable sources. Given the current state of technology, however, natural gas-based H<sub>2</sub> production in large industrial plants is considered to be cheapest method available [4]. With the growing concern about global climate change, alternatives to the atmospheric release of CO<sub>2</sub> when H<sub>2</sub> is being produced from fossil fuels are being investigated. Sequestration of CO<sub>2</sub> could be an option as a viable near-term solution [6]. Among the three main fossil fuel reforming technologies (i.e., steam reforming, partial oxidation, and autothermal reforming), steam methane reforming is the least expensive and most common method for producing H<sub>2</sub> [4]. Natural gas steam reforming is currently

**Table 1 – Overview of seven major technology pathways for H<sub>2</sub> production (partly from Ref. [3]).**

Major technology pathways	Feedstock	Energy source	Production scale	Types of hydrogen production
Distributed natural gas reforming	Natural gas	Natural gas	Distributed	Thermal process
Bio-derived liquids reforming	Biomass	Biomass	Distributed	Thermal process
Coal and biomass gasification	Coal	Coal	Semi-central	Thermal process
	Biomass	Biomass	Central	
Water Electrolysis	Water	Grid	Distributed	Electrolytic process
		Wind	Semi-central	
		Solar	Central	
		Nuclear		
Thermochemical production	Water	Solar Nuclear	Semi-central Central	Thermal process
Photoelectro-chemical production	Water	Solar	Semi-central Central	Photolytic process
Biological <sup>a</sup> production	Water	Solar	Semi-central	Photolytic process
	Biomass	Biomass	Central	

<sup>a</sup> Biological production includes other biological approaches that are not strictly photolytic; e.g., dark fermentative, microbial-aided electrolysis.

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