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Detlef Stolten, Thomas Grube (Eds.):
18th World Hydrogen Energy Conference 2010 - WHEC 2010
Parallel Sessions Book 3: Hydrogen Production Technologies - Part 2
Proceedings of the WHEC, May 16.-21. 2010, Essen
Schriften des Forschungszentrums Jülich / Energy & Environment, Vol. 78-3
Institute of Energy Research - Fuel Cells (IEF-3)
Forschungszentrum Jülich GmbH, Zentralbibliothek, Verlag, 2010
ISBN: 978-3-89336-653-8

Demonstration of Highly-Efficient Distributed Hydrogen Production from Natural Gas with CO_2 Capture

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1 Introduction

To reduce carbon dioxide (CO_2) emission in wide range of fields and industries, various efforts have been accelerated including research and development on hydrogen and fuel cell technologies. Hydrogen is expected to play a significant role as an energy carrier to mitigate CO_2 emission in the coming decades. Though hydrogen has no environmental impacts at the point of use, most of hydrogen is produced today from fossil fuel sources and used mainly in industries [1,2]. Further utilization of hydrogen requires key technology breakthroughs and device cost reduction in all domains over the hydrogen energy chain including fuel supply infrastructure. On-site hydrogen production from natural gas using the existing pipeline networks is one of the most promising pathways to produce and supply hydrogen to fuel cells and vehicles. Since hydrogen production from natural gas is a practical and reasonable way for early introduction of hydrogen energy into society, further CO_2 reduction must be considered in the future because carbon dioxide is released during hydrogen production. Carbon capture technologies are important at upstream of CCS (Carbon Capture and Sequestration) chain and lowering the energy penalty and capture cost is a key issue for practical application. Hydrogen separation membrane technology can be a solution to the issue and provide an efficient carbon capture in hydrogen production from natural gas.

A membrane reformer for hydrogen production is more compact and more highly-efficient than the conventional SMR (Steam Methane Reforming) with PSA (Pressure Swing Adsorption) system because steam reforming reaction of natural gas and hydrogen separation process proceed in a single reactor simultaneously without a separate shift converter and a purification system. A schematic diagram and principle of the membrane reformer is shown in Figure 1. In the membrane reformer, natural gas is reformed in the catalyst bed and only hydrogen can be separated from a reformed gas mixture by a hydrogen separation membrane. The reforming process with simultaneous hydrogen separation can be free from the limitation of chemical equilibrium [3] and provide higher energy efficiency than the conventional SMR technologies. Due to these advantages, the membrane reformer is expected to be applied to on-site hydrogen production including hydrogen refuelling stations for fuel cell vehicles. The membrane reformer offers another significant advantage of high CO_2 concentration of 70~90% in the reactor off-gas. Even under the rated operating condition where the CO_2 concentration of the reactor off-gas is the lowest, the CO_2 concentration is significantly higher than that of fossil fuel combustion systems, i.e., natural gas fired boiler: 7~10%, gas turbines: 3~4%, and coal fired boilers: 12~14% [4]. Due to the high CO_2 concentration of the off-gas, CO_2 purification is not required and CO_2 can be captured easily by only compression and liquefaction and the cost for carbon capture can be reduced significantly compared with the conventional reforming systems. The membrane reformer is a very promising technology not only to achieve the highest efficiency in hydrogen production from natural gas but also to capture CO_2 efficiently. In the present study, we have demonstrated highly-efficient distributed hydrogen production with CO_2 capture using an advanced membrane reformer system.

2 Performance of 40 Nm^3/h-class Membrane Reformer System

Tokyo Gas has been working on an advanced membrane reformer system development for highly-efficient hydrogen production from natural gas for more than 15 years. We have successfully developed and operated a 40 Nm^3/h-class membrane reformer system (1st MRF) and demonstrated its high efficiency of 76.2% (HHV) and the product hydrogen was supplied to fuel cell vehicles in 2004 [5]. Hydrogen production efficiency from natural gas was defined by the following equation:

$$\text{Efficiency}(\%) = \frac{F_{H2} \times Q_{H2}}{F_{NG} \times Q_{NG} + W(AUX)}$$

where F_{H2} is the flow rate of product hydrogen, F_{NG} is the flow rate of natural gas, Q_{H2} is the gross heating value of hydrogen, Q_{NG} is the gross heating value of natural gas, and W(AUX) is auxiliary power consumption. Electricity for hydrogen suction/compression unit was not included in the efficiency defined above. The development of the 1st MRF system also revealed that its compactness reduced the total volume of the system to 1/3 of the conventional reforming system based on SMR-PSA.

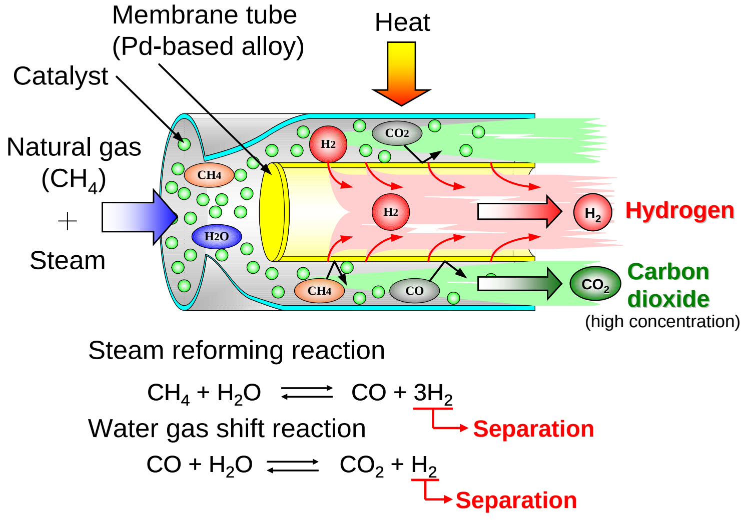

Figure 1: Principle of membrane.

We have achieved a significant milestone of over 80% efficiency with the improved version of the 40 Nm^3/h-class membrane reformer system (2nd MRF) in 2008. When the natural gas feed rate was 11.2 Nm^3/h, the hydrogen production rate was 40.5 Nm^3/h, and the improved system achieved a remarkable gain in hydrogen production efficiency up to 81.4% (HHV), which is about 5% higher than the previous system. The improved membrane reformer system has achieved the world-highest efficiency in hydrogen production from natural gas and has been proved to give the highest efficiency among various competing technologies. These demonstrations proved that the hydrogen production efficiency of the membrane reformer system is about 10~15% higher than the conventional steam reforming technologies with PSA. Fig. 2 shows the off-gas composition of the membrane reformer systems. The concentration of CO_2 in the off gas increased significantly due to enhanced reaction by simultaneous hydrogen separation. When the natural gas feed rate was 3.2 Nm^3/h, the CO_2 concentration in the 1st MRF off-gas was as high as 90%. In the case of the 2nd MRF, the CO_2 concentration was 72% at minimum at the designed hydrogen production capacity of 40 Nm^3/h (natural gas feed rate = 11.2 Nm^3/h).

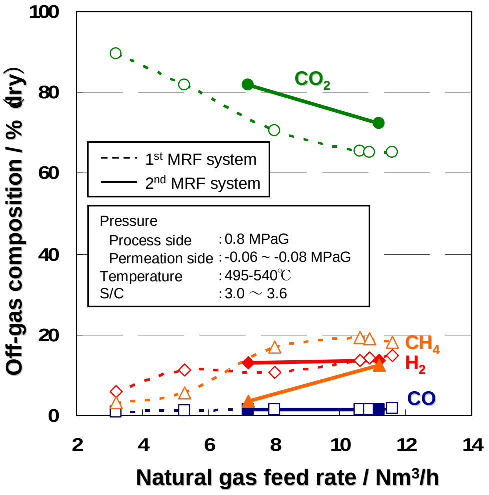

Figure 2: Natural gas feed rate dependence of off-gas composition.

3 Demonstration of Distributed Hydrogen Production with CO_2 Capture

The potential of the membrane reformer system to capture CO_2 was evaluated by calculation prior to the demonstration of distributed hydrogen production with CO_2 capture. CO_2 emission from each part and possible CO_2 reduction rate of the membrane reformer system were calculated by assuming that 100% CO_2 in the reactor off-gas can be captured by liquefaction process. The material balance of the 40 Nm^3/h-class membrane reformer system was calculated based on the actual operation data of the membrane reformer system. The CO_2 capture system is assumed to be composed of a water removal equipment, a compressor, a chiller and a gas-liquid separation unit. The off-gas from the membrane reformer system was assumed to be compressed up to 7.0 MPaG at over 29°C, then cooled down to room temperature to liquefy and separate CO_2 in the gas-liquid separation unit. CO_2 emission from the electric power consumption was calculated from the average emission factor of the Tokyo area in 2008 (0.332 kg/kWh) [6]. The required electricity for the CO_2 capture system was also estimated with the theoretical power consumption of the compressor. The calculated CO_2 emission and possible reduction rate at the designed capacity of the membrane reformer are shown in Figure 3. The amount of CO_2 emission from the reactor in the reformer system, which can be captured by liquefaction, was 56% at the designed capacity of 40 Nm^3/h. The estimation revealed that CO_2 emission in hydrogen production can be reduced significantly by capturing CO_2 from the off-gas.

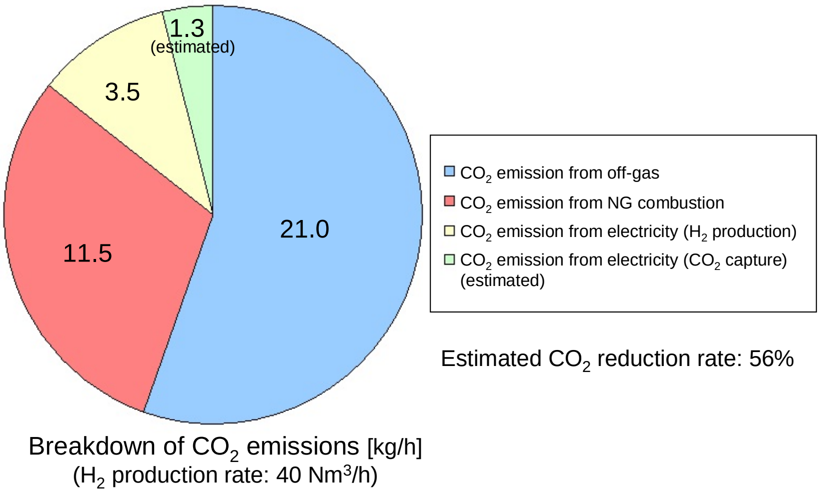

Figure 3: Estimated CO_2 emission and CO_2 reduction rate of the 40 Nm^3/h-class membrane reformer.

To confirm the CO_2 reduction potential of the membrane reformer system, a test equipment for CO_2 capture was developed for a demonstration test and attached to the 40 Nm^3/h-class membrane reformer system. Figure 4 shows the improved 40 Nm^3/h-class membrane reformer system and CO_2 liquefaction test equipment. Hydrogen production operation was carried out and part of the reactor off-gas was introduced into the test equipment to separate and capture CO_2. The obtained and calculated data from the demonstration at 75% of the designed capacity (hydrogen production rate = 30.6 Nm^3/h) are shown in Table 1. When the system was operated with CO_2 capture at 75% of the designed capacity, over 90% of CO_2 in the reactor off-gas was captured and CO_2 emission decreased from 25.1 to 12.6 kg/h, while electricity consumption was increased from 6.6 to 13.6 MJ/kg-H_2 by applying CO_2 capture. The total CO_2 emission of hydrogen production was decreased by 50% with only 3% energy loss. The total energy efficiency of hydrogen production was still as high as 78.6% (HHV) even with CO_2 capture, which is 5~10% higher than the conventional SMR + PSA technologies without CO_2 capture.

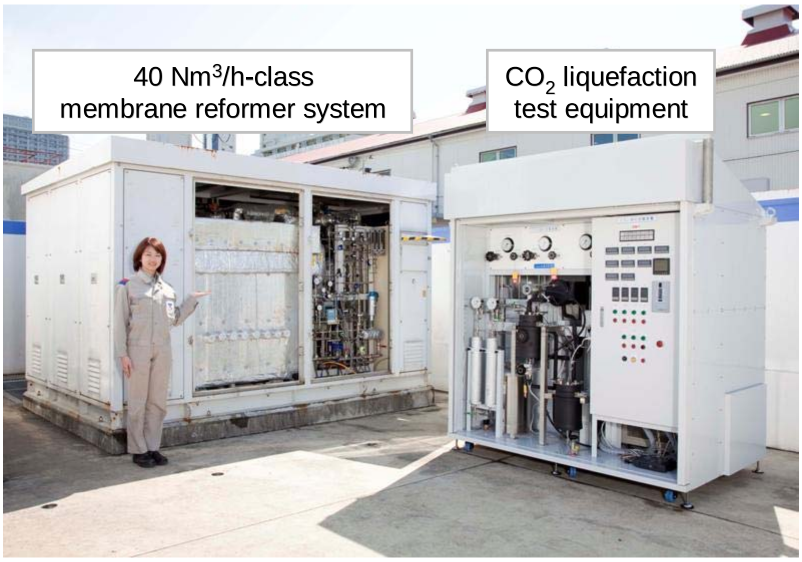

Figure 4: **40 Nm3/h-class membrane reformer system and CO_2 liquefaction test equipment.**

Table 1: **Energy efficiency of hydrogen production and CO_2 reduction rate.**

		MRF 75% load (individual operation)	MRF 75% load + CO_2 capture (calculated from experimental data)
Input Energy ($MJ/kg-H_2$)	Natural gas	168.5	168.5
	Electricity	6.6	13.6
Hydrogen production rate (Nm^3/h)		30.6	30.6
Efficiency (%, HHV)		81.7	78.6
CO_2 emission (kg/h) [*1]		25.1	12.6
CO_2 reduction rate (%)		-	50

[*1] CO_2 emission from the electric power consumption was calculated from the average emission factor of the Tokyo area in 2008 (0.332 kg/kWh) [6]

4 Summary and Future Prospects

In the present study, distributed highly-efficient hydrogen production with CO_2 capture has been demonstrated with the 40 Nm^3/h-class membrane reformer system. The experiment was the first demonstration in the world in terms of small-scale on-site hydrogen production with CO_2 capture. The advanced membrane reformer system can reduce CO_2 emission to half with only 3% energy loss by applying the CO_2 capture system. CO_2 removal in on-site distributed hydrogen production used to be considered unrealistic with conventional technologies because of large energy loss, costly and space-consuming CO_2 purification and capture facilities. This study revealed that the advanced highly-efficient membrane reformer system has a potential to make it feasible to capture CO_2 efficiently even in on-site distributed hydrogen production and proved the possibility of feasible small scale CO_2 capture.

During the introduction period of hydrogen energy, local hydrogen network, which supplies hydrogen not only to fuel cell vehicles but also residential and stationary fuel cells, is expected to be a regional hydrogen supply model to facilitate the spread of hydrogen energy into the society. On-site hydrogen production is a key technology in the local hydrogen network and the advanced membrane reformer system with CO_2 capture is a promising technology to produce “low-carbon” hydrogen efficiently.

Acknowledgement

The development of the 40 Nm_3/h-class membrane reformer system was supported by the New Energy and Industrial Technology Development Organization (NEDO). The authors acknowledge their financial and technical supports.

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