

A STUDY ON HYDROGEN CARRIER SYSTEM FOR FUEL CELL VEHICLE

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Abstract

A hydrogen carrier system that was utilized chemical reactivity was discussed for fuel cell (FC) vehicle in this study. One of the key that will make the widespread use of FCs possible is a hydrogen supply system. Then, possibility of new hydrogen system for FC vehicles, which utilized chemical reactions and capable to realize efficient hydrogen transportation with zero carbon dioxide emission and small explosion risk, was discussed in this study. The reactivity of calcium oxide to carbon dioxide was used for the carbon dioxide fixation and also for heat source of fuel reforming of methane. The reaction process realized high-purity hydrogen production under mild operation conditions, and was regenerative thermally by using high-temperature thermal energy produced from renewable energy or nuclear power. To estimate the efficiency of the fuel reforming system using the reaction process, the reactivity of hydrogen production was examined experimentally. The proposed reforming system was expected to be applicable as a hydrogen carrier system in zero carbon dioxide emission. This system would show one of possibilities of chemical energy conversion technologies.

KEYWORDS

Hydrogen, Hydrogen Carrier, Carbon Dioxide, Zero Emission, Fuel Reforming

INTRODUCTION

FC offers the possibility of expanding the electricity utilization market. Vehicles are seen as particularly good candidates for FC application, because FC is more compact, quieter and emit cleaner exhaust gas than conventional internal combustion engines. One of the key technologies that will make the widespread use of FC possible is a hydrogen (H_2) supply system. The uses of liquefied or compressed H_2 are candidates for this technology. However, the storage and transportation of either of these forms of H_2 require large amounts of energy as well as stringent safety precautions. These drawbacks make steam reforming of common fuels, such as methane, propane, methanol and kerosene, more practical solution for storing and supplying H_2 . Steam reforming can occur at the site of the FC.

The use of any of these chemical reactants as a H_2 storage medium presents the possibility of a safe H_2 carrier and supply system. On the other hand, the reforming requires additional apparatuses for H_2 production, including at least three: a steam reforming reactor, a burner for reforming heat supply and a carbon monoxide converter. In vehicles, it is especially important for a FC reformer to be compact and lightweight. A concept of a thermally regenerative steam fuel reformer for a vehicle had been proposed [1]. This paper showed that a H_2 carrier system for FC vehicles using the reformer could achieve zero carbon dioxide by using chemical fixation of carbon dioxide and utilize of thermal output form a high-temperature heat source, and estimated experimentally a possibility of the H_2 carrier system.

Separation process is key technique for high-efficient fuel reforming. **Figure 1** shows concept of the separation processes to enhance H_2 yield. Conventional steam reforming in Fig. 1 (a) for hydrocarbons attains to equilibrium and produces H_2 , carbon dioxide (CO_2) and carbon monoxide (CO). The yield of H_2 is restricted by equilibrium. Removing H_2 from reforming reaction system induces the system into non-equilibrium state, and H_2 production is going on to establish the next equilibrium state in Fig. 1(b). The yield of H_2 will be enhanced in the result. Removing CO_2 in Fig. 1(c) induces also non-equilibrium state and enhances CO_2 production, then H_2 productivity and purity are also enhanced. These separation processes would realize not only high-yield of H_2 , but also decrease of temperature of the endothermic reforming. This study aims the demonstration of the CO_2 separation process for high-yield production of high-purity H_2 under lower temperature.

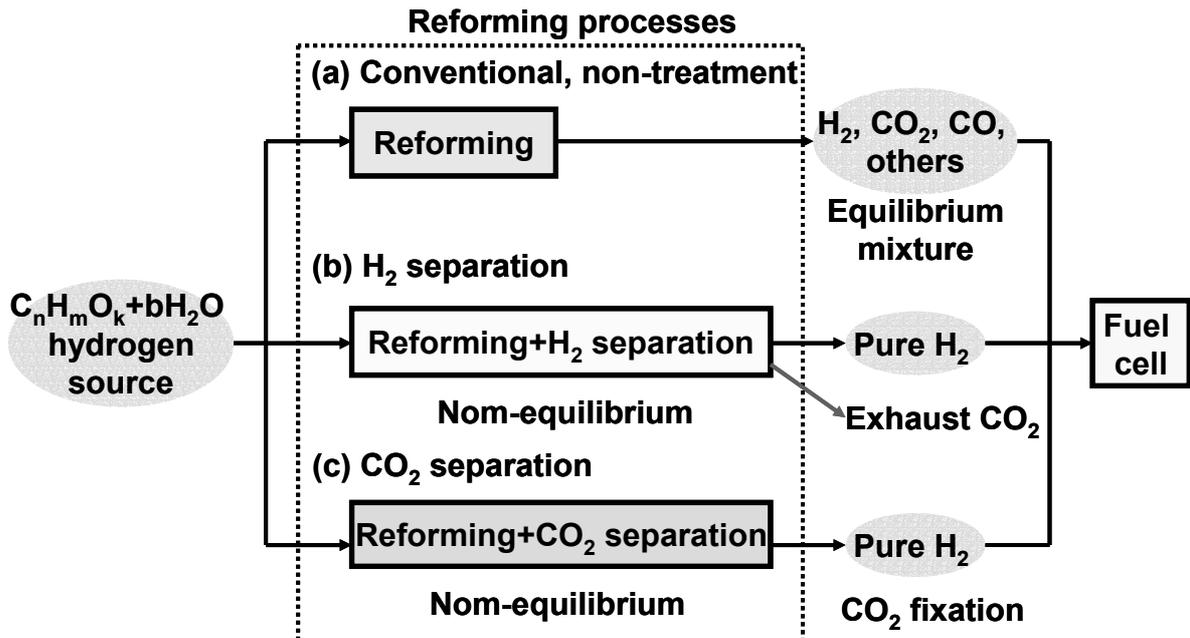


Fig. 1. Separation processes for enhancement of H₂ yield in fuel reforming

CONCEPT OF THERMALLY REGENERATIVE REFORMER FOR A VEHICLE

Regenerative Reformer

In this study, methane (CH₄) was chosen at first as a candidate reactant for steam reforming, because it is the most popular natural fuel resource and has a simple hydrocarbon fuel structure. The following regenerative reformer methodology is applicable also to kerosene and propane, both of which have reforming temperatures in the range of 700-900°C, similar to that of methane. The CH₄ steam reforming process consists of the following two gas phase reactions with various catalysts.

Methane steam reforming:



Carbon monoxide (CO) shift reaction:



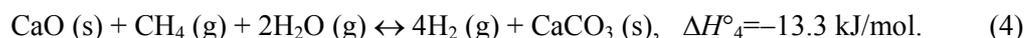
The study attempts to use calcium oxide (CaO) carbonation to remove carbon dioxide (CO₂) from the reformed gas and fix it.

Carbonation of calcium oxide:



This study aims to cause Eqs. (1), (2) and (3) reactions in the same reactor at once. These reactions, taken as a whole, are defined as regenerative reforming.

Regenerative reforming:



Conventional steam reforming is depicted in **Figure 2 (a)**. CH₄ and water (H₂O) react by Eq. (1) in a catalytic reformer, and the generated CO is shifted by Eq. (2) into CO₂ and H₂ in a catalytic converter. The endothermic reforming process needs a heat supply of ΔH^o₁. The proposed process is shown in **Figure 2 (b)**. This process consists of a reforming process (Fig. 2 (b-1)) while the vehicle is driving and a regenerating process (Fig. 2 (b-2)) for calcium oxide regeneration and CO₂ recovery while the

vehicle is turned off. CaO and a reforming catalyst mixture are packed in a regenerative reformer. Reactants are reformed by Eq. (1), and generated CO₂ is removed from the gas phase by the CaO carbonation of Eq. (3). The CO shift reaction of Eq. (2) is enhanced under the non-equilibrium condition realized by the CO₂ removal. Purified H₂ is generated from the reactor finally. The whole reaction of Eq. (4) is exothermic, hence the reaction needs no heat supply and can proceed spontaneously. A zero CO₂ emission drive is possible due to CO₂ fixation resulting from the carbonation. In the regenerating process, CaCO₃ is decomposed endothermically into CaO in the reactor using high-temperature heat, which is assumed to be supplied as heat from high temperature gas reactor, or as joule heat generated from off-peak electric output of other type nuclear power plants. The reformer is regenerated, and used again for the reforming. The proposed regenerative reformer is intended to be contained in a removable package for use in a FC vehicle. The package is loaded into and recovered from a vehicle at a regeneration station that supplies new packages and regenerates used ones. Regenerated CO₂ is managed according to a CO₂ recovery process.

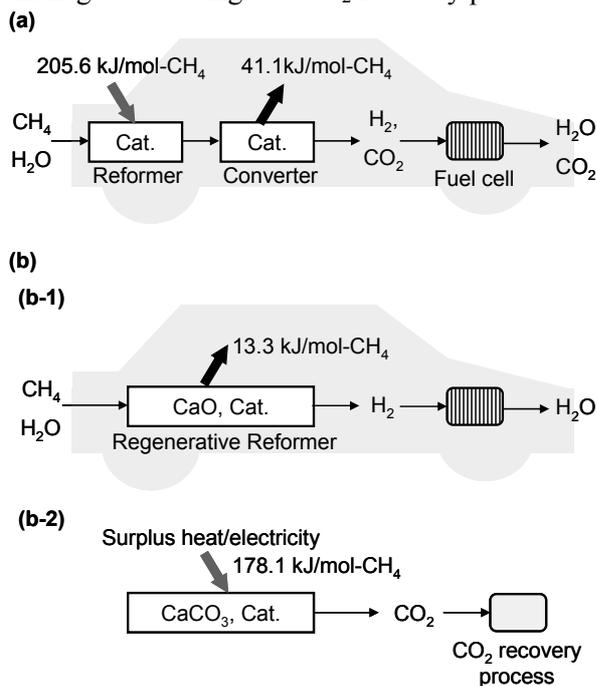


Fig. 2. Concept of a zero CO₂ emission FC vehicle using a thermally regenerative reformer; (a) conventional reforming, (b) proposed thermally regenerative reforming, (b-1) reforming mode, (b-2) regenerating and CO₂ recovering mode

Zero CO₂ Emission H₂ Carrier System

The concept of a zero CO₂ emission H₂ carrier system using the regenerative reforming process depicted in Fig. 2(b) is proposed in **Figure 3**. The zero CO₂ emission system consists of FC vehicles using packages of the regenerative reformer, a decentralized package regeneration station, and nuclear power systems for thermal energy supply. The regeneration station plays central role in the system. The packages are loaded in FC vehicles. The vehicles are driven by H₂ fuel produced from the packages. The packages after reforming are collected to the regeneration station. The packages are regenerated, that is, decarbonated thermally using thermal output or joule heat produced from renewable energy or nuclear power plant, and heat output from high-temperature industrial process. Regenerated packages are reused repetitively in the vehicles. Generated CO₂ is recovered in a storage vessel, and is regenerated in hydrocarbons at a hydrocarbon regenerator by hydrogenation process using H₂, which is generated from water electrolysis consuming the power plant output. Regenerated hydrocarbons are reused cyclically in the vehicles. A comprehensive zero CO₂ emission system is formed using the hydrocarbon regeneration process. The system was expected to contribute on load leveling of renewable energy or nuclear power plant operations by utilizing off-peak electricity or thermal output of the plants as heat source for the CaO and hydrocarbons thermal regeneration

processes. Because the system is acceptable unstable thermal energy input also, the system would realize stable load operation for renewable energy systems. Utilize of off-peak electricity in the system would be advantageous for economical power plant operation.

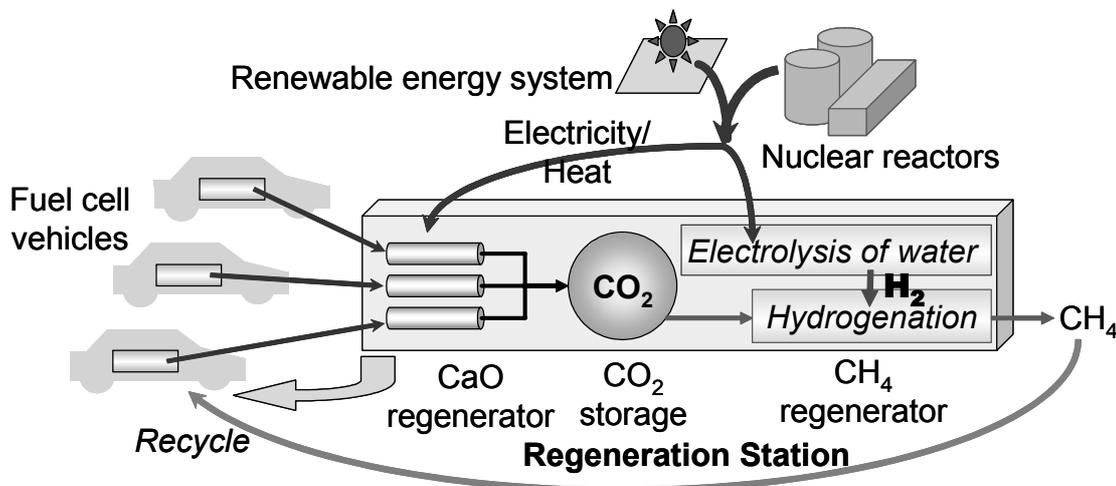


Fig. 3. Zero CO₂ emission H₂ carrier system using the thermally regenerative reformer driven by off-peak electricity and heat from renewable energy and nuclear power

The proposed H₂ carrier system for FC vehicles has several merits. Firstly, because the FC system using the proposed process emits no CO₂ during operation, a zero CO₂ emission vehicle system could be established, so long as the treatment of CO₂ is managed well after it is removed from the package, and the system can transport H₂ safely under low-pressure and as low-explosive chemicals. The reforming process is simpler than conventional reforming systems. Because the regenerative reforming is exothermic, the reforming proceeds automatically by self-heating, then, heat conduction control step arisen by heating for conventional reforming can be removed. The power-generation efficiency of a FC after with the reformer will be enhanced by the supply of the reformed high-purified H₂. In a conventional system, H₂ effluent gas from the FC is burned for the next reforming, because the CO₂ concentration in the gas is too high for the cell to use. In the proposed system, on the other hand, the H₂ concentration is higher enough than one of a conventional system, enabling the H₂ to be consumed highly in the cell. Therefore, the proposed system also enhances the H₂ economy of the FC. Because the reformation equilibrium temperature is shifted to a lower temperature than in a conventional system, the exothermic CO shift reaction is enhanced naturally. Furthermore, the CO reduction induced by the enhancement is advantageous for FC durability.

The simultaneous reaction concept for H₂ production from methane steam reforming had been patented by Williams [2]. A fluidized bed concept using a reforming catalyst and carbon dioxide acceptor for H₂ production was patented by Gorin and Retallik [3]. Shift reaction and carbon dioxide removal in a single-reactor packed with a calcium oxide mixture were examined by Chun Han [4]. Calcium oxide as a CO₂ absorber was also applied to regenerative H₂ production by Balasubramanian [5]. Those proposals were based on the use of the regenerative reforming process in fixed plants for H₂ production, such as a fluidized bed or a combined system of a packed-bed reactor and gas turbine. Continuous batch-wise H₂ production system using two regenerative reformers is proposed for a vehicle use by Specht [6]. This study proposed a new concept of zero CO₂ emission H₂ carrier system utilizing the reaction concept.

EXPERIMENT OF THE REGENERATIVE REFORMER

Experimental Discussion

A flow-type experimental apparatus for the steam reforming process in **Figure 4** was used for the reforming study (Kato *et al.*, 2003). A quartz tube, 15 mm in inner diameter was used as a reactor. CaO particles and a reforming catalyst are packed in various forms in the reactor. Reaction gases controlled by mass-flow meters and water supplied by a micro-feeder were mixed previously in an

evaporator and supplied into the reactor. The temperature at the upstream end of the packed reactants, T [°C], was referred to as a representative bed temperature and was controlled by the furnace. The reacted effluent gas from the reactor was separated into liquid and gas. The separated gas flow rate was measured by a soap film flow meter, and the gas was analyzed by a TCD-type gas chromatograph (Shimadzu GC-8A). Natural calcium carbonate (average diameter of 1.5 mm, purity over 98%, produced in Hiroshima, Japan, supplied by Nitto Funka Kogyo Co. Ltd.) was used in the reactor as the precursor to CaO. Nickel catalyst particles, screened to between 0.3 and 0.5 mm, were used for the reforming. A mixture of 2.7 g Ni catalyst, 21.9 g alumina balls as a diluent and 8.1 g CaCO₃ was packed into the reactor after the alumina ball pre-heating layer.

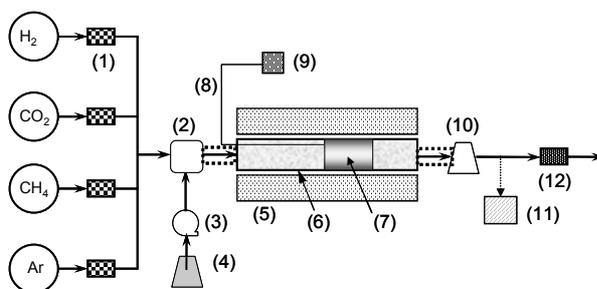


Fig. 4. Experimental apparatus for the regenerative reforming

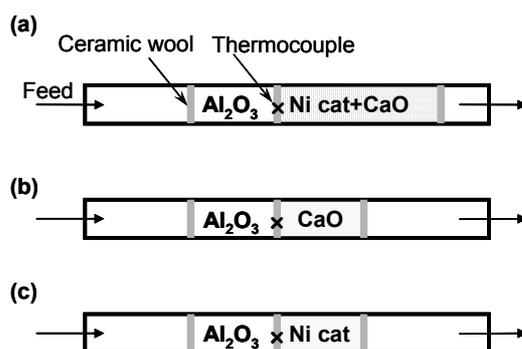


Fig. 5. Packed bed structures of the reformer in the experiment: (a) regenerative fuel reformer (RGR), (b) carbon dioxide absorber (CDA), (c) conventional reformer (CVR)

The decarbonation of CaCO₃ was proceeded prior to the reforming experiment, at 830 °C under argon flow. The reduction of the catalyst was proceeded under H₂ flow after the decarbonation process. For reforming processes, methane, water and argon were supplied to the reactor under atmospheric pressure.

Three kinds of packed bed structures, shown in **Figure 5**, were discussed: (a) a regenerative reformer type, (b) a CO₂ absorber type, and (a) a conventional reformer type.

- (a) Regenerative reformer (RGR): The regenerative reforming process was examined in this reactor form. Alumina balls, 2.0 mm in diameter, were packed for pre-heating of the reaction gas in the layer. A mixture of 2.7 g Ni catalyst, 8.1 g CaCO₃ as the precursor of CaO and 21.9 g alumina balls as a diluent was packed into the reactor after the alumina ball layer. Ceramic wool pieces, 10 mm thick, were interposed between the ball layer and the mixture layer, and were placed also at both ends of the packed bed.
- (b) CO₂ absorber (CDA): The CO₂ removal performance of CaO carbonation was examined. At the downstream of the alumina ball pre-heating layer, CaCO₃ as the precursor was charged with the ceramic wool pieces.
- (c) Conventional reformer (CVR): The reactor was examined as a conventional reformer to allow for comparison with the regenerative reformer. 2.7 g of nickel catalyst, diluted with 21.9 g of alumina balls, was packed after the alumina ball layer.

For the RGR and CDA type reformers, the decarbonation of CaCO_3 was proceeded prior to the reforming, at 830°C under argon flow of 100 ml/min for 4 hours. The reduction of the catalyst was proceeded under H_2 flow of 100 ml/min for 60 min after the decarbonation process. For all reforming processes in the RGR and CVR, methane of $8.9 \times 10^{-4}\text{ mol/min}$ (20 ml (STP)/min), water of $3.6 \times 10^{-3}\text{ mol/min}$ (0.064 g/min) and argon of $2.2 \times 10^{-3}\text{ mol/min}$ (50 ml (STP)/min) were supplied to the reactor at bed temperatures of $550\text{--}750^\circ\text{C}$ under atmospheric pressure.

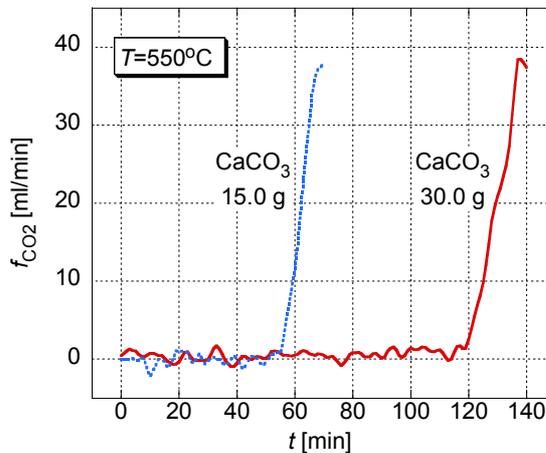


Fig. 6. Carbon dioxide chemical absorption ability of the CaO bed by CDA

Figure 6 shows the CO_2 absorption ability of the beds. The abscissa and ordinate show the carbonation time and the CO_2 effluent flow rate from the exit of the bed reactor. CO production by the reverse reaction of Eq. (2) was not detected in these operations. During initial carbonation, small amount of CO_2 was detected. CO_2 appeared clearly after 57 min and 120 min for 15.0 and 30.0 g of CaCO_3 , respectively. The appearance times were almost proportional to charged amount of reactant. The initial supplied CO_2 was considered to be absorbed well in the reactant until the reactant saturation period because the completion times with the CDA were close to the calculated ones. It was expected that the packed bed had enough CO_2 absorption ability and the chemical absorption were proceeded under a zone reaction.

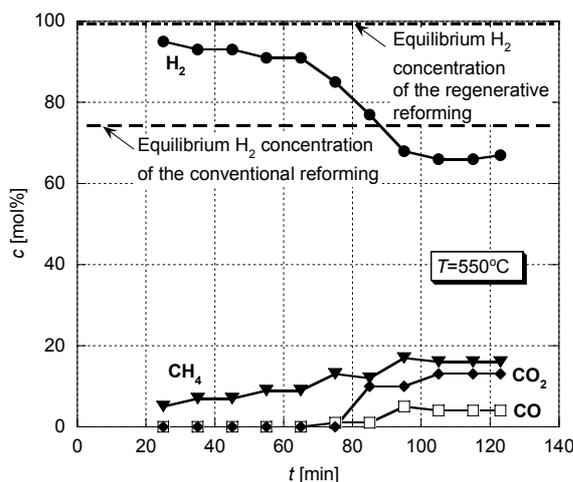


Fig. 7. Temporal change of effluent composition of the regenerative reformer at 550°C by RGR

Figure 7 shows an example of a temporal change in the concentrations in effluent of the regenerative reforming at 550°C . Time 0 min. indicates the start of reactants supply to the reactor. The dashed lines show H_2 equilibrium concentrations in regenerative reforming and conventional one, respectively. During the initial 60 min., charged CaO absorbed well CO_2 by carbonation, then, the

concentration of CO_2 and CO were less than 1% each, and the H_2 production was higher than the equilibrium concentration of conventional reforming. Because CO_2 adsorption kept the gas phase non-equilibrium, the shift reaction, steam reforming reaction and H_2 production all proceeded beyond the original standard equilibrium composition. As the carbonation was saturated after 70 min., the steam reforming and shift reactions changed into a conventional reforming condition, and the H_2 production decreased below the conventional equilibrium.

Comparison of reforming performance

The reforming performances of the RGR and CVR were compared from analysis of the gas products of these reformers.

(1) Change in H_2 concentration

Figure 8 compares the H_2 concentration in the effluent gas from each reactor. The bold and dashed lines show, respectively, measured and calculated equilibrium H_2 concentrations in the effluent gas, excluding inert gas. The concentration of H_2 with the RGR was higher than one with the CVR. The H_2 production performance of the RGR was outstanding at low temperatures. Especially, 94% of H_2 was measured in the RGR at 550°C . CaO carbonation removed CO_2 from the gas phase, and enhanced CO shift reaction in Eq. (2) and also CH_4 reforming in Eq. (1). Simultaneously, the exothermic heat of carbonation and the heat of the shift reaction compensated well for the endothermic reforming. The heat being exchanged between the reactions enhanced both reactions each other. At 650°C , the exothermic CO shift reaction and CaO carbonation progresses were smaller than they were at 550°C , and CH_4 reforming was inhibited due to the surplus CO . Thus, the H_2 production with the RGR was close to that with the CVR. The unique performance of the RGR was available at a lower temperature. The equilibrium line of the RGR, which shows the reactions in Eq. (4), is bent at 725°C because above this temperature the equilibrium CO_2 pressure of Eq. (3) is higher than the partial pressure of CO_2 in the reforming equilibrium composition, and the CaO effect on the reforming disappears.

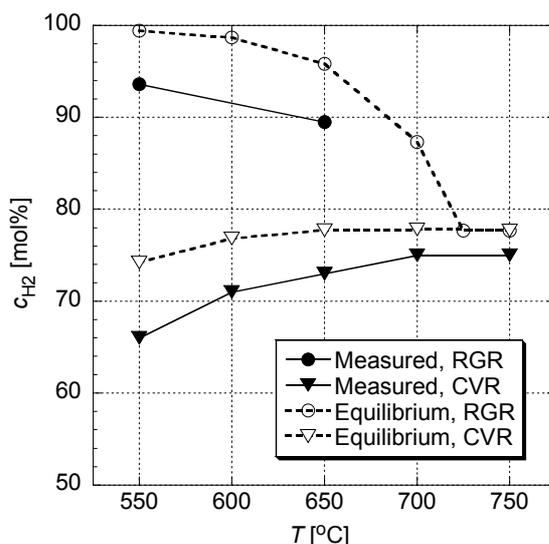


Fig. 8. Effect of bed temperature on H_2 production concentration of the reformers

(2) Change in carbon dioxide concentration

Figure 9 shows the CO_2 concentration change in the effluent gas. The CO_2 concentration of the RGR at 550°C was less than 1%. In the RGR, the exothermic heat of CaO carbonation was consumed well by the endothermic CH_4 reforming, enhancing the carbonation. The CO_2 concentrations with the RGR at 550 and 650°C are lower than that with the CVR because of the CO_2 absorption resulted from the CaO carbonation. The CO_2 concentration with the CVR decreased at a higher temperature, because of an increase in CO concentration. The CO_2 concentration with the RGR increased at a higher temperature on the contrary, because of a decrease in CO_2 absorption capacity of CaO with a rise in equilibrium CO_2 decomposition pressure. Then the CO_2 concentration difference between both reactors became smaller at a higher temperature.

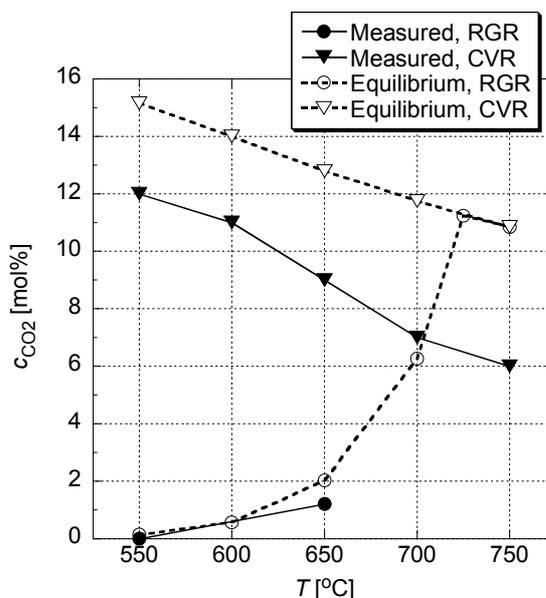


Fig. 9 Effect of bed temperature on effluent carbon dioxide concentration of the reformers

Performance Evaluation

Performance evaluation for the regenerative reformer based on the experimental results of this study is shown in **Table 1**. The reactor volume required for a vehicle FC, which needs 1.0 kg of H_2 in order to drive for 100 km, was evaluated from the experimental analysis. The estimation assumed that all of the produced H_2 was consumed in the FC. From this estimation, 7.94 kg (9.62 liters) of CaO was required for the reformer. The CaO requirement calls for similar amounts of total reforming reactants both CH_4 and H_2O . The capacity of a conventional H_2 bomb under 70.0 MPa requires 25.6 liters to store the same amount of H_2 . Thus the regenerative reforming system is expected to provide a more compact and lightweight container than the conventional H_2 storage system. A battery-powered vehicle is a practical zero emission car. Advanced lead-acid and Li-ion batteries are required of amount of 400 kg and 100 kg for the same amount electricity power storage, respectively. Compared with a battery-powered car, the reforming system could be lighter and more compact, and also regenerative by surplus thermal energy.

Estimated contribution of a nuclear power plant on the FC system is shown in **Table 2**. The estimation assumed that off-peak electricity from a nuclear power plant of 1 GWe electric output was used for the regeneration of the reformer for 8 h. 1.3×10^6 pieces of the reformer package are regenerative by the power plant. Over 7,100 tons of CO_2 is recoverable from the process. The FC system with a nuclear power plant would have effective contribution on reduction of CO_2 emission and protection against the global warming. The enhancement of durability of CaO reactant to the repetitive reversible reaction would be important subject also for practical use. Conventional CaO reactant loses reactivity easily under repetitive reaction cycle. The reactivity disappearance would be arisen from sintering between reactant primary particles by relatively high-temperature reaction. An economical enhancement of reaction durability to the sintering is required.

The regenerative reforming is applicable for higher-hydrocarbon fuels, such as propane and kerosene. Because CaO is capable of reacting with sulfur components, the reforming can contribute to the purification of H_2 product by removing sulfur components contained in an original fuel. The reforming system offers the possibility of realizing a zero CO_2 emission H_2 carrier system for FC vehicle and enhancing the utilization of surplus energy.

Table 1. Estimated size of the regenerative reformer for FC vehicle

Vehicle mileage [km]	100
H ₂ requirement [kg]	1.0
CaO requirement mass [kg]	7.94
CaO requirement volume [liter]	9.62
Recovered CO ₂ [kg]	5.5

Table 2. Estimated contribution of a nuclear power plant on the FC system

Power plant	
Power plant electric output [GW]	1.0
Night time operation duration [h]	8.0
Regeneration station	
Requirement reaction heat [MJ/piece]	22.3
Number of cell piece [pieces]	1.29×10 ⁶
Recovered CO ₂ amount [ton]	7.12×10 ³
[m ³ (STP)]	3.62×10 ⁶

EVALUATION OF THE H₂ CARRIER SYSTEM

Comparison between the H₂ carrier system depicted in Fig. 3 and other conventional H₂ systems is discussed to evaluate the carrier system.

Figure 10 shows enthalpy balance for conventional H₂ production using water electrolysis, the proposed carrier system and on-board fuel reforming. Those values in the figure show enthalpy changes per 1 mole of H₂ production. Conventional water electrolysis consumes electricity for H₂ production of 282 kJ/H₂-mol, and H₂ compression 28 kJ/H₂-mol. The compression is assumed isentropic and 5 stages compression up to 700 bar. Total of 311 kJ/H₂-mol is required.

The H₂ carrier system needs thermal input of 44.6 kJ/H₂-mol for CaO regeneration, electricity input of 282 kJ/H₂-mol for water electrolysis. 327 kJ/H₂-mol is needed totally. The H₂ carrier system needs enthalpy input slightly larger than the electrolysis. On the other hand, a methanation process of generated CO₂ and H₂ is exothermic, and produces thermal output of 41.1 kJ/H₂-mol at around 300 °C. When thermal output from the methanation is utilized in some heating process, enthalpy input for decarbonation would be cancelled. Enthalpy input for water electrolysis process is dominant in total input at both the conventional and the carrier systems. Although compression process needs electricity, decarbonation does thermal input. Then, exergy consumption of the carrier system would be smaller than one of the conventional system. On-board reforming needs smaller enthalpy input than one of other systems which have water electrolysis process. However, CO₂ is emitted from the former system. The merits and demerits of the carrier system would be summarized as the following from the enthalpy balance discussion.

Merits of the carrier system:

- Comparing with conventional water electrolysis H₂ system, the carrier system needs similar amount of enthalpy consumption. The latter would be more compact and lighter than the former, and can transport H₂ under lower pressure with small risk of H₂ explosion.
- Comparing with on-board fuel reforming system, the carrier system does not emit CO₂, and is carbon recyclable.
- Comparing with other H₂ storage materials, that is, metal hydroxides, hydrocarbons, the carrier system is operated self-heating with no additional heat input. Reactants in the latter are more environmental friendly, safer and cost lower.
- Comparing with compressed H₂ cylinder system, the carrier system is relatively more compact and lighter, and has small risk of explosion.

Demerits of the carrier system:

- The cylinder requires regeneration process at over 800 °C. Effective high-temperature heat source is required.

- Process for CO₂ conversion into methane or other hydrocarbons is required. The process would be more complicated than water electrolysis, and needs high cost.
- Reinforcement of CaO material to the repetition is required for practical use. Conventional CaO material has small durability to repetitive cyclic operation. High durable material is required to be developed.

The proposed H₂ carrier system has unique performance comparing with conventional H₂ production systems. Improvement of the demerits would enhance possibility for practical use of the system. Not only efficient H₂ production, but also H₂ supply system is needed for development of practical FC vehicle market. Comprehensive feasibility study of H₂ carrier system would be still important.

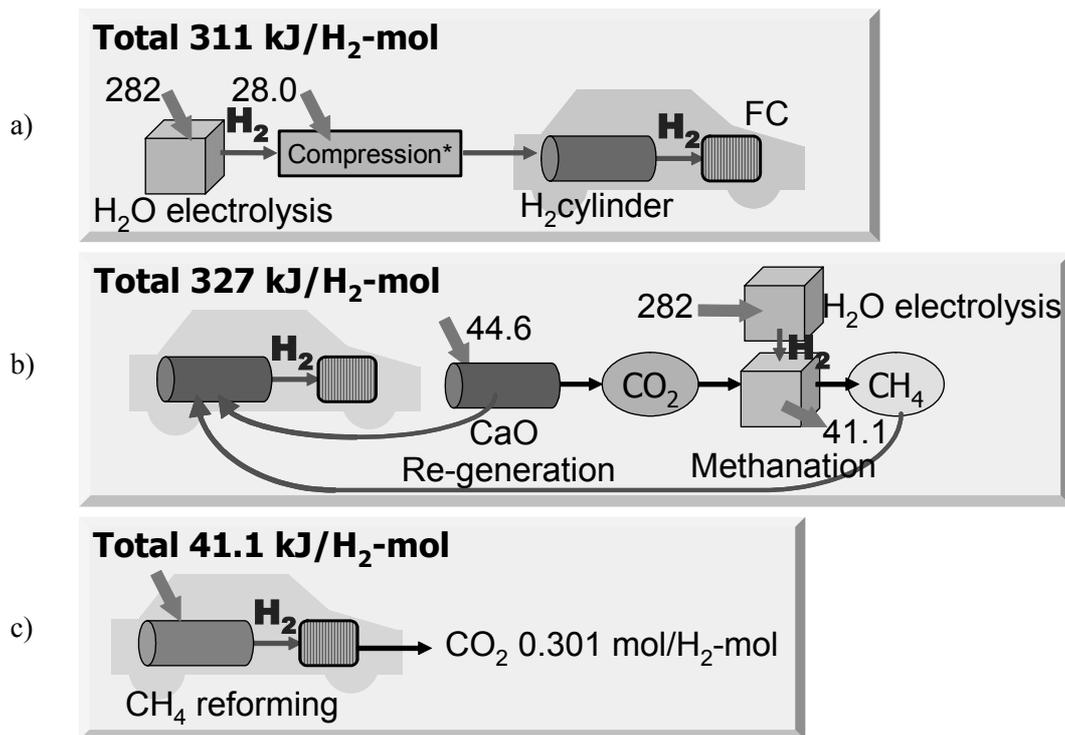


Fig.10. Enthalpy balance of H₂ systems, (a) conventional H₂ system using water electrolysis, (b) the zero CO₂ emission H₂ carrier system, (c) on-board fuel reforming. Values show enthalpy changes per 1 mole of H₂ production.

CONCLUSION

A zero CO₂ emission H₂ carrier system for FC vehicles using a regenerative fuel reformer based on renewable energy and nuclear power was proposed. The regenerative reformer was applicable to a practical zero CO₂ emission reforming with safety H₂ transportation. The required amount of CaO for the reformer was expected to be small enough comparing with other H₂ systems. The system can utilize off-peak electricity generated from renewable energy or nuclear power plant. Electricity of 1 GWe for 8 h can regenerate 1.3 million of reforming packages, and recover 7.1 ton of CO₂. Enthalpy consumption of the H₂ carrier system is similar or less than water electrolysis H₂ system. The H₂ carrier system also would contribute simultaneously on load leveling of power plant operation. The carrier system shows a new possibility of chemical energy conversion. Chemical energy conversion technologies would be expected to be developed in not only thermal energy utilization, but also new area.

NOMENCLATURE

- c* – mole concentration in effluent gas excluding inert gas [mol%]
- CDA – carbon dioxide absorber
- CVR – conventional fuel reformer

f_{CO_2} – CO₂ effluent flow rate from the exit of the bed reactor [ml/min]
RGR – regenerative fuel reformer
 T – bed temperature measured at the upstream end of reactor bed [°C]
 t – reaction time [min]
 ΔH° – standard enthalpy change [kJ/mol]

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