

POSSIBILITY OF A CHEMICAL HYDROGEN CARRIER SYSTEM BASED ON NUCLEAR POWER

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Abstract

Possibility of a chemical hydrogen carrier system for fuel cell vehicles, which utilised chemical reactions and capable to realise efficient hydrogen transportation with zero carbon dioxide emission and small explosion risk, was discussed in this study. The reactivity of metal oxide to carbon dioxide is used for the carbon dioxide fixation and also for heat source of fuel reforming. The performance of the combination of the chemical hydrogen carrier system and a high temperature gas reactor was estimated using experimental results. It was expected that the system realised low-pressure and safe storage of hydrogen carrier media consisting with the same energy efficiency of conventional water electrolysis system.

Introduction

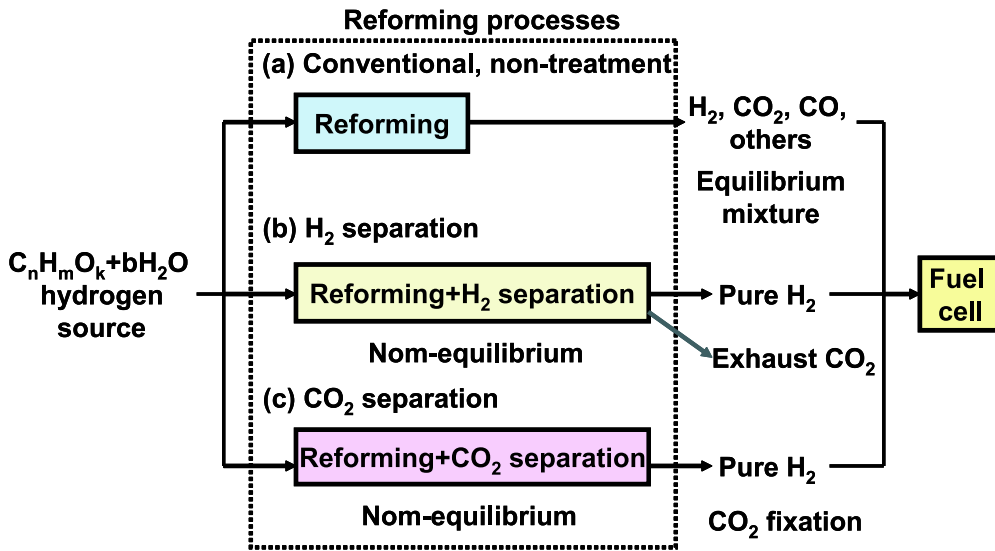
Fuel cell (FC) offers the possibility of expanding the electricity utilization market. Vehicles are seen as particularly good candidates for FC application, because FC is more compact, quieter and emit cleaner exhaust gas than conventional internal combustion engines. One of the key technologies that 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 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. It is especially important for a reformer of FC vehicle to be compact and lightweight. A concept of a thermally regenerative steam fuel reformer for a vehicle had been proposed [1]. The reformer can realize compact reforming and achieve zero carbon dioxide (CO_2) emission by using chemical fixation of CO_2 . Finally, zero CO_2 emission H_2 carrier system using the reformer can be established. The system needs energy input for fuel reforming and fuel regeneration processes. Nuclear power plant is good candidate as energy source because it emits zero CO_2 in operation. This paper discussed the possibility of the chemical H_2 carrier system based on the above concept. The energy balance of the carrier system combined with a high temperature gas reactor (HTGR) was estimated using experimental results. The possibility of the carrier system was compared with other hydrogen system.

Concept of Chemical H_2 Carrier System

Separation Process for Fuel Reforming

Separation process would be key technique for high-efficient fuel reforming. Figure 1 shows concept of the separation processes to enhance H_2 yield. Conventional steam reforming in Figure 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 by Le Chaterier's principle, and H_2 production is going on to establish the next equilibrium state in Figure 1(b). The yield of H_2 will be enhanced in the result. Removing CO_2 in Figure 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.

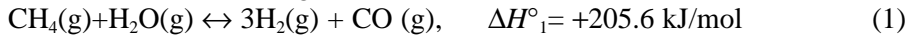
Figure 1. Separation processes for enhancement of H₂ yield in fuel reforming.



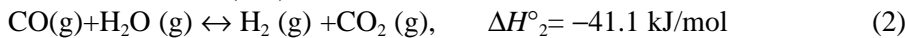
Regenerative Reformer

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:

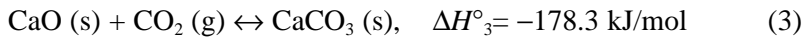


Carbon monoxide (CO) shift reaction:



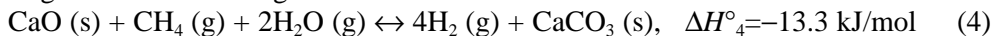
The study attempts to use calcium oxide (CaO) carbonation to remove carbon dioxide (CO₂) from the reformed gas and fix it.

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.

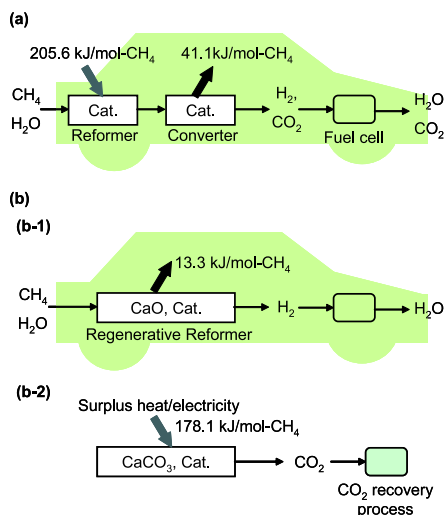
Regenerative reforming:



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°_1 . The proposed process is shown in Figure 2 (b). This process consists of a reforming process (Figure 2 (b-1)) while the vehicle

is driving and a regenerating process (Figure 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.

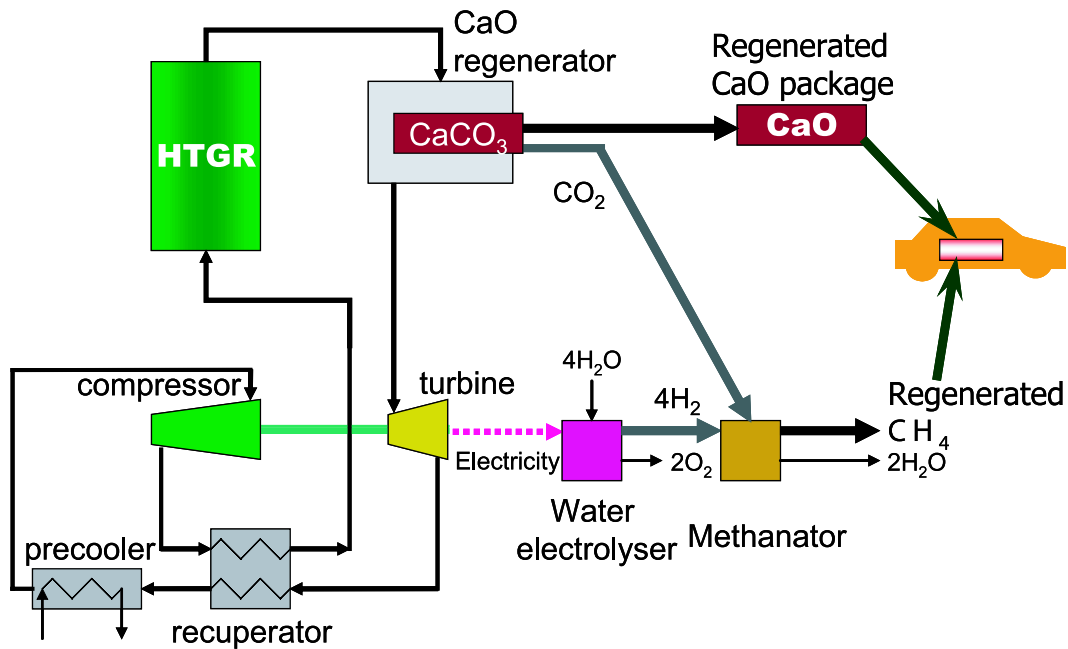
Figure 2. Concept of a zero CO₂ emission FC vehicle using a thermally regenerative reformer; (a) conventional reforming, (b) proposed thermally regenerative reforming, (b-1) reforming mode, (b-2) regenerating and CO₂ recovering mode



Zero CO₂ Emission chemical H₂ Carrier System

The concept of a zero CO₂ emission chemical H₂ carrier system using the regenerative reforming process depicted in Figure 2(b) is proposed in Figure 3. The zero CO₂ emission system consists of FC vehicles using packages of the regenerative reformer, a decentralised package regeneration station, and power systems for energy supply to the system. 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. 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

Figure 4. Combination of the zero CO₂ emission H₂ carrier system using the thermally regenerative reformer driven with a high-temperature gas reactor.



Combination with nuclear reactors

The H₂ carrier system has compatibility with HTGR. Figure 4 shows combination with the H₂ carrier system with an HTGR. The HTGR thermal output is utilised in cascade at the CaO regenerator and gas-turbine. CaO package and hydrocarbon fuels are regenerated in the system. CaO regenerator is placed in the primary coolant loop of the HTGR. Regeneration of CaO from CaCO₃ of the package is proceeded in the reactor. CaO regeneration process is relatively safe process, and then the reactor can be placed in the primary loop directly. The coolant is used secondary at gas turbine for electricity production. Electricity output is used in a water electrolyser for hydrogen production. CO₂ generated from the regenerator and H₂ from the electrolyser are supplied into a methaneator, and then, CH₄ is regenerated. Finally this process regenerates CaO and the reforming fuel.

The HTGR is zero CO₂ emission energy source to establish zero CO₂ emission H₂ carrier system. Although the carrier system is applicable to use renewable energy, nuclear reactor has good combination with the H₂ carrier system on the stand point of stable and large-enough energy supply.

Performance Evaluation

Evaluation of the chemical H₂ carrier system

To evaluate the advantage of the chemical H₂ carrier system depicted in Figure 4, the system was compared with conventional H₂ production process using water electrolysis. Enthalpy balances of those systems per 1 mole of H₂ production were shown in Figure 5. Conventional water electrolysis in Figure 5(a) consumes electricity for water electrolysis process of 282 kJ-electric/H₂-mol, and H₂ compression of 29 kJ-e/H₂-mol. The compression is assumed isentropic and 5 stages compression up to 700 bar. Total enthalpy of 311 kJ/H₂-mol is required. The H₂ carrier system in Figure 4(b) needs

thermal input of 44.6 kJ-thermal/H₂-mol for CaO regeneration, electricity input of 282 kJ-e/H₂-mol for water electrolysis, and 4.9 kJ-e/H₄-mol equivalent for CH₄ compression to 175 bar. The compression is assumed also isentropic and 5 stages compression. Enthalpy of 332 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 produces exothermically thermal output of 41.1 kJ-t/H₂-mol at around 300-700 °C. When thermal output from the methanation is utilized in some heating process, total enthalpy consumption would be reduced. Enthalpy input for water electrolysis process is dominant in total input at both the conventional and the carrier systems. On-board reforming in Figure 5(c) needs smaller enthalpy input than one of other systems which have water electrolysis process. However, CO₂ is emitted from the former system.

Figure 5. Enthalpy balance of H₂ systems, (a) conventional H₂ system using water electrolysis, (b) the zero CO₂ emission H₂ carrier system, (c) conventional methane steam reforming.

* Isenthalpic & 5 stages compression up to 700 bar. ** Isenthalpic & 5 stages compression up to 175 bar.

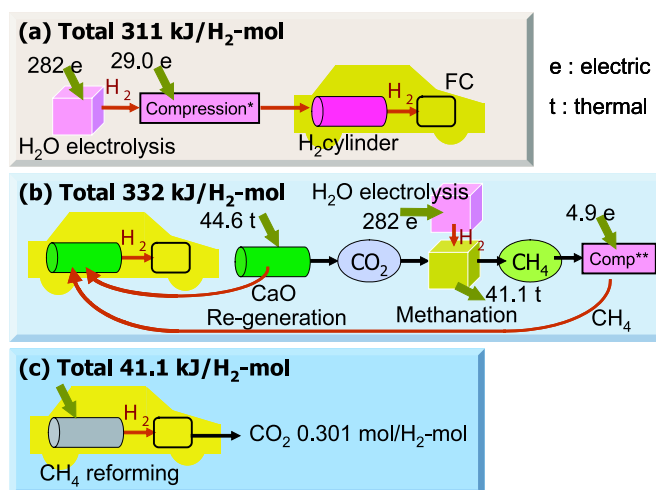


Table 1. Estimated contribution of an HTGR power plant on hydrogen systems

	Proposed H ₂ carrier system	Conventional water electrolysis system
HTGR thermal power ¹		600 MWt
Outlet coolant temperature from HTGR		850°C
HTGR operation duration for hydrogen processes		8 h/day
Inlet/outlet coolant temperature for CaO regenerator	850/835°C	-
Inlet/outlet coolant temperature for gas-turbine	835/587°C	850/587°C
Power input for CaO regeneration	34.8 MWt	-
Plant power input for gas-turbine power generation	565.2 MWt	600.0 MWt
Gas-turbine power efficiency ²	44.3 %	45.0 %
Water electrolysis efficiency (ΔH base) ³		90 %
Compression pressure	175 bar-CH ₄	700 bar-H ₂
Power for H ₂ electrolysis	245.8 MWe	247.1 MWe
Power for compression	3.8 MWe-CH ₄	22.9 MWe-H ₂
H ₂ production/equivalent	2.26E+07 H ₂ -mol equiv.	2.27E+07 H ₂ -mol
Number of FC vehicles for 100 km mileage each ⁴	4.52E+04 -	4.54E+04 -

1. Based on GTHTR300 [7], 2. Power efficiency for the carrier system is estimated by Carnot's efficiency ratio of turbines between the carrier system and the conventional electrolysis system. 3. Ref. [8], 4. It is assumed that H₂ of 500 mol is required for 100 km mileage.

Estimation of the system combined with HTGR

Thermal feasibility of the H₂ carrier system and conventional water electrolysis systems depicted in Figure 5 was evaluated. Optimized energy balances of both hydrogen systems based on HTGR is shown in Table 1. It was assumed that the gas turbine high temperature reactor named as GTHTR 300 designed by JAERI [7] circulating helium coolant was used as the HTGR in the estimation. Off-peak output of 600 MWt for 8 h/day from the HTGR was used for the regeneration of the reformer and hydrogen production.

In conventional water electrolysis system, thermal output from the HTGR at 850°C is used at a gas turbine for electricity power generation with efficiency of 45.0% [7]. Hydrogen is produced by water electrolysis consuming the electric power with efficiency of 90% [8]. Produced hydrogen is compressed for on-board use up to 700 bar by a compressor consuming a part of the electric power. In the chemical H₂ carrier system, thermal output from HTGR between 835°C and 850°C is used for CaO regeneration firstly, and rest of heat is consumed for power production at the gas turbine. Power efficiency for the carrier system is estimated by Carnot's efficiency ratio between the carrier system and the electrolysis system, which is calculated from both inlet coolant temperatures for the turbine.

Hydrogen is produced by the same process of the water electrolysis system [8]. Methane is produced exothermically from recovered CO₂ and produced H₂. Produced methane is compressed for on-board use up to 175 bar by a compressor consuming a part of the power. Finally the carrier system is capable to supply hydrogen of 2.26x10⁷ mol to FC vehicles of 4.52 x10⁴, assuming that H₂ of 500 mol is required for 100 km mileage.

The vehicle number is almost same with the value of the conventional electrolysis system of 4.54×10^4 . Required amounts of electric and thermal energy are shown also in the table. Because thermal energy consumption for CaO is only 5.8% of the whole HTGR output, and the compression work is fairly smaller than one of the electrolysis system, then, energy for both hydrogen production processes are closed, and finally both vehicles number are quite similar. It is reduced from the evaluation that the carrier system can deliver hydrogen to FC vehicles on-board under the same efficiency with conventional water electrolysis system. The carrier system stores fuel source, CH₄, under relatively lower pressure at 1/4 of the storage pressure for the conventional electrolysis H₂ system. The result shows that the H₂ carrier system has advantage to conventional H₂ system in the point of the reduction of fuel storage pressure and the compressor work cost. The regenerative reforming is applicable for higher-hydrocarbon fuels, such as propane and kerosene. Especially the carrier system based on liquid fuel would be candidates of attractive hydrogen carrier media because the compression work would be reduced more.

Conclusion

The proposed zero CO₂ emission H₂ carrier system for FC vehicles using a regenerative fuel reformer based on nuclear power has unique performance comparing with conventional H₂ production systems.

The carrier system is capable to reduce hydrogen media storage pressure, and realise more safety H₂ transportation to FC vehicle.

The carrier system has good compatibility with a high temperature gas reactor, because the regeneration process, which is relatively safe, can be applicable at the primary loop of the reactor. Off-peak power of 600 MWt for 8 h from a reactor can supply H₂ for FC vehicles of 45 thousands, which is the similar number for conventional electrolysis system under the same thermal input from the reactor. The carrier system shows a new possibility of chemical energy carrier system.

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SESSION V

BASIC AND APPLIED SCIENCE IN SUPPORT OF NUCLEAR HYDROGEN PRODUCTION

Chairs: Y. Kato, P. Anzieu, Y. Sun

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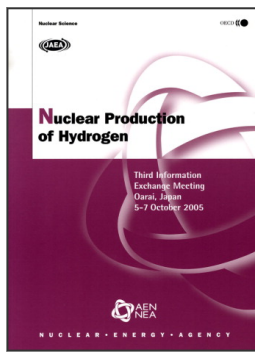
SESSION V

Basic and Applied Science in Support of Nuclear Hydrogen Production

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