NUCLEAR HEAT UTILIZATION FOR NATURAL GAS STEAM REFORMING TO PRODUCE HYDROGEN

by
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ABSTRACT

The assessment of nuclear heat utilization for natural gas steam reforming to produce hydrogen has been carried out. Most of hydrogen production in the world, is produced by steam reforming of natural gas. This process is an endothermic reaction at high temperature that needs a huge amount of heat energy to proceed the reaction. Conventionally, the heat energy needed is supplied by direct burning of fossil fuel. If the huge amount of those heat energy can be substituted by nuclear process heat, some advantages can be obtained such as, reducing combustion of fossil fuels that give implication of significant decreasing of CO_2 *emission to the environment. On application of nuclear process heat to steam reforming of natural gas, there are some inferior conditions related to the limitation of temperature and pressure provided by nuclear reactor which directly gives impact on lower thermal efficiency (~50%) compared to the fossil-fuelled plant (80-85%). Some modification design and operation of reformer can improve the lack condition, and capable to increase the thermal efficiency of nuclear heated natural gas steam reformer become about 78%.*

Keywords: *nuclear process heat, natural gas steam reforming, HTGR reactor*

I. INTRODUCTION

In recent years, a rapid increase in CO_2 emission resulted mainly from increasing of fossil fuel consumption as energy sources bring about a significant increase in global temperature, that is, global warming. The air temperature is estimated to increase by approximately 2°C in the year of 2050. While, fossil resources are not unlimited and, in particular, oil is said to be consumed out in the middle of this century for the annual consumption rate as of the year 2000 (US-DOE, 2002). Since early of 2000, Indonesia has become the net oil importer country, because of the rapid growth of the fuel consumption exceeds national oil production. Hence, some alternative energy should be assessed to substitute the use of fossil resources, including production synthetic fuel from coal, and hydrogen production by utilization of nuclear heat.

Hydrogen is of great importance among clean energy sources, is not only due primarily to its high energy density but also offer potential attractiveness because of production capability from an abundantly recyclable resource of water nor from fossil resources. However, high thermal efficiency of hydrogen productions from water is still under development (Walter, L., et. al., 2002; and Charles, W.F., 2002). At present, almost hydrogen demand is produced from the process of natural gas steam reforming. This process is endothermic chemical reaction, operating at high temperature, and consequently requires heat energy process at high temperature. Conventionally, the need of the huge amount of heat energy is supplied by direct burning of fossil fuel. However, this method has disadvantage of releasing CO_2 emission and limitation of fossil fuel. If the required heat energy can be substituted by nuclear reactor,

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there will be some advantages such as: fossil fuel burning reduction, which will give implication of decreasing CO_2 emission rate, and diversification of nuclear energy.

High temperature gas cooled reactor (HTGR) is one of the type of advanced nuclear reactor which operates at high temperature and uses helium as cooler. The outlet temperature of helium, about 900°C, can be used for either industrial heat process or to produce electricity by cogeneration system. This reactor is expected to be the most promising reactor to generate the thermal energy up to 950°C without any emission of CO_2 gas and other greenhouse effect gases as well as sulfur oxide and nitrogen oxide gases. Accordingly, hydrogen production by the process of natural gas steam reforming in which an HTGR is used as the heat source is anticipated to play a key role in resolving the global warming and oil shortage problems.

The policy of nuclear energy utilization for electricity generation and cogeneration system in Indonesia (Soentono, S., 2006) is realizing symbiotic and synergetic in utilizing of nuclear energy with renewable and non renewable energy sources to fulfill national demand of energy to support sustainable development. Hence, besides promoting the realization of first nuclear power plant in Indonesia, BATAN also should do some assessment of future utilization of nuclear reactor such as: nuclear cogeneration for desalination, nuclear heat process for industry like hydrogen production, coal liquefaction, coal gasification.

This paper describes the utilization of nuclear process heat for operating steam reforming of natural gas to produce hydrogen. The goal of the study is to understand the couple system of nuclear process heat with steam reforming of natural gas, as the important candidates of high temperature nuclear application for industry. The result of the study can be useful to support the policy of nuclear technology development program in Indonesia.

II. STEAM REFORMING OF NATURAL GAS

Raw material in this process is natural gas. The primary ways in which natural gas, mostly methane,

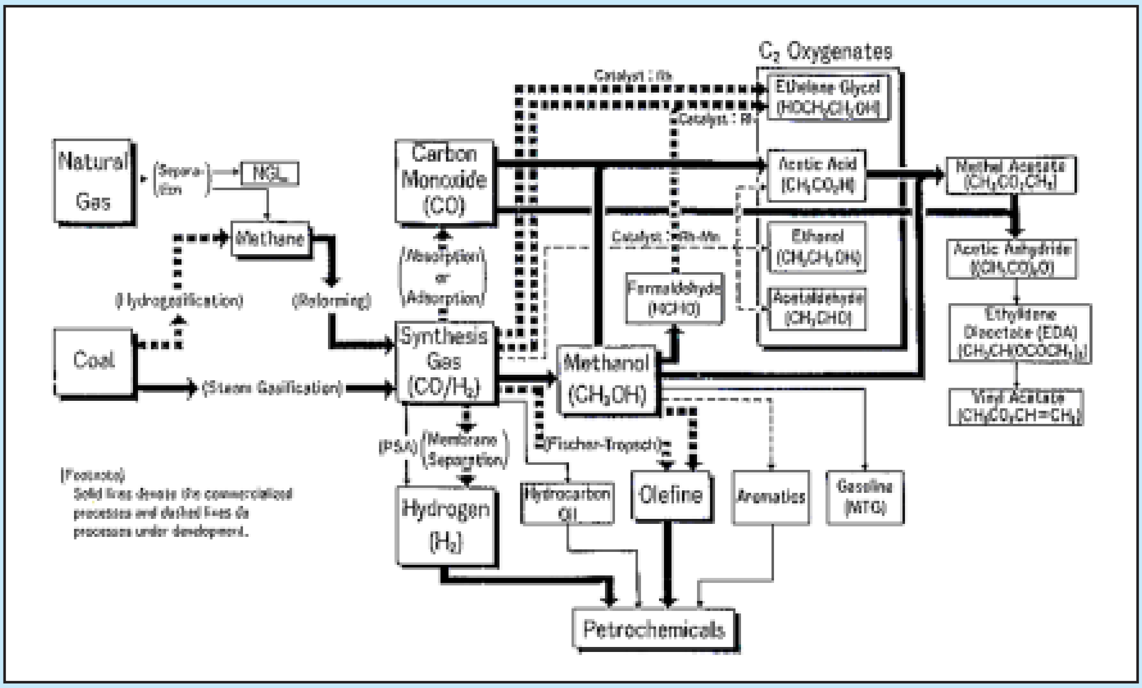
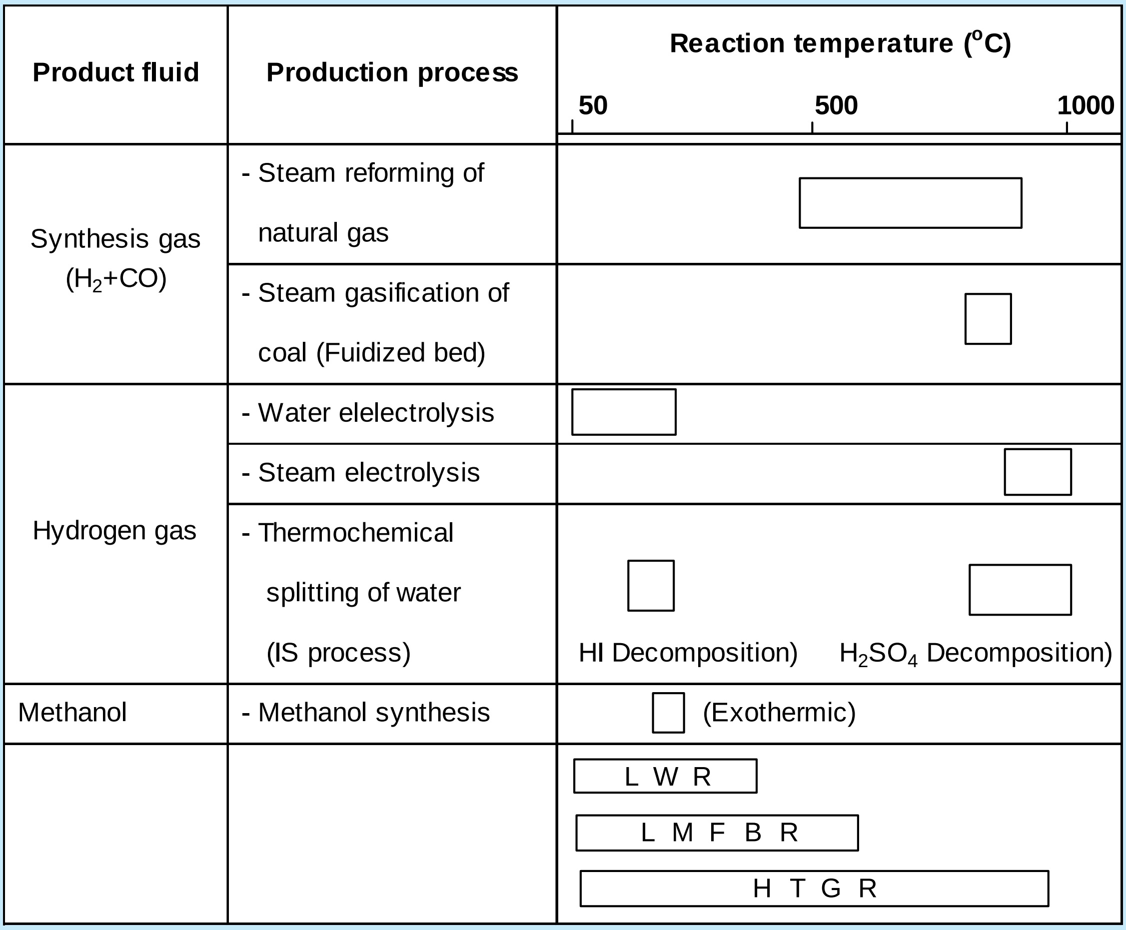


Figure 1
Key Technologies for Synthesis Gas, Hydrogen, and C_1 Chemistry Production

Table 1
Temperature Range of Typical Nuclear Process Heat



Product fluid	**Production process**	**Reaction temperature (°C)** **50** **500** **1000**
Synthesis gas (H_2+CO)	- Steam reforming of natural gas	
	- Steam gasification of coal (Fuidized bed)	
Hydrogen gas	- Water elelectrolysis	
	- Steam electrolysis	
	- Thermochemical splitting of water (IS process)	HI Decomposition) H_2SO_4 Decomposition)
Methanol	- Methanol synthesis	(Exothermic)
		LWR LMFBR HTGR

is converted to hydrogen involves reaction with steam, as follow (Hada, K., et.al., 1992):

$$CH_4 + H_2O = CO + 3H_2 - 206.20 \text{ kJ/mol} \quad (1)$$

$$CH_4 + 2H_2O = CO_2 + 4H_2 - 165.03 \text{ kJ/mol} \quad (2)$$

$$CO + H_2O = CO_2 + H_2 + 41.20 \text{ kJ/mol} \quad (3)$$

In practice, gas mixtures containing carbon monoxide (CO) as well as carbon dioxide (CO_2) and unconverted methane (CH_4) are produced and require further processing. The reaction of CO with steam (water-gas shift) over a catalyst produces additional hydrogen and CO_2, and after purification, high-purity hydrogen (H_2) is recovered. Composition of reformer gases is determined by reaction equilibrium of reaction 1 and 3. While stoichiometry of produced gas is determined by controlling the operation condition of water gas shift reaction (3). To achieve the ideal composition in producing hydrogen, operating condition required at temperature of 800-950°C and pressure of 1-3 MPa. Empirical rule tells that decreasing temperature or increasing pressure decreases the concentration of hydrogen in the equilibrium mixture. Hence the higher pressure and lower temperature of process gas results in less productivity of hydrogen.

Figure 1 shows steam reforming of natural gas which is also well known as state of the art of key technologies for synthesis gas, hydrogen, and methanol production and also for other C_1 chemical feedstock (Hada, K., et.al., 1992). Recently, more than 85% hydrogen demand is supplied by this process production.

III. NUCLEAR HEATED NATURAL GAS STEAM REFORMING

A. *The Role of Nuclear Process Heat*

At present, process heat is generated by burning fossil fuels even in producing hydrogen. Another critical issue for meaningfully reducing CO_2 emission is how to generate process heat without any emission of CO_2 as a substitute for burning fossil fuels. Nuclear energy is now being used to produce about 17% of the world's electricity. As of 2006, this included 442 nuclear reactors, with total capacity of about 354 GWe. Yet, only a few of these plants are being used to supply hot water and steam. The total capacity for these purposes is only about 5 GWt, and they are operating in just a few countries, mostly in Canada, China, Kazakhstan, Russian, and Ukraine (IAEA TECDOC-1085, 1999).

Specific temperature requirement for nuclear heat application range from low temperature (~ room temperature), for application such as hot water and steam for agro industry, district heating, and sea water desalination, to ~1000^{o}C for process steam and heat for the chemical industry (i.e. natural gas steam reforming) and high pressure injection steam for enhanced oil recovery, oil refinery process, and refinement of coal and lignite. One of the advanced nuclear reactor, called as high temperature gas cooled reactor (HTGR) which have capability to provide process heat until 950^{o}C, is appropriate for endothermic chemical process, such as production hydrogen by high temperature electrolysis, production synthesis gas and/or hydrogen by natural gas steam reforming. Table 1 shows nuclear process heat associated with typical temperature of the process for syngas, hydrogen, and methanol production (IAEA TECDOC-1085, 1999).

B. *Nuclear Heat Utilization for Natural Gas Steam Reforming*

Figure 2 illustrates the couple of Japanese HTGR (or HTTR, high temperature testing reactor) with

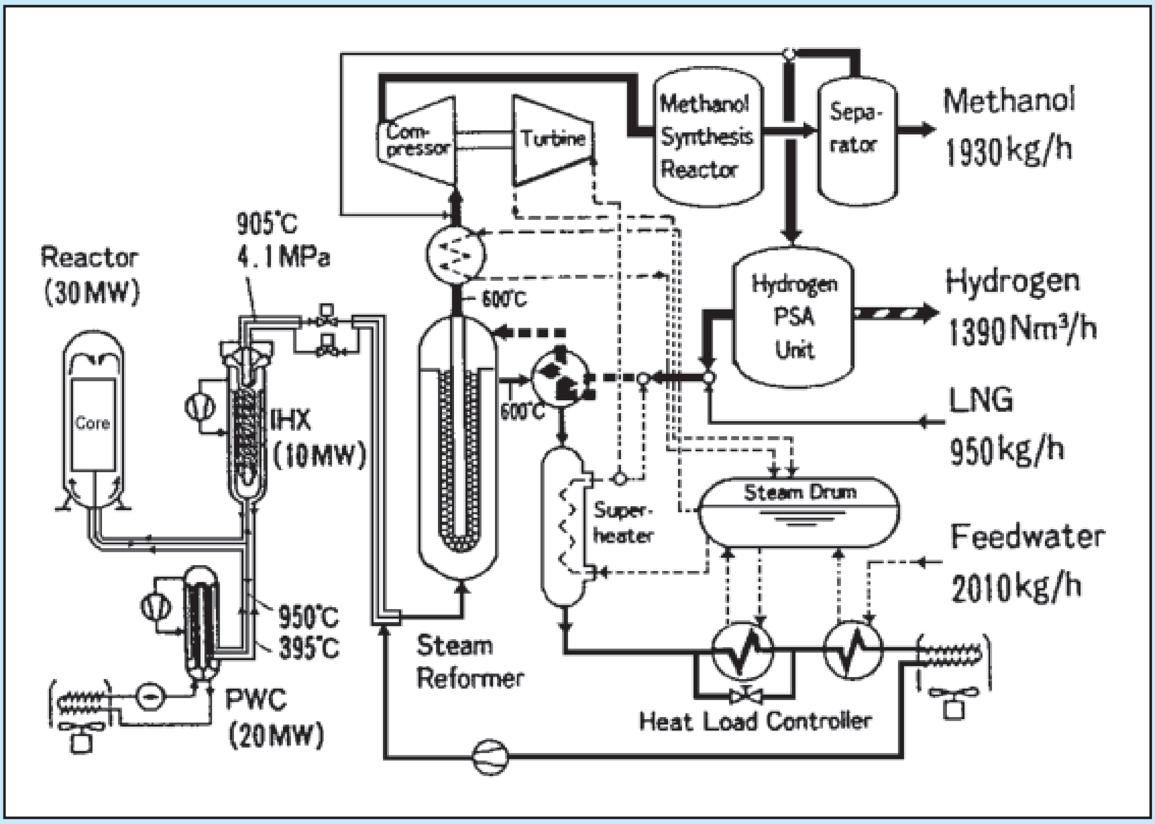


Figure 2
Schematic Illustration of HTTR Heated Natural Gas Steam Reforming

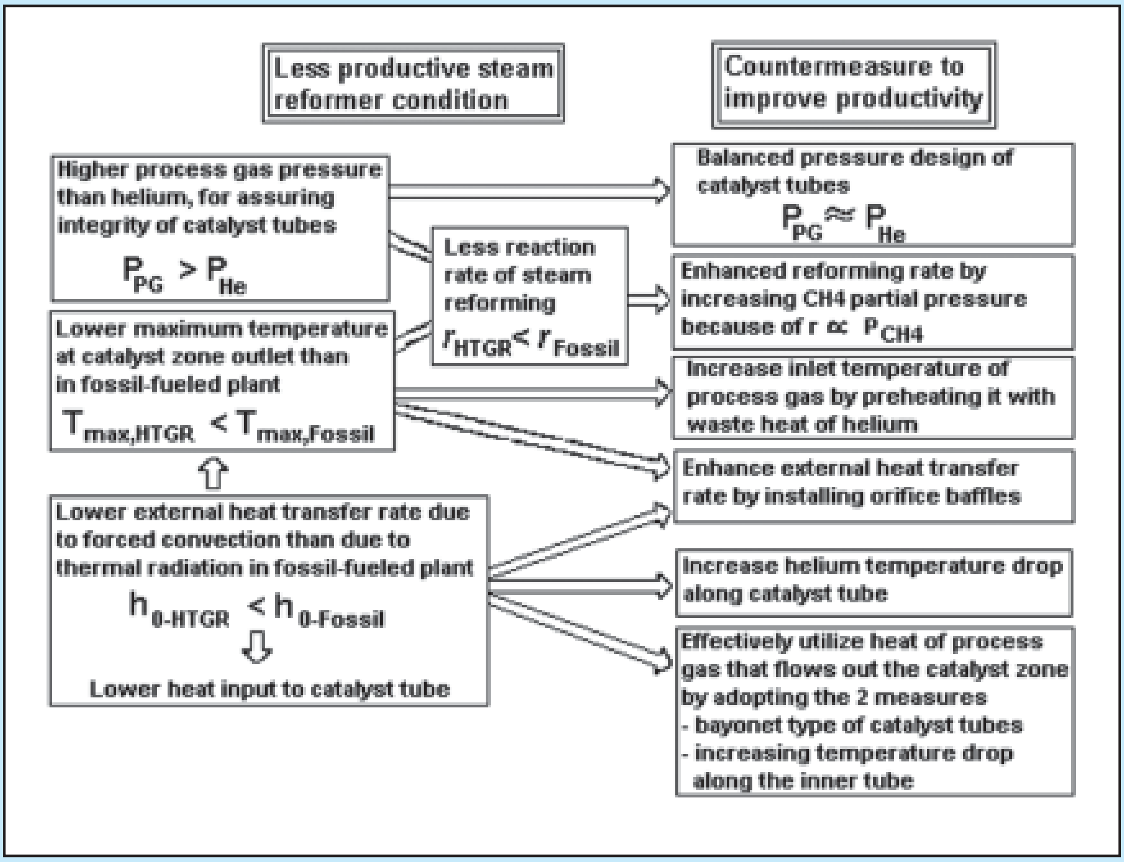


Figure 3
Schematic Diagram of Improvements Nuclear-heated Steam Reformer

natural gas steam reforming plant to produce hydrogen and methanol (Hada, K., et.al., 1992; Masao, H., 2002). The figure show that utilization of high temperature nuclear heat is done by utilize an intermediate heat exchanger (IHX) which also has a function as an interface to separate nuclear zone and chemical process zone. IHX is helium-helium heat exchanger. In IHX, the primary helium from nuclear reactor is flowed to transfer the heat to the secondary helium. This secondary helium which is relatively free from radioactive contaminants is utilized as process heat to fulfill heat requirement at chemical processes. The chemical process at Figure 2 is the conventional natural gas steam reforming which is used widely in almost fertilizer or petrochemical industries worldwide. With the natural gas feed rate of 950 kg/h, the production rate of hydrogen and methanol (as side product) are 2390Nm^3/h and 1930 kg/h respectively.

In conventional process of natural gas steam reforming, the optimum operating conditions is temperature and pressure in the range of 850 – 900^{o}C and 1 – 3 MPa respectively. Decreasing temperature and increasing pressure will reduce hydrogen production. Unfortunately, the outlet temperature of HTGR coolant carried by primary helium is approximately 905^{o}C at high pressure of 4.1 MPa. Due to safety consideration, utilization of helium heated is the secondary helium in Intermediate Heat Exchanger, called as IHX. Since heat loss to the air, secondary helium temperature is about 890^{o}C, lower 15^{o}C than temperature of primary helium coolant of HTGR (905^{o}C). This secondary helium from IHX at 890^{o}C enter the reformer and flows outside the catalyst tubes, while a process feed gas comprises mainly of steam and methane does inside the tubes (see Figure 4). Hence, heat transfer from helium to catalyst tubes is predominantly attributed to forced convection. As the result, the maximum process gas temperature at the catalyst zone barely exceeds 800^{o}C, lower about 50 – 100^{o}C than in fossil fueled plants. Also heat flux to catalyst tubes much lower than 50000 - 80000 W/m^2 in fossil fueled plants, around one fifth to a half of these, about only 10000 – 20000 W/m^2. In conventional fossil-fueled steam methane reformer, catalyst tubes are heated through thermal radiation from burning natural gas or other burnable gas.

The optimum pressure condition for fossil fueled natural gas steam reformer is around 1 – 3 MPa. For safety reason of preventing the possibility of fission-product leakage of the helium gas circuit, and to ensure structural integrity of catalyst tube in high temperature and pressure, the pressure of nuclear heated plant (4.5 MPa) must be set higher than the pressure of secondary helium (4.1 MPa). Ones more, this higher pressure condition than conventional plants give a result less productivity of hydrogen, because the optimum pressure condition in the established conventional plant is in the range of 1 – 3 MPa.

IV. IMPROVEMENT OF NUCLEAR HEATED STEAM REFORMING

As mentioned before, Figure 3 illustrates the schematic diagram of lack condition of nuclear heated steam reforming of natural gas (Hada, K., et.al., 1992; Masao, H., Shiozawa, S., 2005; Fujimoto, et.al., 2005). To counter this inferior condition resulting lower thermal efficiency of nuclear heated plant, some countermeasures are applied to improve the productivity. The countermeasures include:

- Balance pressure. It is a natural idea to reduce the process gas pressure P_{PG} close to the helium gas pressure P_{He}, that is, balance pressure$P_{PG} \approx P_{He}$. The design-by-analysis realizes the reasonable application of the balanced pressure design in which the balanced pressure between primary and secondary side fluids is applied to the design of structural part so as to drastically reduce the primary stress and its design pressure is specified based on the differential pressure but not on the maximum of both fluid pressure values. The catalyst tube wall thickness of 16 mm meets requirement on design limits for pressure retaining.
- Enhanced of reforming rate by increasing a partial pressure of methane. Many chemical researches have proposed rate equation for steam methane reforming reaction. The following expression is recognized the basic pseudo-first-order form.

where $-r_{CH4}$: demethanation rate
k : rate constant
P_{CH4}, P_{H2}, .. : partial pressure of methane, hydrogen, and others
$P_{CH4,E}$: equilibrium methane partial pressure

The basic equation means that demethanation rate is roughly proportional to the difference between the actual and equilibrium methane partial pressure.

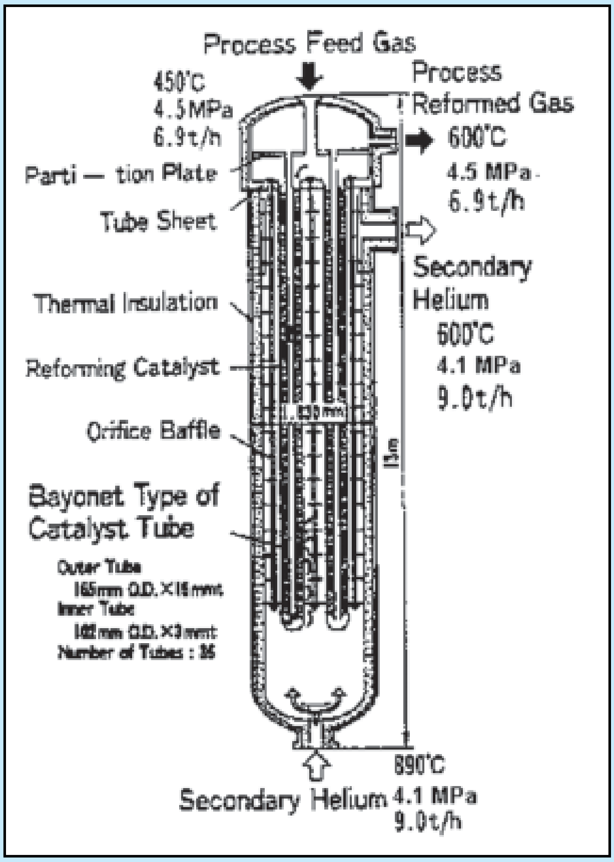


Figure 4
Sectional View of Steam Reformer

Table 2
Comparison of Steam Reformer Performance

Reformer type	Fossil-fueled	Helium-heated	Improved helium-heated
Process gas pressure	1 – 3 MPa	> Helium pressure P_{He} of 4.1 MPa	Balanced Pressure ($\cong P_{He}$) → 4.5 MPa at the inlet of steam reformer
Maximum process gas temperature, ^{o}C	850 – 900	≤800	830
Maximum heat flux to catalyst zone, W/m^2	50000 – 80000	10000 – 20000	40000
Steam reformer thermal efficiency, %	80 ~ 85	~50	78

- Preheating process feed gas is done by helium for the purpose of increasing process gas temperature over the catalyst zone. Feed gas of steam and methane is preheated to 450^{o}C before entering the catalyst region of the inner tubes and heated up to 830^{o}C in the catalyst zone. The reformed gas transfers its heat to the process gas when flows up in the center tube. Lowering helium outlet temperature also will increase heat input from helium gas to catalyst tubes. It can be done by utilizing flowing out helium from steam reformer, to preheat the process gas to 450^{o}C as mentioned above.
- Enhancement of external heat transfer rate. At a high temperature, less heat transfer rate by single gas phase forced convection than by thermal radiation results in lower heat flux of heat input to catalyst tubes. In fossil fueled reformer, the type of shell-and-tube of heat exchanger with straight catalyst tube should be applied because of easily handling catalyst. Forced convection heat transfer is capable of being enhanced mainly by means of cross flow, accelerating local flow velocity and inducing flow disturbance. Japanese design adopted the bayonet type of reformer tubes and embodies segmental baffles with orifice baffles.

The sectional diagram of the improved nuclear steam reformer is shown in Figure 4 (Hada, K., et.al., 1992; Fujimoto, N., et.al., 1992). The secondary helium at the temperature of 890^{o}C, flows into the steam reformer at the bottom and then flows up outside the catalyst tubes, squeezed by multiple plates of orifice baffles. Finally, the helium flows out to a super-heater at the temperature of 600^{o}C. On the other side, the process gas is preheated to 450^{o}C at a balance pressure of 4.5 MPa, higher than helium pressure of 4.1 MPa and then flows into the steam reformer from the top. It flows down through catalyst in annuli of the bayonet type of catalyst tubes, and methane, other light hydrocarbons and steam of the process gas are reformed to hydrogen, carbon monoxide and carbon dioxide. The reformed gas, then, flows up inside the inner tubes and transferred its heat to the reforming process gas across the tube wall. Finally, the reformed gas flows out at the top of the reformer at the pressure of 4.1MPa little bit higher than helium so as to ensure the structural integrity of tubes. The tubes are made of Hastelloy XR, a Nickel-base Cr-Mo-Fe heat resistant super alloy, which has industrially applied to high temperature structural parts of the IHX.

As the result of some improvements above, the thermal efficiency of nuclear heated steam reforming become higher than the system without improvements. Table 2 shows a comparison of natural gas steam reforming performance among fossil fuel-heated, helium-heated, and the improved helium heated steam reformers (Hada, K., et.al., 1992; Salimy, D.H., 1994; Masao, H., Shiozawa, S., 2005).

V. DISCUSSION

Recently, any high nuclear process heat utilization, including the nuclear hydrogen production by natural gas steam reforming process has not been operated in the world, even though a wide range variety of research and development works have been devoted to realizing it. The establishment of Nuclear Hydrogen Society in Japan on 2001 triggers the extensive collaboration between scientists and engineers from hydrogen and nuclear fields in promoting the realizations of nuclear utilization to produce hydrogen.

Beside Japan, some countries like China, United State, and South Africa are the countries that develop R and D in utilizing nuclear process heat to produce hydrogen by process of methane steam reforming (IAEA TECDOC 1085, 1999; US-DOE; 2002). For the example, China recently operate the HTGR type reactor named HTR-10 and do extensive study in developing the process technology of fossil fuel conversion such as coal liquefaction/gasification to produce clean transportation fuels and or hydrogen by utilizing nuclear process heat as heat energy sources. South Africa also develops the conceptual design of commercial HTGR called as PBMR (Pebble Bed Modular Reactor). Some study to couple PBMR with steam methane reforming have been carried out to realize the application nuclear reactor for non electricity purposes (Bolthrunis, C. O., et.al., 2006; Kriel, W., et.al., 2006).

The most advanced country in developing the technique of producing hydrogen by nuclear process heat is Japan. Since the mid of 1990 decade, Japan already operating the engineering testing HTGR which is officially called as the High Temperature Engineering Testing Reactor, HTTR. One of the most creative objectives of the HTTR is to demonstrate high efficiency of CO_2 free application of high temperature nuclear process heat with utilizing HTGR, and provide technical information for proceeding the commercial deployment of HTGR process heat utilization. Recently, besides operating HTTR, Japan has already built demonstration plant of natural gas steam reforming nearby HTTR. Some experiment associated to couple HTTR with reforming plant has been proceeding extensively since early of 2000. A computational prediction suggests that commercialization of the HTGR process heat utilization be required to be achieved in the year of 2015 or earlier so as effectively reduce CO_2 emission. Demonstration plant of HTTR heated hydrogen production should be made in the beginning of the early 2010 decade (Masao, H., Shiozawa, S., 2005). This is become the first demonstration plant of symbiotic utilization between nuclear process heat and chemical process in the world.

Indonesia is the country that has abundant natural energy resources like natural gas, oil, and coal. The direct burning of these resources to provide process heat for industrial process is a wasting if the process heat can be substituted by nuclear reactors. From the environment point of view, the direct burning will increase the CO_2 emission rate. Unfortunately, the nuclear energy program in Indonesia is constrained by crucial factor including financial and pros-and-cons problems. If the utilization of nuclear energy in Indonesia is not realizing soon, Indonesia will be left behind other countries, due to the rapid development in nuclear energy utilization. In near future, nuclear reactor is not only to produce electricity, but also develop as heat process to produce hydrogen, coal liquefaction, coal gasification, and so on. Anyhow, the development of nuclear energy utilization in the world should be followed continually. The evolution in utilization of nuclear reactor should be anticipated in supporting the national policy in development of nuclear energy in Indonesia.

VI. CONCLUDING REMARKS

- Steam reforming of natural gas to produce hydrogen is one of the promising candidates for the nuclear process heat utilization, since the products of hydrogen is an alternative fuels in the future to relax the global warming.
- High Temperature Gas Cooled Reactor, HTGR, is expected to play an important role in the utilization nuclear process heat at high temperature. This heat energy can substitute the conventionally direct burning of fossil fuels.
- Application of nuclear process heat for industrial purposes are advantage from the point of reducing direct burning of fossil fuels, which give an implication to decrease the rate emission of CO_2.

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