

CONVERSION OF CO₂ TO HYDROCARBON SYNFUEL BY UTILIZING NUCLEAR HYDROGEN COGENERATION

APLIKASI KOGENERASI HIDROGEN NUKLIR UNTUK KONVERSI CO₂ MENJADI BAHAN BAKAR SINTETIS HIDOKARBON.

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ABSTRAK

Telah dilakukan studi pemanfaatan kogenerasi hidrogen dengan energi nuklir sebagai teknologi kunci untuk proses konversi CO₂ menjadi bahan bakar cair sintetis hidrokarbon. Tujuan studi adalah untuk memahami konversi CO₂ dan H₂ menjadi bahan bakar sintetis, serta peran kogenerasi hidrogen nuklir untuk produksi hidrogen dan sebagai sumber energi panas proses. Metode yang digunakan adalah studi pustaka berdasarkan hasil penelitian yang sudah ada. Secara konvensional, produksi bahan bakar sintetis dari batubara diproduksi melalui proses gasifikasi batubara, dilanjutkan dengan mereaksikan gas sintesis (campuran CO dan H₂) dalam reaktor Fischer-Tropsch menjadi bahan bakar sintesis. Dalam studi ini, dipelajari produksi bahan bakar sintetis dengan bahan baku CO₂ dan H₂. CO₂ berasal dari emisi pembangkit berbahan bakar batubara, sedangkan H₂ dihasilkan dengan sistem kogenerasi hidrogen nuklir. Hasil kajian menunjukkan bahwa dibanding proses konvensional, proses berbasis CO₂ dan H₂ dengan didukung sistem kogenerasi memberikan keuntungan yang sangat signifikan dari sisi emisi CO₂. Proses berbasis gasifikasi batubara dan kogenerasi nuklir, mampu menurunkan emisi sampai 75% dan menghemat konsumsi batubara sampai 40%. Sedang proses hanya berbasis CO₂ dan H₂ kogenerasi nuklir tanpa batubara, sama sekali tidak mengemisi CO₂ bahkan dapat memanfaatkan emisi CO₂ pembangkit listrik batubara sebagai bahan baku proses.

Kata Kunci: sistem kogenerasi nuklir, konversi CO₂, emisi CO₂, bahan bakar sintetis

ABSTRACT

A study of the utilization of hydrogen cogeneration with nuclear energy as a technology for the conversion of CO₂ into synthetic liquid hydrocarbon fuels has been carried out. The aim of the study is to understand the conversion of CO₂ and H₂ into synthetic fuels, as well as the role of nuclear hydrogen cogeneration for the production of hydrogen and as a source of process heat energy. The method used is literature study based on the results of existing research. Conventionally, synthetic fuel production from coal is produced through coal gasification process, followed by reacting synthesis gas (mixture of CO and H₂) in FT reactor to synthesis fuel. In this study, we studied the production of synthetic fuels with CO₂ and H₂ raw materials. CO₂ comes from emissions of coal-fired plants, whereas H₂ is produced by nuclear hydrogen cogeneration systems. The results show that compared to conventional processes, CO₂ and H₂-based processes supported by coal cogeneration systems provide significant advantages in terms of CO₂ emissions. The process based on coal gasification and nuclear cogeneration, capable of reducing emissions by up to 75% and saving up to 40% of coal consumption. While the process based only on CO₂

and nuclear hydrogen cogeneration (without coal gasification), teoretically can operate witout any CO₂ emission at all. Even this process can captured and utilize CO₂ emissions from coal fired plant, and use it as a raw material for the process.

Keywords: nuclear cogeneration system, CO₂ conversion, CO₂ emission, synfuel

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I. INTRODUCTION

Cuurrently, fossil fuels are still the primary energy source of the earth's surface, although it has since long been recognized fossil fuels are not renewable materials and a time will run out. Among fossil fuels, liquid type fuel is the fuel most used primarily for the transportation sector. Due to the pace of demand always exceeds the production rate, various means are used to meet the very high demand. Converting solid form of energy source (coal) to liquid (petrol, diesel) is an alternative to support the supply demand continues to increase.

Conventionally, synfuel can be made by converting coal (Patzek 2009; Xu et al. 2013). In principle, the production begins with the coal gasification process to produce syngas, a mixture of CO and H₂. Then the Fischer-Tropsch (F-T) process, can produce a variety of products derived, one of them is hydrocarbon synfuels. In the conventional process, either CO, or CO₂ and H₂ are produced from the coal gasification process as an intermediate product before converting them to synfuels. In several decades, the process was carried out by reacting H₂ and CO₂ originating from the outside of the system. CO₂ emissions can be obtained from industrial or power plants, which is abundant and to date become the problem because it is a greenhouse gas that is damaging to the environment. While the H₂ can be produced from the decomposition of water, electrolysis or steam reforming of natural gas.

Nuclear cogeneration for the decomposition of water is thermochemical hydrogen production process by utilizing nuclear energy as a source of heat energy to run the process (Merajin et al. 2014; Borgard & Tabarant 2011). Among the decomposition of water by nuclear, process with iodine-sulfur cycle is relatively advanced R & D process. The process was originally developed by General Atomic in America in 1970, then adopted and developed by

several countries such as Japan, Germany, China, France, and South Korea. In addition, the United States, and South Africa are also developing a hybrid process cycle of sulfur to produce hydrogen with nuclear energy. Hybrid process is combination of electrolysis and thermochemical processes. Nuclear cogeneration system is also used to develop high-temperature electrolysis process, which is believed to generate hydrogen with better thermal efficiency.

The issue of global warming induced by CO₂ emissions has become a global issue that is increasingly important in the world and is considered as the main cause of climate change and the negative impact on human life. One of the technologies used are carbon capture and storage technologies are often referred to as Carbon Capture and Storage (CCS). In principle, the CCS technology is to capture emissions out of the emission sources (plant or power plant), to separate CO₂ from other gases, and then transporting and storing the CO₂ to CO₂ storage (Hong et al. 2015).

Since several decades, CCS technology developed to the technology Carbon Capture and utilization (CCU) (Yan at al. 2014). In the CCU technology, the captured CO₂ will be used as industrial raw materials. In this way, beside getting the the advantages obtained in the form of decreasing the rate of emission of CO₂, the process also obtained added value from the use of CO₂ for industrial processes.

This paper discusses the process production of liquid hydrocarbon synfuel from H₂ and CO₂, by utilizing nuclear as a heat source, because it is believed that nuclear energy to an important source of heat energy to produce hydrogen. While the use of CO₂ as a raw material becomes an important part solution addressing greenhouse gas emissions. Conventional processes based on coal gasification technology is used as a benchmark, to analyze the positive aspects of the alternative process in terms of environmental issue.

II. METHODOLOGY

Nuclear Cogeneration for Co₂ Conversion to Hydrocarbon Synfuel

This chapter describes an alternatif technology to produce hydrocarbon synfuel from CO₂ and H₂. An assumption used is that H₂ obtained from the process of nuclear water splitting, while CO₂ come from emission from coal filled plant. The conventional technologi of hydrocarbon synfuel production is used as comparison.

A. Nuclear Hydrogen Cogeneration

Cogeneration, also known as combined heat and power (CHP), is very efficient, clean, and reliable approach to generate power and thermal energy from a single fuel source (Khamis 2013). Countries embarking on nuclear power should consider cogeneration and the use of waste heat from a nuclear power plant (NPP) to increase energy utilization and overall efficiency. In a number of countries, cogeneration and heat production using nuclear reactors is an effective way to meet the variety types of energy needs. Currently, there are 79 reactors operating the cogeneration mode. In addition, is the potential for applying this technology more widely promising (Hong et al. 2015). Heat applications cover a wide range of specific temperature requirements starting from low temperatures i.e., just above room temperature for applications such as hot water and steam for agro-industry, district heating, and sea water desalination; reaching more than 1000°C for process steam and heat applications i.e., for chemical industry and high-pressure injection steam, enhanced oil recovery, oil shale and oil sand processing, oil refinery, refinement of coal and lignite, and water splitting for the production of hydrogen. With the rapid increase in energy demand (for both electricity and heat), concern over global warming could pave the way for nuclear energy to exert a major positive impact on energy security and climate change.

The R & D of nuclear water splitting to produce hydrogen from water has been developed very intensively in the developed countries. One of the promising process is the process of water splitting thermochemical iodine-sulfur cycle (IAEA 2012; Noguchi et al. 2016). The process was originally developed by General Atomics in America in the 1970s. The technique then adopted and developed by several countries such as Japan, Germany, China, France, and South Korea. The other process is hybrid process that known as sulfur cycle developed by Westinghouse (IAEA 2013). The hybride process is the combination between thermochemical and electrolysis. This process was also developed intensively in South Africa (Botha 2012; Telsnig et al. 2013).

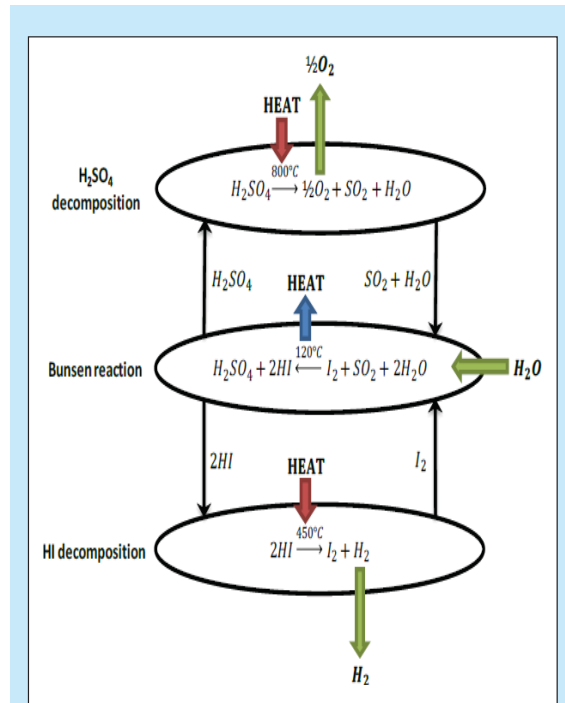


Figure 1
The sulphur iodine cycle (Botha, 2012).

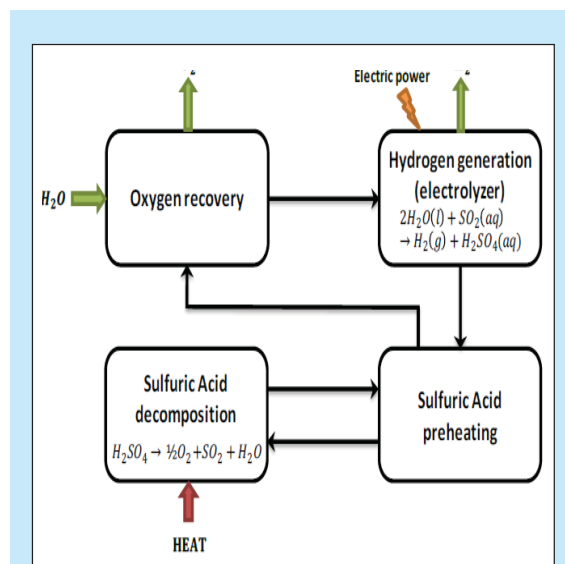


Figure 2
The hybride sulphur cycle (Botha, 2012).

The sulphur-iodine (SI) cycle consists of three steps, as shown in Figure 1. The second step occurs at 800°C and requires the addition of heat. The High Temperature Gas-cooled Reactor (HTGR) is the only one type of nuclear reactor that have capability to supply energy at thus temperature (Khamis et al. 2013; Verfondern et al. 2017).

The Hybrid Sulphur (HyS) cycle, is the combination of thermal and electricity processes. The HyS cycle is shown in Figure 1. Consists of 2 steps, in the first step, water and sulphur dioxide is electrolysed at around 87°C to produce hydrogen and sulphuric acid. The second step is the decomposition of sulphuric acid into sulphur dioxide, steam and oxygen at around 800°C. Optimisation studies suggested that thermo-dynamic efficiency for the HyS cycle of 47% is possible by using a temperature of 930°C and 10 bar pressure in the decomposition step (Botha 2012; Chiuta 2010). Again, HTGR is the only one type of nuclear reactor that have capability to supply energy at thus temperature.

B. Capture and Utilization of CO₂

The emergence of the concept of CO₂ captured and storage (CCS) as a potentially viable technology for the reduction of greenhouse gas emissions (GHG) emissions in the world have spawned a parallel interest in the concept known as the CO₂ captured and utilization (CCU) [16]. In contrast to the CCS which only mitigate, CCU seeks to convert CO₂ on a large scale into value-added products. In other words, CO₂ is not to be stored or mitigate but used as raw materials. In spite of carbon dioxide is considered to be a thermodynamically and chemically stable molecule under standard conditions, it can react with

other chemical feedstocks given sufficient energy or using a catalyst to produce value added commodity chemicals (Aresta et al. 2013) and fuels (Jiang 2010).

In principle, the CCS is technology to capture and separate CO₂ from other gases, and then transported to another place to be stored in the stable geological disposal (Telsnig et al. 2013; Li et al. 2013). While, in the CCU technology, captured CO₂ will be used as raw materials for industries. In this way, beside the advantages of a decreasing the rate of CO₂ emissions, also obtained added value of CO₂ as raw materials. Some industries that use CO₂ as the raw material become the target of this program.

The concept of CO₂-recycled synthetic hydrocarbon fuels is not new. Likely spurred by the oil crisis of the 1970s, Steinberg and Dang first envisioned the closed-loop version of CO₂-recycled synthetic fuels with CO₂ captured from ambient air. In a series of journal articles and patents, they explored various options to capture CO₂ from the atmosphere using hydroxide or carbonate absorbents, produce H₂ by water electrolysis, and synthesize methanol by reaction of the CO₂ with H₂, with nuclear fission or fusion supplying the electricity and heat (Graves 2010).

Figure 3 shows the possible pathways of CO₂ and H₂O utilization as raw materials for producing synfuel. This concept is interesting because it is

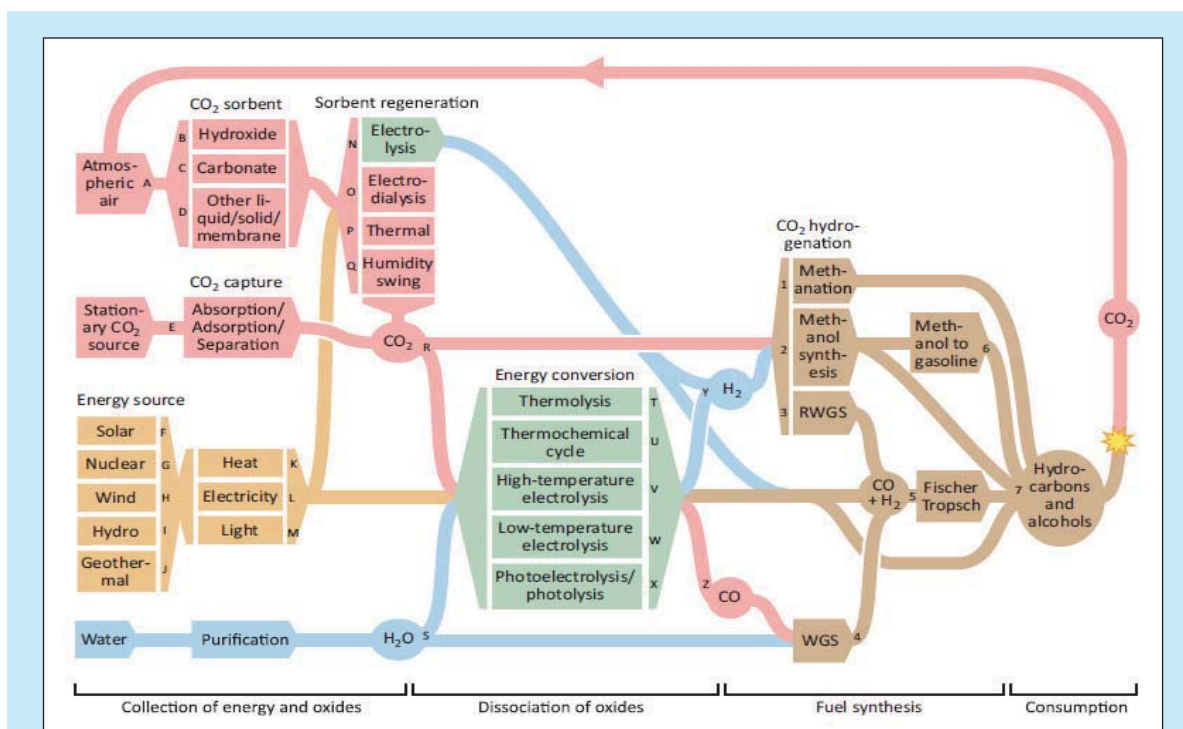
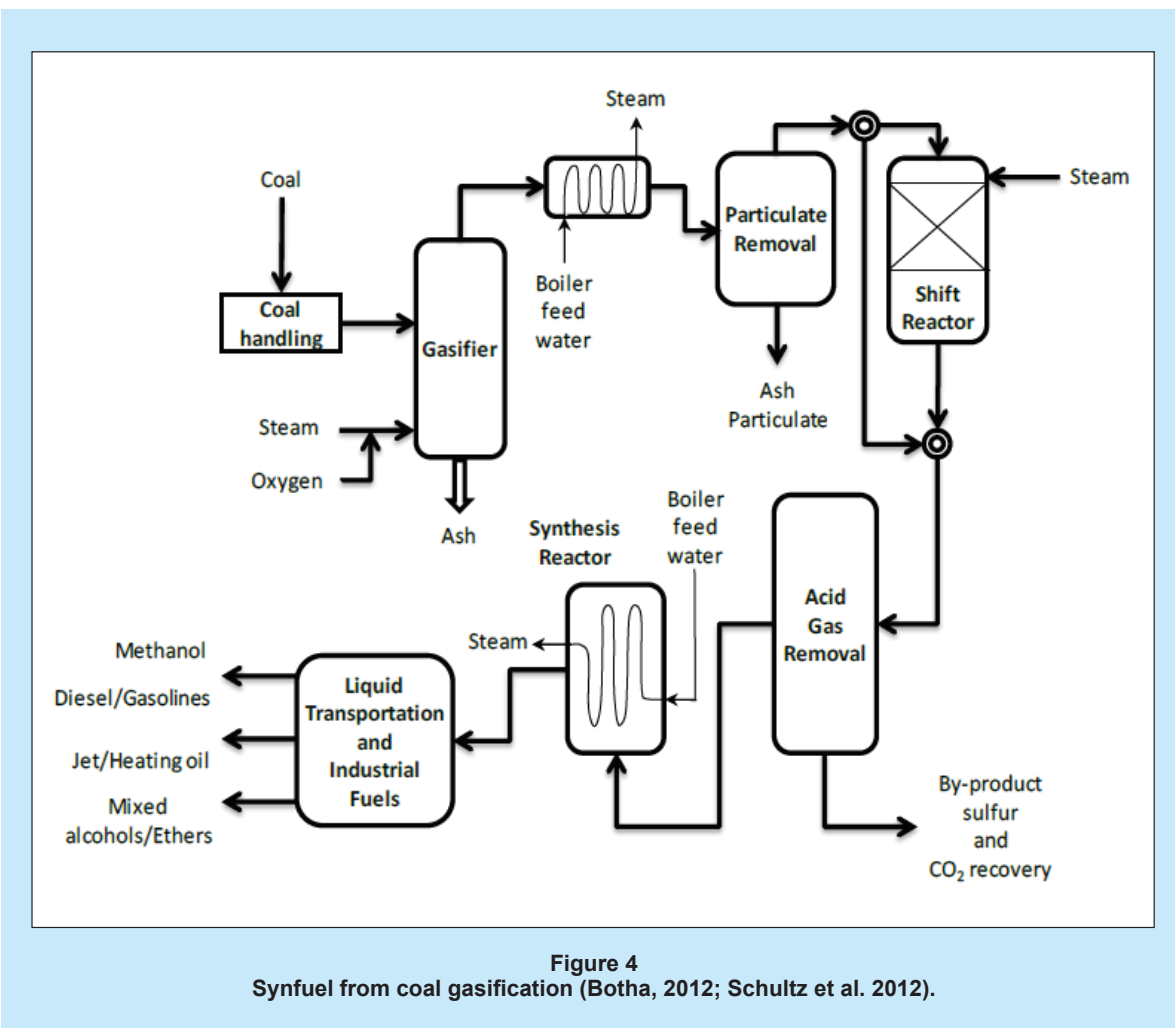


Figure 3
Map of the possible pathways from H₂O to CO₂ to hydrocarbon synfuels (Graves, 2010).



based solely on relatively abundant raw materials of H₂O and CO₂. Energy sources without no CO₂ emissions are chosen as energy sources to generate hydrogen from water. Nuclear, solar, wind, hydro, and geothermal energy are attractive options because they do not emit CO₂ (Chiuta 2010; Choi et al. 2017).

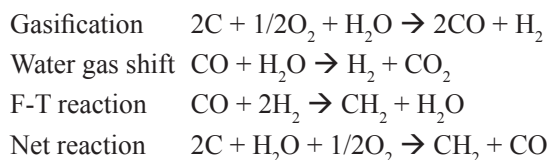
C. Synthetic Fuel Production

Liquid hydrocarbons synfuel have been synthesized for more than three-quarters of a century from non-liquid feedstocks, principally coal but also natural gas in recent years. The leading process is the F-T process which uses synthesis gas, hydrogen, and carbon monoxide, as its feed and produces a synthetic “crude” that undergoes further processing to a range of commercial finished products.

D. Conventional Process

The conventional process that commercially in operation now, is based on the process of coal

gassification to produce synthesis gas, the is mixture of H₂ and CO. This reaction than followed by water gas shift reaction to produce CO₂ and additional H₂. Main reaction of synfuel production is allowed in synfuel reactor following the F-T reaction (Chiuta, 2010; Schultz et al. 2012). The complete reactions of the process are as follow:



From stoichiometric of reaction, side product of the process is CO₂. In this conventional process, beside as a raw material, coal also use as heat energy souce by means of direct burning. The negative effect of this process production of synfuels in a coal to liquid (CTL) plant produces enormous volumes of CO₂. Figure 4 shows the diagram of conventional synfuel from coal.

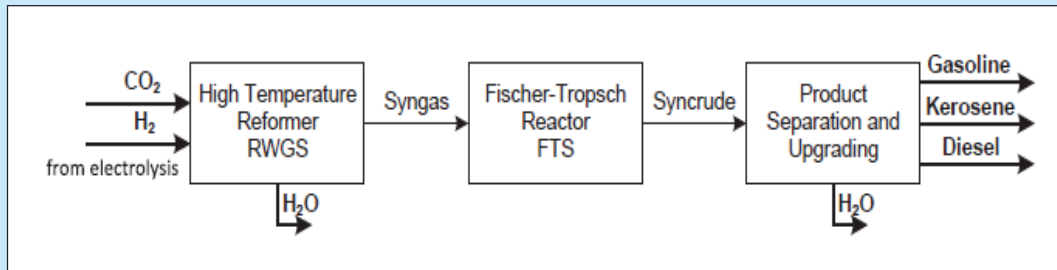


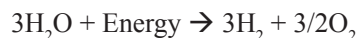
Figure 5
Diagram of the synfuel generation from CO₂ and H₂ (Konig et al. 2015).

E. Synfuel from CO₂ capture and nuclear hydrogen system

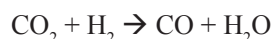
The process now under R&D is the process that utilizes CO₂ capture and nuclear hydrogen system. Nuclear hydrogen system is hydrogen production from water by utilizing high temperature nuclear reactor as a heat source. One important process is thermochemical water decomposition of iodine sulfur cycle.

As raw material in this process, CO₂ is captured from fossil fired power plants or other industrial plant. By utilizing the reverse reaction of water gas shift reaction, CO can be produced. Then CO and H₂ feed to the F-T reactor to produce synfuel. The reaction occurs in the process are as follows (Schultz et al. 2012; Chen et al. 2015).

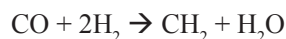
Nuclear Water-splitting



Reverse Water Gas Shift



F-T reaction



Net reaction



In this case, no coal is needed at all, and CO₂ is consumed rather than produced. The excess O₂ would be used in the fossil power plant that provides the CO₂, simplifying CO₂ capture. There is currently considerable effort underway on developing CO₂ capture systems for new and existing power plants. The increasing concern with Global Climate Change suggests that there is a reasonable likelihood of such plants operating in the timeframe associated with synthetic fuel from carbon dioxide. Such a synergistic system, dubbed twice burned coal or

recycled coal, has the potential to significantly reduce current emissions of CO₂ since the carbon in the coal is used once for power production and then again for liquid hydrocarbon fuel synthesis (Schultz et al. 2012).

The F-T reactor contains only gas and catalyst (iron-based) with no liquid phase outside the catalyst pores. The operating temperature is between 320°C and 350°C and the reactor is typically operated at a pressure of about 25 bars to yield a highly olefinic hydrocarbon product. Furthermore, the F-T reactor requires syngas with high hydrogen concentration to avoid rapid catalyst deactivation due to carbon formation (Chiuta 2010; Graves 2010; Konig et al. 2015).

The diagram of the process based on H₂ and CO₂ captured is shown in Figure 5. The raw material of CO₂ is coming from industrial or fossil power plant emission, while H₂ is produced by utilization of HTGR. Besides to produce H₂, the heat from HTGR is also can be used as heat source for the F-T, and RWGS reactors (Kreutz 2011).

III. RESULTS AND DISCUSSION

The use of nuclear energy has mostly concentrated on producing electricity. Another established but much smaller application has been as a power source for ships: submarines, icebreakers, and even merchant ships. Non-electricity applications based on direct use of heat generated in the fission process of water cooled nuclear reactors could include process heat for various low temperature industrial applications, desalination, and district heating. The creation of entirely new applications for nuclear power, such as process heat and hydrogen, is one of the main objectives in the development of Generation IV technologies (El-Genk & Tournier 2017; Locatelli et al. 2013).

Designed from the start to enable deployment in proximity of industrial sites and agglomerations, the Very High Temperature Reactor (VHTR) capability to deliver heat above 600°C to the end user makes them uniquely suited for cogeneration of process heat and electricity (Futterer et al. 2014; Yan et al. 2014). This feature enables them for instance to efficiently produce hydrogen through steam electrolysis or thermochemical processes, to supply both hydrogen and high temperature heat for producing synthetic fuels from coal, biomass or captured CO₂, or to deliver high temperature heat and hydrogen or syngas as chemical reactants to a variety of industrial plants including petrochemical, fertilizer production and steel-making. According to market studies performed in several countries, the potential market for this process heat is approximately as large as the electricity market and currently almost exclusively provided by fossil fuels with the concomitant CO₂ emissions

The efficiency of electricity production can be increased by using higher temperatures, but this also expands the range of possible applications for process heat. Industrial processes which need process heat at high temperatures include wood pulp manufacture (around 400°C), desulphurization of heavy oil in petroleum refining (around 500°C), production of syngas, styrene, ethylene and hydrogen via steam reforming, copper-chlorine (around 550°C) or iodine-sulphur thermochemical processes (around 800°C), and the manufacture of iron, cement and glass (around 1000°C) (Locatelli et al. 2013). In nuclear terms such temperatures can only be achieved by the various Generation IV reactors, which also introduce entirely new operating principles and safety features (Verfondern et al. 2017).

Nuclear hydrogen cogeneration by means of nuclear water splitting, is relatively the most developed technology of high temperature nuclear reactor application. This technology is interesting from the point of hydrogen and thermal energy output from the system. Hydrogen will be used as important feedstocks for CO₂ hydrogenation, while thermal energy from HTGR (in the form of heat, steam or electricity) is absolutely needed to run the process. As nuclear hydrogen cogeneration do not utilize carbon source as raw material, the system need CO₂ source from the other system. Fossil power plant, cement industries, or the other industry that emit CO₂ in a huge quantity can be used as CO₂ source. By utilizing technology of CCS and or CCU, CO₂ can be captured

and separated from the other gases, and use them as material of carbon source for petrochemical.

Some studies indicated the important of nuclear hydrogen cogeneration as the key technology for CO₂ conversion in the future. Study in US, indicated the role of nuclear hydrogen cogeneration in conversion CO₂ emission from cement plant to produce synthetic fuel (Bicer & Dincer, 2017). The other similar studies, also have been doing in around the world such as nuclear hydrogen cogeneration for CO₂ conversion to: ammonia and urea production (Salimy, 2014; INL-DOE, 2010), methanol (Salimy & Alimah 2015; Faberi et al. 2014), synthetic fuels (Stangeland 2017; Verfondern 2013).

Theoretically, to produce hydrocarbon synfuel from CO₂ and H₂, the ideal ratio of CO₂/H₂ is 1:3 (Konig et al. 2015; Schultz 2012). In other words hydrogen will be needed in large quantities to meet the needs of transportation fuel in the future. The application of nuclear cogeneration to hydrogen production is expected to meet that demand. If nuclear water splitting technology does not reach the commercial level, conventional nuclear-based electrolysis technology can be utilized as intermediate technology for the supply of hydrogen. The operation of the electrolysis process by nuclear power plant operators may be the solution to obtain hydrogen by electrolysis process. When the demand for electricity is high, the electrolysis unit is lowered in capacity, while when the demand for electricity is low, the hydrogen production capacity can be raised.

The efficient utilization of CO₂ has attracted considerable attention from fundamental research to industrial application in recent years (Lanjekar, 2011; Ravanchi & Sahebdehfar, 2014). The efficient utilization of (renewable) CO₂ will not only help to alleviate greenhouse effect, but also to obtain useful chemicals (Prokofyeva & Gürtler, 2014). Heterogeneous catalysis, electrocatalysis and photocatalysis are presently the three predominant chemical methods for converting CO₂ into some useful chemicals, such as methanol, formic acid and formaldehyde, etc. (Faberi et al. 2014). This CO₂-to-fuel integrating the heat from nuclear plant reactor could lead towards the sustainable development in the long term since cement industries, pulp and paper industries, as well as steel industries are the boundless sources of CO₂ (Graves 2010). Until now, CO₂ to fuel has been usually considered coupled to renewable sources such as wind power. In France 14% of electrical power is produced by renewable

energy but also around 78% is provided by nuclear reactors (IAEA 2012). Nuclear power is also a low CO₂ footprint energy source, which can be used massively for CO₂ to fuel processes (Borgard 2011).

Compared to conventional coal-based gasification processes, alternative CO₂ hydrogenation processes promise significant reductions in CO₂ emissions. From the stoichiometry of the reaction it is seen that in the conventional process, CO₂ produced in large quantities that must be removed from the system. While in the process of hydrogenation of CO₂, CO₂ is not produced but instead used as raw material. Intensive study of HTGR applications for synfuel production has been conducted in South Africa based coal gasification process. The study indicates significant potential benefits compare to the conventional process. The CO₂ emission can be decrease about 75%, and coal requirement can be reduced about 40% (Botha, 2012; Chiuta, 2010). The other study indicate, for the process based on CO₂ capture and H₂, the process will not emit CO₂ at all, and also no requirement of coal. Even this process will utilize CO₂ from the other plants (Schultz, 2012).

IV. CONCLUSION

The technology of hydrocarbon synfuel production from CO₂ and H₂ will play an important key in the future in supplying the need of liquid transportation fuel. Nuclear hydrogen will also to be important key technology to realize the process. From the point of environment, the process can mitigate CO₂ emission in very significant amount. Even this technology can clean the electricity production from coal power plant, by capture the CO₂ emission to be raw material of the process.

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