The assessment of nuclear hydrogen cogeneration system (NHCS) for CO₂ conversion to urea fertilizer

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INTRODUCTION
One way to reduce CO₂ emissions is to turn it into a product that has added value. In addition, CO₂ can be used as an abundant and renewable raw material for various petrochemical and synthetic fuels. CO₂ has advantages over other C1 toxic materials such as carbon monoxide and phosphagen [1]. A study indicates that the production of chemicals from CO₂ can significantly influence mitigation of global CO₂ emissions because the amount of CO₂ used is quite large [2]. To produce chemical products from CO₂, large amounts of hydrogen are needed. Water splitting by utilizing high-temperature gas cooled reactors (HTGR) is an interesting technology to supply hydrogen for CO₂ conversion [3, 4]. HTGR is a reactor that operate at high temperature (~1000 °C). This reactor is dedicated not only for electricity generation, but also to utilize high temperature heat energy for industries.

Water splitting by utilizing HTGR is the process of thermochemical decomposition of water to produce hydrogen and oxygen which occurs at high temperatures [5]. Heat energy coming out from HTGR can serve as heat energy source to run the process. Among the nuclear thermochemical process, a process with iodine-sulfur cycle is the most advanced. The process was originally developed by General Atomics in America in the 1970s, later adopted and developed by several countries such as Japan, Germany, China, France, and South Korea. Nuclear hydrogen cogeneration system (NHCS) is a system that has capability to supply hydrogen and high temperature thermal energy. NHCS can then play an important role in the application of nuclear energy to industrial processes.

From the environmental side, expanding the use of nuclear energy not only for electricity but also for industry, aims to reduce CO₂ emissions in the atmosphere. If NHCS can be combined with CO₂ conversion technology, it is hoped that significant CO₂ emissions savings will be obtained. One of the important uses of NHCS for CO₂ conversion is the production of urea fertilizer. Urea production is a process that takes place at high temperatures which consume large amounts of natural gas, both as raw materials, as well as source for process heat, and utilities. The study of NHCS applications and CO₂ conversion can save large amounts of natural gas which has a significant effect on reducing CO₂ emissions.

This paper reviews previous studies related to the application of the NHCS for the conversion of CO₂ to urea. The future limitation of natural gas can be a threat to the sustainability of the urea fertilizer plant if it only relies on conventional processes made from natural gas. For this reason, it is necessary to study various alternative processes for producing urea fertilizer with raw materials other than natural gas. Studies in America indicate that urea fertilizer can be made with coal or water [11]. In principle, the key technology for urea fertilizer plants is the production of hydrogen in the ammonia plant unit. Because hydrogen can be produced from coal (through coal gasification processes), or with water raw materials (thermochemical processes), it means that natural gas as a raw material for fertilizer plants can also be replaced with other materials that can produce hydrogen.

This review provides important input to the stakeholders in formulating policies of nuclear energy development in Indonesia, particularly the development of the use of high-temperature nuclear reactor in the future.

NHCS FOR CO₂ CONVERSION
Nuclear hydrogen cogeneration

Cogeneration is a way to use a single source of energy efficiently to produce power and useful thermal energy [7]. This system is a very efficient, clean, and reliable approach to generate power and thermal energy from a single fuel source [8]. Typically, cogeneration plants recover “waste heat” that is otherwise discarded from conventional
power generation. The recovered waste heat is effectively utilized in other thermal energy applications. Current nuclear power plants have low thermal efficiencies generally in the range of 30 to 35% compared to other steam cycle energy conversion systems. Countries embarking on nuclear power should consider cogeneration and the use of waste heat from a NPP to increase energy utilization and overall efficiency.

In a number of countries, cogeneration and heat production using nuclear reactors is already an effective way to meet different types of energy needs. Currently, there are 79 reactors operating in cogeneration mode and the potential for applying this technology more widely appears promising [9,10,11,12]. 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.

Utilization technology [16,17]. In CCU technology, the captured CO2 is processed using various processes to convert CO2 into usable materials, such as urea, methanol, or inorganic carbonates. The process is complex and energy-intensive, but it provides a way to sequester CO2 and reduce the amount of greenhouse gases in the atmosphere.

One of the technologies used is carbon capture and storage technology (CCS) that captures CO2 emissions from industrial processes and stores them underground or in other long-term storage sites. CCS is considered a crucial technology to mitigate climate change by reducing CO2 emissions. However, CCS technology has limitations such as high cost and low energy efficiency.

In principle, CCS technology works by capturing CO2 emissions that emanate from sources of emissions (factories or power plant), separating CO2 from other gases, then transporting and storing the CO2 to sustainable storage [13,14,15]. This technology is still relatively expensive and does not provide direct added value. Since decades, CCS technology has been developed into CCU (Carbon Capture and Utilization) technology [16,17]. In CCU technology, the captured CO2 gas will be used as industrial raw material. This way, in addition to obtaining profits in the form of decreasing the rate of emissions, additional value is also obtained from the use of CO2 emissions as raw material for industrial processes. While CO2 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 [15] and fuels [16]. Figure 4 demonstrates the potential use of CO2 as raw material for industrial processes [18]. It can be seen that urea is one of the important products that can be produced from CO2 conversion.

**Fig. 1** HTGR for hydrogen cogeneration, thermochemical IS process [11].

Figure 1 shows the ability of high temperature nuclear reactors to provide heat for hydrogen production, petrochemical processes, and iron industries, as well as clean water. High temperature thermal energy and hydrogen produced from nuclear can be a basic component to operate various processes in the petrochemical industry, NHCS by means of nuclear water splitting of iodine sulfur cycle is the most developed technology of high temperature nuclear reactor application.

**CO2 conversion**

The issue of global warming induced by CO2 emissions has become a global issue that is increasingly important in the world. It can be a major cause of climate change and the negative impact on human life. One of the indicators used in analyzing the issue of global warming is the increase of greenhouse gases, especially CO2, as a result of human activities. So far, efforts have been started to reduce the impact of global warming, such as reforestation, energy saving, the use of new and renewable energy, and utilization of engineering and technology. One of the technologies used is carbon capture and storage technology named CCS (Carbon Capture and Storage).

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**Fig. 2** Applications of CO2 Utilization [18].

**NHCS for CO2 conversion to urea**

The study of NHCS applications for the conversion of CO2 into urea has been carried out in Indonesia [19]. In this study, a conventional process of urea fertilizer plant with a capacity of 1725 tons per day was set as a reference. In general, urea is obtained when reacting ammonia with CO2 through a 2-step reaction [20].

\[
2\text{NH}_3 + \text{CO}_2 = \text{NH}_2\text{COONH}_4 \quad (1)
\]

\[
\text{NH}_2\text{COONH}_4 = \text{H}_2\text{O} + \text{NH}_2\text{CONH}_2 \quad (2)
\]

In conventional processes, ammonia is obtained from the reaction of hydrogen with nitrogen sourced from the air. Hydrogen is obtained from the process of steam reforming natural gas, which also produces CO2. Unlike conventional processes, the NHCS do not produce CO2. For this reason, CO2 needs must be supplied from outside the system. In the study, it is assumed CO2 was obtained from emissions of coal-fired power plants. The application scheme of the NHCS in the process of converting CO2 into urea can be seen in Figure 3.
CCU system is needed to accommodate CO₂ emissions from power plant. Studies in America have shown that the efficiency of CCU utilization is about 70% (which is a combination of capture, storage, and utilisation). So, to supply the CO₂ requirement of 417450 ton/year, coal power plant should be capable of emitting CO₂ at 596357 ton/year (417450 / 0.7 = 596357). With 1.09 kg/kWh emission factor, 70% capacity factor [21,22], and conversion factor of 8760 hr/year, the power capacity of the power plant can be calculated as follows: [596357 / (1.09 x 0.7 x 8760)] = 89,223 ≈ 90 MWe. In other words, if the urea fertilizer plant is coupled with an NHCS established near the coal power plant of 90 MWe, the fertilizer plant will be able to absorb all CO₂ emissions from this power plant.

In this study, NHCS was powered by HTGR with a capacity of 2x600 MWe. Nuclear energy balance is shown in Table 1. This table shows that HTGR with a capacity of 2x 600 MWt can be used for the production of urea fertilizer with a capacity of 1725 ton/day. In the process, there is still 425.65 MWt of heat energy remaining which can be converted into electricity of 140 MWe.

### Table 1 Nuclear Energy Balance [19].

| Energy Supply | Conventional | NHCS for CO₂ conversion [19] | Nuclear steam reforming [23] |
|---------------|--------------|-----------------------------|-----------------------------|
| Power Capacity | 2x600 MWe   | Natural gas, air, water     | Natural gas, air, water     |
| Recuperator Eff. | 90%        | Nuclear steam reforming of natural gas to produce H₂ and CO₂ | Nuclear steam reforming of natural gas to produce H₂ and CO₂ |
| Output from recuperator | 1080 MWe | Ammonia production by react H₂ with N₂ from air. | Ammonia production by react H₂ with N₂ from air. |
| Steam Gen. Eff. | 90%        | CO₂ + NH₃ → urea | CO₂ + NH₃ → urea |
| Process heat for ammonia plant | 505.36 MWt | CO₂ come from side product of steam reforming | CO₂ come from side product of steam reforming |
| Steam for process and electricity | 149 MWt | Supply the need of process heat, process steam, and electricity by direct burning of natural gas | Supply the need of process heat, process steam, and electricity are com from HTGR. |
| Total Demand | 654.36 MWt | Supply the need of process heat, process steam, and electricity by direct burning of natural gas | Supply the need of process heat, process steam, and electricity are com from HTGR. |
| Remain | 425.65 MWt ~140.46 MWe | Nuclear steam reforming of natural gas to produce H₂ and CO₂ | Nuclear steam reforming of natural gas to produce H₂ and CO₂ |
| NHCS for CO₂ conversion | | Nuclear water splitting of iodine sulphur process to produce H₂ | Nuclear water splitting of iodine sulphur process to produce H₂ |
| Ammonia production by react H₂ with N₂ from air. | CO₂ come from coal power plant | Ammonia production by react H₂ with N₂ from air. | Ammonia production by react H₂ with N₂ from air. |
| Urea plant | | CO₂ + NH₃ → urea | CO₂ + NH₃ → urea |
| | | CO₂ come from side product of steam reforming | CO₂ come from side product of steam reforming |
| Utility | | Supply the need of process heat, process steam, and electricity by direct burning of natural gas | Supply the need of process heat, process steam, and electricity by direct burning of natural gas |
| Annual consumption of natural gas | 21,252,188.82 MMBTU | 8,945,131.08 MMBTU | 0 |
| (as raw material, process heat for steam reforming, direct burning for electricity and process steam) | (as raw material) | (as raw material) | (as raw material) |
| Annual saving of CO₂ emissions | 718,192.42 ton | 0 | (1.24x10⁶ + 417,450) ton |

### DISCUSSION

In addition to the study of NHCS applications for the conversion of CO₂ into urea, study of HTGR for the process of steam reforming of natural gas to replace the conventional process of steam reforming of natural gas, has also carried out. In this process, natural gas as a raw material for the process is still needed, but the usage of natural gas has been replaced with HTGR which supplies heat, steam, and electricity. As comparison, summary of study results can be seen in Table 2.

In Table 2, it can be seen that the conventional process requires natural gas of 21.25 million ton/year which is used as raw material (8.95 million tons), process heat for steam reforming (6.16 million tons), and utilities (6.15 million tons). The replacement of the conventional to nuclear steam reforming process, only requires natural gas as a raw material of 8.95 million ton/year, will save natural gas by 12.3 million ton/year which can be replaced by nuclear heat. In terms of the potential reduction in the rate of CO₂ emissions, urea production with a capacity of 1725 ton/day requires natural gas of around 21.25 MMBTU/ton/year which is equivalent to CO₂ emissions to the environment of around 1.24 million ton/year. The process with nuclear steam reforming, which replaces nuclear heat for the steam reforming processes and utility, can reduce the rate of CO₂ emissions to around 718.2 thousand ton/year. Besides able to reduce the rate of emissions up to 1.24 million tons a year, the application of the NHC system is also able to absorb CO₂ emissions from coal power plants amounting to 417.5 thousand ton/year.

From the point of urea fertilizer demand, urea fertilizers are one of the major inputs, which help in increasing the yield of the crops. Fertilizers consumption rate in Indonesia has been increased over the last decade. Growing population and limited agricultural land have accelerated demand for agricultural land and proper utilization to increase crop yields. In terms of raw materials, fertilizer industry is an industry that absorbs the largest number of natural gas consumption after the need for electrical energy. BPPT Energy Outlook data states that Indonesia will soon become the net importer of natural gas [24]. Hence, it is necessary to conduct various studies to find sustainable alternatives to reduce the dependency on natural gas. The use of nuclear energy is expected to reduce the consumption of natural gas in significant amounts which has an impact on reducing the rate of CO₂ emissions to the environment.

From the point of CO₂ conversion, recovery and utilization of CO₂ is a well-known technology since the second half of 1800’s [15]. The efficient utilization of CO₂ has attracted considerable attention from fundamental research to industrial application in recent years [25,26]. The efficient utilization of renewable CO₂ will not only help to alleviate greenhouse effect, but also to obtain useful chemicals [1].
As previously stated, NHCS by means of nuclear water splitting of iodide sulfur cycle is 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. As nuclear hydrogen cogeneration does not utilize carbon source as raw material, the system needs CO₂ source from the other system. Fossil power plant, cement industries, or the other industry that emit CO₂ in a combination technology of NHCS and CO₂ conversion, are expected to use them as a carbon source for petrochemical. The combination technology of NHCS and CO₂ conversion, are expected to become the key technology to support sustainable development.

Some studies have highlighted the importance of NHCS as the key technology for CO₂ conversion in the future. A study in the US has indicated the role of nuclear hydrogen cogeneration in conversion CO₂ emission from cement plant to produce synthetic fuel [27]. Other similar studies also have been performed using nuclear hydrogen cogeneration to convert CO₂ into new products such as ammonia and urea [6, 19], methanol [28, 29], and synthetic fuels [3, 9, 27, 30, 31].

Indonesia is currently developing an experimental power reactor design that is expected to operate co-generatively to generate both electricity and heat for industrial applications [32, 33]. The continuous study on the application high temperature nuclear energy for industry must be carried out in anticipation of possible applications in Indonesia in the future.

CONCLUDING REMARK

From the analysis, it can be understood that the NHCS combined with the technology of CO₂ conversion will play the important role in the future of chemical industries. Adopting this technology will lead to the reduction in dependency on fossil fuel which ultimately will reduce the CO₂ emission to the atmosphere.

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