Utilization of HTGR for phosphate fertilizer production and uranium recovery

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Abstract. A study of the utilization of HTGR for phosphate fertilizer production and uranium has been carried out. The objective of the study is to understand the high temperature nuclear reactor applications to explore and process phosphate rocks into phosphate fertilizers with additional uranium products that can be used for the reactor fuel itself. The method used in this study is literatures studies, and analysis of previous research. The mining and processing of phosphate rock mines utilizing HTGR will benefit from two sides. First, phosphate fertilizer products are useful to improve agricultural products. Second, the uranium by-product of phosphate rocks is often referred to as unconventional uranium resources. Studies conducted abroad indicate that HTR 3x50MW is able to provide energy to be integrated to run the production process of phosphate fertilizers, and uranium. Factory units coupled with cement and sulfuric acid indicate the need for fossil fuels. In the study, an energy balance analysis was performed on the equivalent electrical energy required. With electricity production of 407 GWh/year from HTR 3x50 MW and 72 GWh/year of cogeneration of sulfuric acid plant, it can be used to run fertilizer, uranium and cement production units. Excess energy of 98 GWh/year can be connected to the power grid. In the paper also discussed if the hydrogen nuclear system can be realized, this system can be the backbone of the chemical industry because it can provide electrical energy, hydrogen, and heat energy simultaneously. With such models it is estimated to be able to save the use of fossil fuels that have an impact on emissions reduction with a very significant.

1. Introduction
Energy is an intangible currency essential for sustaining the life. More than one billion people are expected to be added in coming 15 years, making world population about 8.5 billion in 2030 [1]. Energy consumption is anticipated to increase by 34 percent till 2035. To support the high demand for energy, NPPs must be considered as important energy suppliers.

If the first generation nuclear power plant is fully dedicated to producing electricity, the 4th generation nuclear reactor is designed as a safe, economical, environmentally friendly, and sustainable nuclear energy supply system and can supply energy for various needs, both for electricity generation and industrial process heat energy including hydrogen production, coal liquefaction and gasification, refinery operations, mining and mineral processing. Adopting the concept of nuclear reactor 4th generation, currently Indonesia is developing Reaktor Daya Eksprimen (RDE) technology based on HTGR type PBR [1]. HTGR is a High Temperature Gas-cooled nuclear Reactor operating at high temperatures. The high temperature output brought by helium gas as reactor coolant can be utilized for various industrial applications. One interesting application to study is the utilization of nuclear energy
HTGR for phosphate stone development process to be converted into phosphate fertilizer, with additional products of uranium contained in phosphate rocks.

HTGRs are thermal reactors that use graphite as a moderator and helium as an inert coolant. The greenhouse gas lean energy source can provide process heat as high as 1000°C and electricity for cogeneration (combined electricity and process heat generation) while retaining desirable safety characteristics when built in a modular design [2, 3]. In the case of HTGRs the potential to participate in the larger energy market (not only electricity) has been recognized for a long time with concept designs like the German PR-500 [4] made in 1973 for steam-reforming, etc. Many other projects, like the experimental High Temperature Engineering Test Reactor (HTTR) in Japan, support hydrogen production research [4]. Beside HTTR, Japan also developed HTGR type reactors for multiple heat applications, included in this study are: the High Temperature Reactor 50S (HTR50S) [5] and the Gas Turbine High Temperature Reactor 300-Cogeneration (GTHTR300C). In Indonesia, National Nuclear Energy Agency of Indonesia (BATAN) also developed the design of HTGR type named Indonesia RDE. At this time, the analysis of detail engineering design has been ongoing.

In this paper, HTGR applications for the mining process of phosphate stone will be discussed. From the process will obtained phosphate fertilizer, and the side product of uranium. Phosphate fertilizer is needed to support the intensification of agriculture, given the increasing population and the narrowing of agricultural land. Meanwhile, the uranium present in phosphate rock, which ranges from 50-200 ppm [2], should be utilized to support the long-term availability of uranium. The International Atomic Energy Agency (IAEA) is promoting the idea to use High Temperature Reactors (HTRs) to provide process heat and/or electricity for mineral development processes whilst recovering and using the accompanying uranium/thorium as nuclear reactor fuel for the HTR employed and/or other Nuclear Power Plants (NPPs) [6].

The objective of the study is to understand the high temperature nuclear reactor applications to explore, and process phosphate rocks into phosphate fertilizers with additional uranium products that can be used for the reactor fuel itself. The method used in this study is literature studies, and analysis of previous researches. The results obtained are expected to support the development policy of nuclear energy program in Indonesia, especially the development of experimental power reactor which is currently entering the stage of detail engineering design.

2. HTGR for phosphate rock conversion

Schematic diagram of HTGR utilisation for phosphate rock conversion is shown in Figure 1. HTGR takes a role as an energy source for process. The energy output brought by helium gas coolant in the temperature range (700-1000°C) can be utilized as heat energy. For the needs of high temperature, heat energy is supplied in the form of heat processes with helium gas media. As for low and intermediate temperatures, hot helium can be converted into steam processes. Some process equipment can also be operated with electricity generated by HTGR [2].

![Figure 1. Schematic diagram of HTGR utilisation for phosphate rock conversion [2]](image-url)
From the picture, it could be seen that processing phosphate rocks produce phosphate fertilizers and uranium. Uranium is further processed into HTGR fuel. With this system, HTGR can be utilized for a process (fertilizer production), with the need for HTGR fuel supplied from the byproducts of the process.

2.1 Conversion phosphate to fertilizer
One type of phosphate fertilizer that is widely produced is diamonium phosphate (DAP). DAP production process starts from the formation of phosphate acid. In the wet phosphoric acid production process, the phosphate concentrate slurry is dissolve in a strong acid, usually sulphuric acid $\text{H}_2\text{SO}_4$, producing phosphoric acid $\text{H}_3\text{PO}_4$. Sulfur from the sulphuric acid combining with the calcium phosphate concentrate to produce phosphogypsum sulphate as a waste product $[7]$. 

$$3\text{H}_2\text{SO}_4 + \text{Ca}_3(\text{PO}_4)_2 + 6\text{H}_2\text{O} \rightarrow 2\text{H}_3\text{PO}_4 + \text{CaSO}_4 + 6\text{H}_2\text{O}$$

Then, the phosphoric phosphate acid is treated with ammonia to produce the diammonium phosphate or DAP as final product.

$$2\text{NH}_3 + \text{H}_3\text{PO}_4 \rightarrow (\text{NH}_4)_2\text{HPO}_4$$

Since 1960s, when ammonium phosphate fertilizers became available, DAP quickly became the main phosphate based fertilizer. It is produced by combining phosphoric acid with ammonia, in such a way that different manufacturing routes are related to different sources of such materials. Figure 3 presents different pathways for DAP production $[8]$. 

![Figure 2. Block diagram of DPA production by 3 pathways $[8]$](image)

In general, the unit process in DAP plants are as follow: Phosphoric acid plant, ammonium sulfate plant, granulation plant, sulphuric acid plant. Since the plant is combine with the unit of uranium recovery, in this facility also have a uranium recovery unit.

2.2 Phosphate as uranium resources
The IAEA states that uranium in phosphate rocks is an unconventional source of uranium, as is the presence of uranium in seawater. However, the presence of uranium in phosphate rocks is thought to account for about 92% of total unconventional U resources. In general, the content of uranium in
phosphate rocks varies depending on the location and history of the formation of the rock. Table 1 indicated the uranium content in phosphate rocks for different locations [2]. By processing phosphate rock into phosphate fertilizer, uranium byproducts can be obtained that can be used as nuclear fuel. This will mean increased levels of nuclear fuel availability in anticipation of growing fuel demand.

![Table 1. Uranium in phosphate rock by country [2]](image)

Although predictions about the overall amount of uranium in phosphate rock are challenging as countries often do not report or mention unconventional resources in their records [9] it is estimated that the world total amount of available recoverable uranium from phosphate rock reserves is roughly 5,700,000 tons [10] compared to 7,096,600 tons reasonably assured conventional resources [9].

### 2.3 Phosphate fertilizer production by utilizing HTGR

The study of HTGR application for phosphate rock conversion process to phosphate fertilizer has been done in several countries [2, 6, 11,12, 13, 14]. In principle, the calculation of energy requirements is done by converting nuclear energies into electrical and heat energy to run the process. A study showed that a Japanese design HTR with a capacity of 50 MW of 3 units is sufficient to run the operation of plant units to convert phosphate rock into phosphate fertilizer, including picking up uranium contained in phosphate rock as the fuel stock of the HTR nuclear fuel [4]. Figure 3 present the schematic diagram of HTGR coupling with phosphate rock conversion, uranium recovery, and cement plant. While the energy balance of the process is presented at Table 2.

As seen at the Figure 3, HTRs with a capacity of 3x50 MW can be utilized to meet the energy requirements of the process of making phosphate fertilizer and uranium collection. In the study, the plant is also integrated with a cement plant to utilize the sulphur from sulfuret recovery plant. In cement plant, almost the process heat is still generating by fossil fuel, that can not yet fully be replaced with nuclear energy. In sulphuric acid plant, the excess heat energy production from exothermal reaction of sulphuric acid formation, can be utilize to generate energy by cogeneration system.
Table 2. Energy balance of HTR utilisation for phosphoric rock processing [14]

| Component / Process Unit                      | Electricity consumption, GWh/year |
|----------------------------------------------|----------------------------------|
| Sulfuric recovery plant                      | 217                              |
| Cement plant                                |                                  |
| Phosphoric acid plant                        | 144                              |
| Ammonium sulfate plant                       |                                  |
| Granulation plant                            |                                  |
| Sulfuric acid plant (cogeneration system)    | -72                              |
| Uranium recovery                             | 20                               |
| 3x HTR 50MW                                  | -407                             |
| Sum, connected to the grid                   | -98                              |

The Table 2 shows that the energy source of 3x50 MW HTR nuclear equivalent to 407 GWh/year energy, plus energy from the cogeneration plant utilizing the sulfuric acid plant’s heat energy of 72 GWh/year, total energy of 479 GWh/yr is obtained to operate the process. The energy is used to run the sulfuric recovery plant and cement plant (217 GWh/yr), phosphoric acid plant, ammonium sulfate plant, granulation plant (144 GWh/yr), and process of uranium recovery (20 GWh/yr). There is still an excess of 98 GWh/yr of energy that can be connected to the power grid.

3. Discussion

HTRG utilization for phosphate rocks processing aims at addition to generating useful phosphate fertilizer to support agricultural intensification, as well as to benefit unconventional uranium resources contained in the phosphate rocks to support long-term uranium availability.

In terms of production processes utilizing 3x50 MW HTR, energy requirements are supplied in the form of converted electrical energy to drive low-temperature operating units. High-temperature operating units such as cement plants and sulfuric acid factories are still run with fossilized materials.
to produce high-temperature heat energy [5,14]. If hydrogen production processes with high temperature heat energy such as natural gas steam reforming processes and / or thermochemical processes can be applied, then operating units such as sulfuric acid plants, ammonia plants may be operated with nuclear energy heat sources. This means it can significantly reduce the use of fossil fuels.

If nuclear hydrogen cogeneration based on thermochemical and / or steam reforming processes can be realized, nuclear hydrogen cogeneration can be the backbone of the petrochemical industry. In the fertilizer industry, for example, the cogeneration system can convert hydrogen and nuclear heat into ammonia, sulfuric acid which can be converted into urea or phosphate fertilizers [4, 5,15, 16]. The system can also be applied to reduce CO₂ emissions by converting CO₂ into high value-added products [17, 18, 19, 20].

In terms of uranium recovery, the process is also profitable. Phosphate rock has been known as unconventional uranium resources. A number of studies have shown a considerable amount of uranium that can be obtained from the collection of uranium present in phosphate rock [21, 22, 23, 24]. Indonesia is also a country rich in natural resources. The HTGR application for mineral processing also needs to be considered, in addition to obtaining the main mineral yields, it is also important because the minerals contain nuclear fuel. The concept of energy neutral mineral processing based on NPP is attractive to be adopted in Indonesia. This concept in principle utilizes high-temperature nuclear energy to process minerals (gold, copper, phosphate) until the final product is obtained, plus the additional amount of nuclear fuel contained in the mine [6].

Indonesia is a country that has a nuclear program. In addition to continue conducting conventional nuclear power plant implementation studies to meet the surge in electricity demand, Indonesia is also continuing to conduct study on high temperature reactor that can be used not only for electricity but for various industrial thermal energy needs. Currently BATAN continues to finalize detailed engineering design of experimental power plant (RDE) based on high temperature nuclear technology. In addition to detailed design related to nuclear island, the study related to RDE utilization need to be continued in the future. As an agriculture-based country, the HTGR application for fertilizer production seems feasible for further study [25]. Population growth and reduction of agricultural land area, will encourage more intensive farming patterns to increase productivity. Therefore, various studies of HTGR applications for fertilizer production need to be continued.

As an agricultural country, with a high level of fertilizer consumption, Indonesia still imports fertilizer from outside [27], especially phosphate fertilizer. Phosphate fertilizer demand in Indonesia is currently filled with imports from other countries such as China, Thailand, Taiwan, Germany and the United States. Whereas in terms of raw materials, Indonesia has mineral reserves of phosphate stones that can be utilized as raw materials [26, 27]. The uranium content in phosphate rock is also worthy of scrutiny as a future nuclear fuel reserve in the future when Indonesia has operated nuclear power plants.

4. Concluding Remark
Obtained an understanding of the importance of HTGR utilization for processing phosphate rocks to be phosphate fertilizer. This process will play an important key in the future. In addition to meeting the needs of phosphate fertilizers to improve agricultural products, the process also produces uranium byproducts that can be used as fuel for HTGR. Uranium in phosphate rocks that are large enough, need to be taken into account to maintain the long-term sustainability of nuclear energy.

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