Investigation on neutronic properties of ZrC coated advanced TRISO fuel for high-temperature gas-cooled reactors

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Abstract. An experimental 10 MWt power reactor based on high-temperature gas-cooled reactor technology has been planned for Indonesia. The concept was initiated at the end of 2014, and the basic engineering design was completed in 2017. Currently, the development program is focused on detailed engineering design. The type of fuel aimed in the design is a pebble-bed type that contains fuel kernels made of uranium dioxide (UO\(_2\)) coated in four layers of three isotropic materials. Those layers are a porous buffer layer made of carbon, usually followed by a dense inner layer of pyrolytic carbon (PyC), followed by a ceramic layer of silicon carbide (SiC) to retain fission products, and after that by a dense outer layer of PyC. One of the issues of the fuel system is the problem of corrosion caused by the interaction of fission products such as silver (Ag) and palladium (Pd) with the SiC layer. One of the candidates to resolve the issue is to replace SiC with zirconium carbide (ZrC) which is more resistant to corrosion at high temperatures. In this study, we investigate the effects of the replacement on the neutronic properties of the reactor design at different operating temperatures. For the purpose, we use the SRAC (Standard Reactor Analysis Code) system to calculate the energy spectrum and multiplication factors of the advanced TRISO fuel design. The result of the investigation showed that, in terms of the Doppler coefficient of reactivity, the use of ZrC seems to act more favorably than that of SiC. However, ZrC increases the parasitic neutron capture in the fuel system resulting in lower initial core reactivity which was also confirmed by a slight hardening of the neutron spectrum.

Keywords: HTGR, SiC, ZrC, SRAC, neutronic properties.

1. Introduction

High-temperature reactors are increasingly gaining attention today. Its safety characteristics are inherently safe and can produce energy economically, attracting the interest of various countries in the world to research it, especially in Asia. Today, Indonesia is one of many countries in this regions that has the highest interest in the development of HTGR (High-Temperature Gas-cooled Reactor), with its plan to build a 10 MWth experimental power reactor, including its fuel, called Experimental Power...
Reactor (Reaktor Daya Eksperimental, RDE) which is expected to give an electrical output of 3 MWe [1].

The general purpose of RDE’s development is to verify and demonstrate the inherent safety features and demonstrate the technical characteristics of HTGR in generating electricity safely. The RDE is also to be dedicated to establishing an experimental base for the development of applications for nuclear process-heat for industrial applications. The specific purposes of development of RDE are, among others: to demonstrate the safety in operating a small nuclear power plant; to show our participation in R&D of new and renewable energies; to improve our NPP technology know-how and know-why in the design, construction, operation and maintenance; and to acquire knowledge in project management of NPP construction. Besides, the development is in line with current HTGR development around the world [2].

The fuel for the RDE is of pebble shape, or a spherical fuel element that utilizes the TRistructural-ISOtropic (TRISO) coated fuel particle (CFP) design. A uranium kernel made up of a solid solution of UO$_2$ is located in the innermost of the fuel element. The TRISO coating is deposited onto the kernels using a chemical vapor deposition process. The coating consists of four layers, namely a porous carbon layer buffer, an inner isotropic, dense pyrocarbon (IPyC), a silicon carbide (SiC), and an outer isotropic, dense pyrocarbon (OPyC). TRISO particles are overcoated with a graphite matrix material and pressed into a solid spherical shape. The graphite matrix surrounding the TRISO particles moderates neutrons passing through it as shown in Figure 1. The aspherical layer of the graphite matrix is pressed on the outside of the fuel core. This layer is called the fuel-free zone and acts as a barrier to protect the fuel core. The fuel design is similar to an IAEA Benchmark problem for HTGR [3].

The fuel pebble form is a 60 mm diameter sphere with a homogenous distribution of TRISO particles in a graphite matrix in the center, surrounded by a layer of graphite without fuel. The presence of many protective layers around the uranium kernel the fission products will be safely contained in the CFP. Furthermore, in the event of loss of coolant, the reactor temperature will coast down due to the negative temperature effect of the fuel system. HTGR based RDE is expected to be one of the safest reactors, while also having high thermal efficiency, and proliferation resistance characteristics [1].

During the course of its utilization, the behavior of TRISO-CFPs has been tested and investigated by many researchers through experiments and reactor operation experience [4], [5], [6]. The data show in general excellent performance of the TRISO-CFPs [7], although this fuel type is continuously being developed [8], [9]. Its use has also been considered in many types of reactors [10], [11]. However, the crystalline material comprising the SiC layer of the CFP has a tendency to decompose at higher temperatures. SiC is also prone to corrosion by metal fission products, such as palladium.

Furthermore, for the upgrading of HTGR technologies, an advanced and extended burnup
TRISO-coated fuel particle ZrC-coated fuel particle in order to keep the integrity at higher operating temperatures has been suggested. Significant improvement can be achieved by replacing SiC with ZrC because ZrC is one of the transition metal carbides which has good compatibility with structural metals. It is also recognized as having a high melting point as well as good thermodynamic stability. In addition, ZrC has other desirable properties such as good resistance to fission product palladium corrosion and adequate retention capability of other fission products. Hence, interests in substituting SiC with ZrC in TRISO fuel grow [12],[13].

In this study, we examined the neutronic properties of the RDE when the layer of SiC was replaced by ZrC of the CFP using different enrichment of uranium. The work is intended to examine the effects on the neutronics of the reactor, such as on the core reactivity values and the neutron energy spectrum.

2. Methodology

The RDE reactor core observed using graphite moderator pebbles and helium gas coolant with an outlet temperature of 700 °C and a thermal power output of 10 MWt. RDE is based on HTGR type. In case of loss of coolant accident, the reactor does not require active core cooling, the reactor will shut itself down safely and dissipate its heat by the mechanism of passive heat transfer to the surrounding atmosphere. The basic design data of the RDE is shown in Table 1 [14].

### Table 1. RDE Basic Design Data of typical small HTGR.

| Reactor type             | High-Temperature Gas-cooled Reactor (HTGR) |
|--------------------------|--------------------------------------------|
| Thermal Power            | 10 MWt                                      |
| Outlet Coolant Temp.     | 700 °C                                      |
| Inlet Coolant Temp.      | 250 °C                                      |
| Fuel                     | Low enriched uranium                        |
| Fuel element type        | Pebble                                     |
| Pressure vessel          | Steel                                      |
| Electrical output        | Approx. 3 MWe                               |

Fuel balls with a diameter of 6 cm with TRISO coated particles fill the core. The spherical fuel design can be seen in Figure 1. The pneumatic fuel handling system is used continuously to fill and remove fuel elements. Graphite is used as the main material in the core structure consisting of the top, bottom and side reflectors. The structure of the ceramic terrace is confined by steel pressure vessels. In this modeling, we used UO2 kernel with an enrichment 17% $^{235}$U and the fuel to moderator ratio in the core of 57/43 similar to the design value used in the IAEA technical document for Evaluation of High-Temperature Gas-Cooled Reactor Performance [14].

Calculations for evaluating the RDE core neutronics were carried out using the SRAC-2006 diffusion code system. This program package has been successfully installed on a PC based on Unix Bash shell on Windows operating system. The computer code system used consists of CELL and CITATION modules. The nuclear data used is JENDL3.2 from Japan [15]. This code is quite versatile and widely used for core neutronics of many types of reactors [16].

In SRAC Code calculation, the neutron equilibrium equation in the reactor core can be written as,

\[
\frac{1}{v_g} \frac{\partial \phi_g}{\partial t} = \nabla D_g \nabla \phi_g - \sum_{sg} S_g - \sum_{g'} \Sigma_{g'g} \phi_{g'} + \sum_{g'=1}^G \sum_{g'g} \phi_{g'}
\]  \hspace{1cm} (1)

Where $S_g$ is the neutron source term which can be written as
\[ S_g = c_g \sum V_g \sum \phi_g \]  
(2)

Those equations in a steady-state condition can be formulated as

\[ Af = \frac{1}{k_e} Ff \]  
(3)

Where, A is a transport, scattering and leakage operator, while F is a fission neutron source and its distribution. The variable f is a flux vector, and \( k_e \) is the effective multiplication factor. The solution to Eq. (3) is usually written as

\[ f = \frac{1}{k_e} A^{-1} Ff \]  
(4)

Equation (4) involves solving a usually large inverse-matrix of A.

Calculations of the average group constants for fuel mixture, ball moderator and reflector were carried out in accordance with the volume ratio. For moderator cells, the group constants were calculated using a model similar to that of the fuel mixture. The moderator ball consists of graphite whose radius is the same as a fuel element ball. The constants for the moderator ball are needed for cone-shaped areas at the bottom of the core, where for the case of the first core of the RDE, this area is filled with only moderator balls. Data for the fuel and moderator pebbles used in this preliminary calculation were taken from that of HTR-10 shown in Table 2.

The volume of cell units in the moderator ball is equal to \( Vp/f \), where, \( Vp \) is the volume of the ball, and \( f \) is the filling fraction. The volume of empty space associated with a ball of any type is \( = Vp(1-f)/f \). Whereas to get the constants for reflectors and other structural materials, appropriate cell modeling is needed. For this reason, cell modeling can be used similar to that of a moderator ball. In this case, the value of \( f \) is chosen equal to 1.0. And the equivalent radius of the reflector cell is the same as that of the ball, which is 3.0 cm. The modeling of the fuel cell for the spherical type of fuel was done in a similar fashion with J. Susilo et al [17].

| Table 2. Main physics parameters of reactor fuel and moderator balls used in this study. |
|---------------------------------|------------------|
| **Main Parameter**             | **Quantity**     |
| **Fuel Element**               |                  |
| Diameter of ball               | 6.0 cm           |
| Diameter of fueled zone        | 5.0 cm           |
| Density of graphite in fueled zone and outer shell | 1.84 g/cc |
| Heavy metal (uranium) loading per ball | 5.0 g |
| Enrichment of \(^{235}\)U | 18.75\% |
| Natural boron impurities in graphite | 0.125 ppm |
| Volumetric filling fraction of balls in core (f) | 0.61 |
| Radius of fuel kernel           | 0.025 cm         |
| \( \text{UO}_2 \) density in kernel | 10.4 g/cc |
| Coating layer material (starting from kernel) | PyC/PyC/SiC or ZrC/PyC |
| Coating layer thickness (cm)    | 0.009/0.004/0.0035/0.004 |
| Coating layer density (g/cm\(^3\)) | 1.1/1.9/3.18 or 6.73/1.9 |
| **Moderator Balls**            |                  |
| Diameter of pebble ball         | 6.0 cm           |
| Density of graphite             | 1.84 g/cm\(^3\) |
Using the main core parameters shown in Table 2, the multi-group constants for this reactor can be calculated using the CELL module of SRAC-2006 with the selected nuclear data library JENDL3.2. Nuclear constants are then generated using 107 energy groups, comprising of 61 fast groups and 46 thermal groups. Multi-group nuclear data generated then condensed into 6 groups, each with three fast groups and three thermal groups. Calculational flowchart as a whole is shown in Figure 2 [18].

![Figure 2. Calculation flow for the investigation of neutronic properties.](image)

**3. Results and Discussion**

The Indonesian HTGR core modeling was done by using the shape of a cylindrical shape of a reactor. The fuel mixture loaded into the core was approached assuming a homogeneous mixture between fuel elements and graphite moderator balls. The calculation of the reactor multiplication factor (eigenvalue problem) was done using the CITATION diffusion calculation module that analyzed the reactor core in two dimensions R-Z geometry. It is expected that the use of ZrC layers in CFP will not affect the neutron flux distribution in the core, because the size of the fuel pebbles and Uranium enrichments are assumed the same. The effect perhaps exists only on the initial criticality core height.
The energy structures used in nuclear data we used comprise of 107 groups. However, in order to speed up the calculation, we merged them into 6 groups, each with 3 groups in fast and thermal regions. Table 3 shows the energy group structure we used.

Table 3. The six energy groups structure used in the calculation.

| Group | Energy range (eV) | Lethargy range |
|-------|------------------|----------------|
| 1     | 0.10000E+08      | 0.18316E+06 0 4 |
| 2     | 0.18316E+06      | 0.96112E+03 4 9.25 |
| 3     | 0.96112E+03      | 0.18554E+01 9.25 15.5 |
| 4     | 0.18554E+01      | 0.68256E+00 15.5 16.5 |
| 5     | 0.68256E+00      | 0.10963E+00 16.5 18.33 |
| 6     | 0.10963E+00      | 0.10000E-04 18.33 27.63 |

The results of our SRAC calculation for the multiplication factor of the RDE core at different fuel temperature with different enrichment of uranium using SiC and ZrC coated fuel is shown in Table 4. We notice that as fuel temperature rises, the multiplication factor will also increase in all types of fuel and all enrichment. This shows the inherent safety characteristics of the core.

Table 4. Results of SRAC calculation on multiplication factor at different temperatures and enrichments for SiC and ZrC.

| Temperature (K) | SiC fuel-enrichment | Multiplication factor | ZrC fuel-enrichment |
|----------------|---------------------|-----------------------|---------------------|
|                | 18.75%   15%   11.25% |                      | 18.75%   15%   11.25% |
| 300            | 1.3264    1.2685  1.1838 | 1.1189    1.0661  0.9908 |
| 600            | 1.2935    1.2352  1.1506 | 1.0861    1.0332  0.9576 |
| 900            | 1.2694    1.2106  1.1260 | 1.0625    1.0093  0.9339 |
| 1200           | 1.2484    1.1891  1.1044 | 1.0400    0.9867  0.9162 |
| 1500           | 1.2313    1.1715  1.0865 | 1.0221    0.9530  0.8779 |
| 1800           | 1.2167    1.1562  1.0707 | 1.0069    0.9513  0.8761 |

It is quite obvious that from the results of our calculation shown in Table 4 that both using SiC and ZrC as a coating layer in the CFP will result in a negative temperature coefficient of reactivity because of the multiplication factor will decrease with an increase in the fuel temperature. However, using ZrC as a substitute for SiC will cause a lower core reactivity. This is because of the higher neutron capture cross-section of Zr than that of Si. The less favorable effect in the neutron economy can be compensated by better thermal and mechanical properties of ZrC. Our calculation’s result for RDE is somewhat in agreement with a similar approach for the case of a fluidized bed reactor performed by Agung [11].

The core reactivity of the core also can be calculated from the multiplication factor shown in Table 4 using the usual expression that \( \rho = (k - 1)/k \), where \( \rho \) is the reactivity and \( k \) is the multiplication factor. For the case of SiC core, for example, at 300 K the core reactivity is \((1.326360-1)/1.326360=0.24606 \). While for the ZrC core at the same temperature the value is 0.10628, which is substantially lower than that of SiC.
In terms of the core temperature coefficient, defined as the change in reactivity per degree change in the fuel temperature, \( \alpha = \frac{d\rho}{dT} = \frac{1}{k} \frac{dk}{dT} \), the values for both types of fuel core can be calculated. It is usually expressed in units of \( \text{pcm/}^\circ\text{C} \) or \( \text{pcm/}^\circ\text{F} \). The magnitude and sign (+ or -) of the fuel temperature coefficient or also called Doppler coefficient is primarily a function of the fuel composition, especially the fuel enrichment. The temperature coefficient value for SiC core going from 300 to 600K at 18.75% enrichment is \((1.293510-1.326360)/300 = -0.0001095 \) or \(-1.095 \text{ pcm/K} \). While for the ZrC core, the calculated value is \(-10.93 \text{ pcm/K} \).

The values of the temperature coefficient for HTGR is more negative than the values usually found in LWRs. In a power reactor, in which low enriched fuel (e.g. PWRs and BWRs require 3% – 5% of \(^{235}\text{U}\)) is used, the Doppler coefficient is always negative. The value of the Doppler coefficient depends on the temperature of the fuel and also depends on the fuel burnup. In PWRs, the Doppler coefficient can range, for example, from \(-4 \text{ pcm/K} \) to \(-1 \text{ pcm/K} \) \[19\].

Based on this calculation it can be seen that the Doppler effect can provide a negative instantaneous reactivity coefficient at the HTGR and that the value is better in terms of safety. If a power excursion occurs, excessive fission immediately results in an increase in fuel temperature. The temperature rise in fertile isotopes such as at \(^{238}\text{U}\) results in a relatively high increase in the parasitic neutron capture cross-section for this isotope. And hence reducing the neutron population the in general and reducing the power level of the core.

![Figure 3. Reactivity loss due to increase in fuel temperature.](image)

The measure of safety characteristics of the core neutronics can also be shown by calculating the reactivity loss due to temperature increase as shown in Figure 3. It can be noticed that in general, the reactivity loss in the core using ZrC fuel is more significant than that of SiC fuel. Furthermore, the loss in reactivity seemed to be greater when we use lower enrichment of uranium in the fuel. The figure also shows that while the linearity in the loss of reactivity persists in SiC fuel, the ZrC fuel reactivity loss tapers-off at very high temperatures (above 1500 K) at the lower enrichment of 11.25 and 15%. This indicates that for ZrC fuel it is desirable to use the design value of enrichment of around 18.75%.
Figure 4 shows the neutron flux per lethargy comparison of using ZrC and SiC TRISO fuel in HTGR. The fuel that uses SiC and ZrC follows a typical thermal reactor spectrum. However, if examined closely, one can see a slight hardening of the neutron spectrum of the core using ZrC fuel, especially in the thermal region. This is due to the parasitic neutron absorption in the thermal region by zirconium atoms.

The shape of the spectrum for ZrC and SiC coated fuel doesn’t seem to change with changing fuel temperature. However, the relative height of the spectrum in the fast energy region (higher lethargy group) increases at higher fuel temperatures due to a larger population of neutrons with higher energy. In other words, at lower fuel temperature average neutrons energy in become thermalized easily.
4. Conclusion

The core neutronic investigation on the Indonesian RDE based on High-Temperature Gas-cooled Reactor has been performed. The study on the reactor core neutronic properties was carried out using the SRAC-2006 computer code system. The effects of using ZrC layer instead of SiC layers in the CFP of the pebble fuel were examined at different operating temperatures and different levels of enrichment in the fuel. It was shown that the use of ZrC increases the parasitic neutron capture in the fuel system resulting in lower core reactivity which was confirmed by a slight hardening of the neutron spectrum. However, although both types of fuels gave a good Doppler or temperature coefficient of reactivity, the use of ZrC seems to act more favorably than SiC for the RDE in terms of the inherent safety feature.

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