Investigation of graphite matrix activation in the fuel pebble of Reaktor Daya Eskperimental

I Husnayani, P M Udiyani, S Kuntjoro, M B Setiawan

Center for Nuclear Reactor Technology and Safety, Puspiptek Complex, PTKRN BATAN
Tangerang Selatan, 15410, Indonesia

Email: ihda_husnayani@batan.go.id

Abstract. Program of Reaktor Daya Eksperimen (RDE) construction in Indonesia is still on progress and it is continuously supported by research and development activities. RDE will be built based on pebble-bed type of High Temperature Gas-cooled Reactor utilizing UO2 fuel microkernel dispersed in a 6-diameter spherical solid graphite matrix. Graphite is the major composition of the fuel pebble. In addition to the fuel pebbles, there are also dummy pebbles in the RDE core which are fully composed of graphite without fuel kernel inside. During irradiation in the core, there happen activation of graphite due to neutron captured reaction. In this study, the activation of graphite was investigated through the simulation of ORIGEN2.1 code. The graphite matrix in the pebble fuel was irradiated for five cycles and the dummy pebble was irradiated only for one cycle. From the ORIGEN2.1 simulation, the activation of graphite matrix produces many isotopes of light nuclei but the isotopes that have significant half-life and activity are only H-3, Be-10, and C-14. The activities of H-3, Be-10, and C-14 inside the graphite matrix of one fuel pebble are 3.98E-08 Ci, 2.45E-08 Ci, and 8.99E-07 Ci, respectively. The results for the activation of graphite of one dummy pebble for the same isotopes are 3.48E-10 Ci, 5.13E-09 Ci, and 1.89E-07 Ci, respectively. These isotopes deposit in the graphite matrix and might be released into the primary coolant through some mechanisms such as pebble crack, graphite corrosion, and graphite abrasion due to the friction during the pebble shuffling in the core. However, since the activity of isotopes is small, it can be stated that the fuel pebble of RDE is safe.

Keywords: graphite, activation, HTGR fuel, RDE

1. Introduction
National Nuclear Energy Agency of Indonesia (BATAN)’s plan to build an experimental power reactor named Reaktor Daya Eksperimen (RDE) has to be supported through research and development activities. The decision to choose High Temperature Gas-cooled Reactor (HTGR) as the reactor type of RDE gives rise to many topics that are appealed to be deeply investigated because HTGR belongs to Generation-IV reactor category, a promising reactor type to be commercialized in the future [1]. Although the development of HTGR has been started since 1960s, and the interest to continue research and development (R&D) of HTGR is regained in 1990s showed by GT-MHR and PBMR projects [2]. HTGR R&D program is also conducted by Japan through HTTR project [3]. The most recent one is the construction of HTR-10 in China along with the plan to construct HTR-PM [4].
Some researches about RDE has also been performed by many Indonesian researcher with broad topics such as design optimization [5,6] inventory and sourceterm analysis [7,8], accident analysis [9], fuel optimization [10,11], and etc.

The design of RDE which will be constructed is based on pebble bed type of HTGR with thermal power of 10 MW. RDE utilizes UO2 fuel in the form of microkernel coated by three isotopic layers (TRISO). Thousands of UO2 microkernels are dispersed in a solid spherical graphite matrix with diameter of about 6 cm. During irradiation in the core, beside the nuclear fission happened in the UO2 microkernels, there also happened the activation of graphite matrix due to neutron captured reaction. In this study, the activation of graphite matrix was investigated. It is very important to investigate the graphite matrix activation because almost 99% of RDE’s fuel pebble is composed of graphite matrix. In addition, beside the fuel pebbles, there are also dummy pebbles in the RDE’s core, which are solid graphite with the same size of fuel pebble but without the fuel kernels inside it. Therefore, there will be a considerable amount of activated graphite after the fuel and dummy pebbles are discharged out from the core and will be contributed to the amount of radioactive wastes produced by RDE.

Investigation of radionuclide inventory inside a whole fuel pebbles has been conducted before through some researches [7,12], but the analysis in this work will be different because the radionuclides produced from graphite activation will be deposited inside the graphite matrix, meanwhile the radionuclides produced from fission reaction, decay, or neutron captured reaction of UO2 fuel will be retained in the fuel kernel. The investigation of activation products inside the graphite matrix alone is important those activation produced might be diffused into the primary coolant.

The graphite matrix activation was simulated using ORIGEN2.1 code. The composition of graphite in a single fuel and dummy pebble was calculated based on the available specification data of RDE. Since the design of RDE is adopted from HTR-10, the specification of the fuel pebble will be similar with the fuel of HTR-10. Radionuclides produced from graphite activation reaction were analysed in term of its radioactivity during the fuel dan dummy pebble lifetime in the RDE core.

2. Methodology

The graphite matrix activation was simulated using ORIGEN2.1. ORIGEN2.1 is a well-known computer code for simulating nuclear fuel depletion and for simulating neutron irradiation of materials as well [13]. Simulation of ORIGEN2.1 needs the parameters of reactor including thermal power, fuel composition, and irradiation history. The fuel of RDE is a 6 cm diameter spherical pebble contained UO2 kernel dispersed in graphite matrix. UO2 kernel is coated by three isotropic layers (TRISO) which are Inner Pyrolitic Carbon (IPyC), Silicon Carbide (SiC), and Outer Pyrolitic Carbon (OPyC). Meanwhile, the dummy pebbles are composed of solid graphite but contain no fuel kernels. The specification of fuel and dummy pebbles are shown in Table 1.

| No | Parameter            | Fuel pebble | Dummy pebble |
|----|----------------------|-------------|--------------|
| 1  | Heavy metal mass     | 5 grams per pebble | -            |
| 2  | U-235 enrichment     | 17%         | -            |
| 3  | Graphite density     | 1.76 g/cm³  | 1.76 g/cm³  |
| 4  | Graphite mass        | 205 grams   | 195.56 grams|

The ORIGEN2.1 simulation were performed two times, the first simulation is for simulating the graphite activation of the fuel pebble, and the second simulation is for simulating the graphite activation of the dummy pebble. In the first simulation, one fuel pebble was irradiated in the RDE core in for 1080 days, which represents the lifetime of the fuel pebble during five cycles in the core. It can be taken as an average that the fuel pebble takes 216 days of irradiation for each cycle. This period has taken into account the 40 days cooling time of the fuel pebble in the discharged tube after each cycle.
This cooling period was also considered in the ORIGEN2.1 simulation. In the second simulation, the dummy pebble was irradiated for 216 days including the 40 days of cooling time because the dummy pebble pass through the core only one time before it is moved out from the core. Based on ORIGEN2.1 calculation for RDE in the previous research [10], the neutron flux in the RDE core is in the order of 10^{13} neutrons/cm^2s. The output of ORIGEN2.1 that was considered is the radioactivity of isotopes produced from graphite activation. These isotopes were then analysed regarding their radioactivity evolution during the fuel and dummy pebble lifetime in the core.

3. Results and Discussion

The results of ORIGEN2.1 simulation for the graphite matrix irradiation showed that there are some isotopes produced from the graphite activation reaction, both for the graphite in the fuel and dummy pebble. Table 2 shows the list of some of isotopes produced from graphite activation in the fuel pebble that have significant activities. It can be shown that He-6 and Be-8 is the isotopes with the highest radioactivity produced during graphite irradiation. However, the half-life of those two are very short, which is 801 milliseconds for He-6 and 8.2E-17 seconds for Be-8, so that both of them are vanished during the 40 days pebble cooling period in discharged tube. At the end of the first cycle, and also for the next cycles as well, the remaining isotopes left in the graphite matrix are only H-3, Be-10, and C-14. Although only three isotopes remained, there three isotopes are important isotopes which have significant radiological impact, both for human and environment especially the Tritium (H-3) and C-14. Moreover, the half-life of H-13 and C-14 is also quite long, which are 12.3 years and 5.7 years respectively so that their presence must get more attention.

Table 2. Activity (Ci) of some isotopes produced from the graphite activation of one fuel pebble at the end of the first cycle

| Isotopes | 50     | 100    | 150    | 176    | 216    |
|----------|--------|--------|--------|--------|--------|
| H-3      | 7.72E-12 | 6.16E-11 | 2.07E-10 | 3.34E-10 | 3.32E-10 |
| He-6     | 1.69E-06 | 3.37E-06 | 5.06E-06 | 5.94E-06 | 0.00E+00 |
| Be-8     | 2.91E-06 | 5.81E-06 | 8.72E-06 | 1.02E-05 | 0.00E+00 |
| Be-10    | 1.39E-09 | 2.78E-09 | 4.18E-09 | 4.90E-09 | 4.90E-09 |
| C-14     | 5.11E-08 | 1.02E-07 | 1.53E-07 | 1.80E-07 | 1.80E-07 |

Table 3 shows the activity of H-3, Be-10, and C-14 at the end the first to the fifth cycle including the 40 days cooling period. It can be implied that this activity is quite high since this is the activity of isotopes deposited inside the graphite matrix of one fuel pebble. In fact, there are plenty of fuel and dummy pebbles in the core. For example, if there are 100 fuel pebbles discharged out from the core each day, the activity of the isotopes must be multiplied by 100 and this will result in a higher level of radioactivity.

Table 3. Activity (Ci) of isotopes produced from the graphite activation of one fuel pebble at the end of each cycle

| Isotopes | Cycle-1 | Cycle-2 | Cycle-3 | Cycle-4 | Cycle-5 |
|----------|---------|---------|---------|---------|---------|
| H-3      | 3.32E-10 | 2.63E-09 | 8.79E-09 | 2.06E-08 | 3.98E-08 |
| Be-10    | 4.90E-09 | 9.80E-09 | 1.47E-08 | 1.96E-08 | 2.45E-08 |
| C-14     | 1.80E-07 | 3.60E-07 | 5.40E-07 | 7.20E-07 | 8.99E-07 |

The results for activation reaction of the graphite matrix in dummy pebble are showed in Table 4. In general, the isotopes produced and its activities are the same with the results of the activation of the
The graphite matrix in the fuel pebble at the end of the first cycle. The difference between the two is that the fuel pebble will be reloaded into the core when they do not yet reach the targeted burnup but the dummy pebble will be directly discharged out of the core after the first cycle.

**Table 4.** Activity (Ci) of some isotopes produced from the graphite activation of one dummy pebble at the end of the first cycle

| Isotopes | Irradiation time (days) |
|----------|-------------------------|
|          | 50          | 100     | 150     | 176     | 216     |
| H-3      | 8.09E-12    | 6.45E-11 | 2.17E-10 | 3.50E-10 | 3.48E-10 |
| He-6     | 1.77E-06    | 3.53E-06 | 5.30E-06 | 6.22E-06 | 0.00E+00 |
| Be-8     | 3.05E-06    | 6.09E-06 | 9.14E-06 | 1.07E-05 | 0.00E+00 |
| Be-10    | 1.46E-09    | 2.92E-09 | 4.37E-09 | 5.13E-09 | 5.13E-09 |
| C-14     | 5.35E-08    | 1.07E-07 | 1.61E-07 | 1.89E-07 | 1.89E-07 |

In relation to the safety of RDE, the deposition of H-3, Be-10, and C-14 in the graphite matrix will be an important concern and can be taken as the sourceterm for analysing the radionuclide release from the pebble. There is always the possibility that the isotopes deposited in the graphite matrix might be release into the primary coolant or deposited in the core structural materials through some mechanism. The first possible release mechanisms is through the crack of the pebble. The crack on the pebble can be caused by mechanical failure during the pebble manufacturing process [14], failure during the pebble transport and handling process, or the damage caused by neutron irradiation itself. The crack on the pebble might cause the diffusion of the isotopes from inside to the outside of the graphite matrix.

Another mechanism that allows the displacement of the isotopes in the graphite matrix is through the abrasion of the graphite matrix due to the pebble handling process and pebble shuffling in the core. During pneumatic lift, the pebbles undergo multiple collisions with the stainless steel lifting pip, thereby causing abrasion of the graphite and producing graphite dust [15]. Most of the graphite dust is produced from the wear of fuel pebble during the cycle due to the friction and kinetic interaction between the pebbles [16]. Another mechanism is due to the graphite corrosion caused by chemical reaction of impurities or fission products. The corrosion of graphite causes the thinning of the graphite layers so that the isotopes might be diffuse from inside to the outside of the graphite matrix. Another important release mechanism is the abrasion of the graphite matrix due to the friction perceived by the pebbles during the circulation and shuffling process in the core.

The probability of the isotopes deposited in the graphite matrix released into the primary coolant depends on the condition of the fuel pebble during the RDE operation. There must be safety criteria for the fuel pebble to be considered as a failed fuel pebble so that it must be moved out of the RDE core. Although there is always the probability of the isotopes release, the RDE fuel pebble is still considered safe because the isotopes activity is not very significant.

**Conclusion**

The investigation of graphite matrix activation of RDE’s fuel pebble and fuel dummy has been performed by using ORIGEN2.1 computer code. The graphite matrix which constructs almost 99% of fuel pebble and as the whole composition of dummy pebble produces the isotope of some light nuclei as the results of neutron activation reaction. The isotopes with significant activity produced are H-3, He-6, Be-8, Be-10, and C-14. However, due to the short hall-life of He-6 and Be-8 and the 40 days cooling period of pebble in the discharged tube, the remaining isotopes deposited in the graphite matrix are only H-3, Be-10, and C-14. The activity of these three isotopes at the end the fifth cycle of fuel pebble is 3.98E-08 Ci, 2.45E-08 Ci, and 8.99E-07 Ci, respectively. The results for the activation of graphite in dummy pebble for the same isotopes are 3.48E-10 Ci, 5.13E-09 Ci, and 1.89E-07 Ci, respectively. These results are the activities of a single fuel and dummy. Since there are many fuel and
dummy pebble discharged out from the core, these activities should be multiplied by the corresponding number of fuel and dummy pebble. Although only three isotopes remained, there isotopes attract significant concern since they are categorized to isotopes that have impact to human and environment. The release of these isotopes into the primary coolant is possible through some mechanisms including the pebble crack due to manufacturing or fuel handling process, the graphite corrosion due to chemical attack, and the graphite abrasion due to the pebble shuffling in the core.

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References
[1] IAEA, 2010, High temperature gas-cooled fuels and materials, *International Atomic Energy Agency*, Vienna
[2] Dominique H., et al, 2012, R&D needs for near-term HTRs, *Nuclear Engineering and Design*, 251 p. 131 – 138
[3] Atsushi Shimizu, 2014, Development of operation and maintenance technology for HTGRs by using HTTR (High Temperature Engineering Test Reactor), *Nuclear Engineering and Design*, 271 p. 499 – 504
[4] Chao fang, 2016, The R&D of HTGR high temperature helium sampling loop: From HTR-10 to HTR-PM”, *Nuclear Engineering and Design*, 306 p. 192 – 197
[5] Sudadiyo, S., 2018, Preliminary Design Of RDE Feedwater Pump Impeller, *Tri Dasa Mega*, 20 No 1
[6] Dibyo, S., Irianto D., 2017, Design analysis on operating parameter of outlet temperature and void fraction in RDE steam generator, *Tri Dasa Mega*, 19, no 1
[7] Kuntjoro S., Udiyani P.M. 2016., *Analisis inventory Reaktor Daya Eksperimental Jenis Reaktor Gas Temperatur Tinggi*. Urania. 22 p. 53–64.
[8] Pande Made, S kuncoro, 2017, estimation of routine discharge of radionuclides on power reactor experimental RDE, *Urania*, 23 no 1
[9] Tjahjono H. 2017, Investigation of RDE thermal parameters changes in response to long-term station black out. *Tri Dasa Mega*. 19 no2 p. 83–92.
[10] Husnayani I, Udiyani P.M et. al. 2017, Kr-85 activity as burnup measurement indicator in a pebble bed reactor based on ORIGEN2.1 computer simulation, *J. Phys: Conf. Ser* 962 011001
[11] P H Liem, H N Tran, T M Sembiring. 2017, Alternative fuelling scheme for the indonesian experimental power reactor (10MWth Pebble bed HTGR), *Energy Procedia*, 131 p. 69-76
[12] Husnayani I, Kuntjoro S., Udiyani P.M. 2016, Fission products inventory analysis of HTGR fuel. *Proc. of Seminar Nasional Teknologi Energi Nuklir*. Batam
[13] Husnayani, I., 2016, Calculation of radionuclide content of nuclear material using ORIGEN2.1 computer code, *Sigma Epsilon*, 19 no2 p. 83–92.
[14] Zhou Xiang-Wen, Zhang Jie, Lu Zhen-Ming, 2014, Study on the carbonization process in the fabrication of pebble fuel elements, *Nuclear Engineering and Design*, 271, p. 149-153
[15] K. Shen, J. Su, H. zhou, 2015, Abrasion behaviour of graphite pebble in lifting pipe of pebble bed HTR, *Nuclear Engineering and Design*, 293 p. 395-402
[16] L. Xiaowei, W. Xiaoxin, S. li, 2017, Nuclear graphite wear properties and estimation of graphite dust production in HTR-10, *Nuclear Engineering and Design*, 315 p. 35-41.