Kr-85m activity as burnup measurement indicator in a pebble bed reactor based on ORIGEN2.1 Computer Simulation

To cite this article: I Husnayani et al 2018 J. Phys.: Conf. Ser. 962 012005

View the article online for updates and enhancements.
Kr-85m activity as burnup measurement indicator in a pebble bed reactor based on ORIGEN2.1 Computer Simulation

I Husnayani, P M Udiyani, S Bakhri, G R Sunaryo

Center for Nuclear Reactor Technology and Safety, Puspiptek Complex, building no. 80, Serpong, Tangerang Selatan 15314 Indonesia

Abstract. Pebble Bed Reactor (PBR) is a high temperature gas-cooled reactor which employs graphite as a moderator and helium as a coolant. In a multi-pass PBR, burnup of the fuel pebble must be measured in each cycle by online measurement in order to determine whether the fuel pebble should be reloaded into the core for another cycle or moved out of the core into spent fuel storage. One of the well-known methods for measuring burnup is based on the activity of radionuclide decay inside the fuel pebble. In this work, the activity and gamma emission of Kr-85m were studied in order to investigate the feasibility of Kr-85m as burnup measurement indicator in a PBR. The activity and gamma emission of Kr-85 were estimated using ORIGEN2.1 computer code. The parameters of HTR-10 were taken as a case study in performing ORIGEN2.1 simulation. The results show that the activity revolution of Kr-85m has a good relationship with the burnup of the pebble fuel in each cycle. The Kr-85m activity reduction in each burnup step, in the range of 12% to 4%, is considered sufficient to show the burnup level in each cycle. The gamma emission of Kr-85m is also sufficiently high which is in the order of 10^10 photon/second. From these results, it can be concluded that Kr-85m is suitable to be used as burnup measurement indicator in a pebble bed reactor.

1. Introduction

Pebble Bed Reactor (PBR) is a high temperature gas-cooled reactor which uses graphite as a moderator and helium as a coolant. PBR is categorized as a Generation IV reactor because it offers many advances compared to conventional reactor such as Light Water Reactor (LWR). The advantage of PBR over LWR is mainly due to its fuel best performance and reliability to operate in high temperature condition. The fuel of PBR is a 6-cm in diameter graphite sphere (pebble) containing thousands of UO2 micro-kernels coated by Tri-structural Isotropic (TRISO) carbon layers; Inner Pyrolitic Carbon (IPyC), Silicon Carbide (SiC), and Outer Pyrolitic Carbon (OPyC) [1]. Typically, there are two schemes of fuel loading in PBR which are once-through-then-out (OTTO), where the fuel pebbles circulate only one time in the core, and multi-pass scheme, where the fuel pebbles circulate for several cycles in the core before being discharged out of the reactor core [2].

In a multi-pass Pebble Bed Reactor, when the fuel pebble completes each cycle, its burnup level must be measured in order to determine whether the burn-up of the pebble is still below the burn-up target so that it must be reloaded into the core for another cycle or it has reached the burn-up target so that it must be discharged out of the core. Burnup measurement in PBR is performed by online measurement without shutting down the reactor. Burnup is a variable that shows the amount of energy produced by the fuels from fission reaction of the fissile materials. Performing online burnup measurement is usually conducted by means of passive system, which is by detecting signal from the fuels and then relates it with the burnup. Those signals include neutron and gamma rays.
The feasibility of burnup measurement using passive neutron counting technique has been studied by Su, et al. [3] and gamma emission technique has been studied by Yan et al. [4]. Regarding the burnup measurement using gamma emission spectroscopy, type of radionuclide that will be used as burnup measurement indicator must fulfill some criteria. For online burnup measurement, the radionuclide must have a short half-life, simple mode of production and decay, and low neutron absorption cross section [5]. In this work, the feasibility of Krypton-85m (Kr-85m) as a burnup indicator in a Pebble Bed Reactor was studied. The activity and gamma emission of Kr-85m was analysed in relation with the burnup of the PBR fuel pebble in each cycle. The activity and gamma emission of Kr-85m is calculated by using ORIGEN2.1 computer code. ORIGEN2.1 is a well-known and worldwide-used fuel depletion code developed by Oak Ridge National Laboratory [6]. Parameters of Chinese 10-MWth PBR (HTR-10) were taken as a case study and in constructing the input file for ORIGEN2.1 simulation.

2. Theory
Kr-85m is an isomer of krypton produced from fission reaction in nuclear reactor fuels. Some characteristics of Kr-85m are shown in the Table 1. These characteristics mainly fulfill the criteria for burn-up measurement indicator. The half-life of Kr-85m is relatively short, which is less than 24 hours, so that it is suitable for online burn-up measurement. The highest probability of gamma emission is in the energy of 151.195 KeV. The fission yield of Kr-85m is relatively high so that the concentration of Kr-85m in the fuel as well as its activity will be sufficient to be detected. The thermal absorption cross section of Kr-85m is far less than one barn so that neutron activation reaction of Kr-85m can be ignored and the concentration of Kr-85m in the fuel pebble will be mainly related to the burnup level of that fuel pebble.

Table 1. Characteristics of Kr-85m [5]

| Characteristic                          | Value             |
|---------------------------------------|-------------------|
| Half life                             | 268.8 min         |
| Emission probability 151.195 KeV       | 75%               |
| 129.81 KeV                           | 0.3%              |
| 451 KeV                              | 0.01%             |
| 304.87 KeV                           | 14%               |
| U-235 fission yield                   | 5.89E-05          |
| Thermal absorption cross section      | 2.35E-02 barn     |
| Mode of production                    | 1                 |

2.1. ORIGEN2.1 Computer Code
ORIGEN2.1 is a versatile depletion code used for calculating the isotopic composition and some decay characteristics of the reactor fuel. The nuclide density rate calculation in the ORIGEN code is done using formula as follows [7]:

\[
\frac{dN_i}{dt} = \sum_{j=1}^{N} l_{ij} \lambda_j X_j + \varphi \sum_{k=1}^{N} f_{ik} \sigma_k X_k - (\lambda_i + \varphi \sigma_i + r_i)X_i + F_i, \ i = 1, \ldots, N(1)
\]

\(N_i\) = density of nuclide i
\(N\) = number of nuclides
\(l_{ij}\) = fraction of radioactive disintegration by other nuclide, which lead to formation of species I
\(\varphi\) = position- and energy-averaged neutron flux \((n \text{ cm}^{-2} \text{s}^{-1})\)
\(f_{ik}\) = fraction of neutron absorption by other nuclides, which lead to formation of species i
\(\sigma_k\) = spectrum-averaged neutron absorption cross section of nuclide k (barn)
\[ r_i = \text{continuous removal rate of nuclide } i \text{ from the system} \]
\[ F_i = \text{continuous feed rate of nuclide } i \]

The photon release rates (photon/second) of the nuclide \( i \) can be calculated from the equation as below:
\[
P_i = g_i \lambda_i N_i
\]  
where \( P_i \) is the photon release rate, \( g_i \) is the photon released per decay, which is given in ORIGEN2.1 photon library, \( \lambda_i N_i \) are described above.

3. Methodology
HTR-10 was taken as a case study for PBR fuel handling system as well as reactor parameters used in this study. In HTR-10, the fuel pebbles circulates in the reactor core for five cycles. The fuel loading scheme of HTR-10 is shown in the Figure 1. SG01 transports the fuel pebbles one by one to BP01 where the burnup of the pebble is measured. If it has not reached its maximum burnup, the pebble is transported back to the core through the lighting pipe 2. If the fuel pebble has reached its maximum burnup, then it is transported to the FS02 through the lifting pipe 3. If some fresh fuel pebbles need to be added into the core, the fuel pebbles will be loaded into the core from FS01 through the lifting pipe 1.

![Figure 1. Fuel handling system of the HTR-10 [9]](image)

In HTR-10, the average target burnup of the fuel is designed for 80,000 MWD/MTU. In the equilibrium state, the number of fresh pebbles and recycled pebbles needed per EFPD is 25 and 100, respectively [8]. It means that there are 125 pebbles performed burn-up measurement per day. It was assumed that one pebble takes about 11 minutes from leaving the reactor core, performing burn-up measurement, and then recycling into the core or moving out into the discharged tube. During this 11 minutes, because of its position is not in the core, there is no fission reaction in the fuel, it is only the radionuclide decay process that occurs in the pebble fuel.

The irradiation process of the pebble fuels during their residence in the reactor core was simulated by using ORIGEN2.1 computer code, from cycle 1 through cycle 5. The reactor parameters used in creating input data for ORIGEN2.1 simulation are shown in Table 2. The material composition of the fuel pebbles, which is shown in Table 3, was calculated based on the heavy metal loading and enrichment of the fuel. Since the thermal power of the reactor is 10 MW and designed maximum burn-
up of the pebble is 80.000 MWD/MTU, it is assumed that the pebble resides in the core for about 216 days per cycle or 1080 days for full cycle. After each cycle, the fuel pebble was simulated to undergo decay for about 11 minutes. The activity and photon intensity of Kr-85m were acquired during this decay period.

Table 2. Parameters used as input in ORIGEN2.1 simulation [10]

| No | Parameter      | Nominal | Unit   |
|----|----------------|---------|--------|
| 1  | Thermal power  | 10      | MW     |
| 2  | Number of pebble | 27000  | pebbles |
| 3  | Heavy metal loading | 5      | grams   |
| 4  | Enrichment     | 17      | %      |

Table 3. Material composition in the fuel pebbles

| No | Material | Nominal   | Unit   |
|----|----------|-----------|--------|
| 1  | U-235    | 22937.32  | grams  |
| 2  | U-234    | 41.15     | grams  |
| 3  | U-238    | 111946.9  | grams  |
| 4  | Oxygen   | 18180.19  | grams  |
| 5  | Carbon   | 5283195   | grams  |
| 6  | Silicon  | 34818.25  | grams  |

4. Results and Discussion

The results for Kr-85m activity as a function of burnup are shown in Figure 2. Around the first 20 days of irradiation, the activity of Kr-85m increases significantly until it reaches 3.83 Ci. After that, the activity of Kr-85m decreases consistently when the burn-up of the fuel pebble increases. Several sharp activity reduction emerged at each cycle indicate the decay process of Kr-85m when it leaves the reactor core and moves to the burnup measurement position. This rapid reduction is due to the short half-life of Kr-85m. The concentration and activity of Kr-85m reduce rapidly since there is no Kr-85m produced from fission reaction during this decay period. From this Kr-85m activity revolution trend, it can be implied that it has a good relationship with the fuel burnup increment. The percentages of the activity reduction of Kr-85m at several burnup steps are shown in Table 4. This reduction percentage is sufficient to determine the burnup level of the fuel at each cycle. These reduction percentages are considered acceptable since the best range of activity reduction for burnup measurement is between 5% and 35% [11].

This result also indicates that the use of Kr-85m as burnup indicator, from the point of view of its activity reduction in relation with the burnup increment, is suitable for online measurement. Compared to other radionuclide such as Cs-137 whose half-life is about 37 years, the activity of Cs-37 will be steady and not be significantly different at each burnup step. Thus, a short half-life radionuclide, which in this case is Kr-85m, is the potential option for use in the online burnup measurement in pebble bed reactor. In order to obtain a better measurement results, the instrument used for burnup measurement must be quite sensitive with high accuracy and with detection error of less than 4%.

The activity of Kr-85m when performing decay at each cycle is shown in Figure 3. It has been stated before that Kr-85m is assumed to undergo decay for 11 minutes after each cycle. If the burnup measurement is performed within 2 minutes after the pebble leaving the reactor core, the activity of Kr-85m, in approximation, at cycle 1 is 3.38 Ci, at cycle 2 is 3.09 Ci, at cycle 3 is 2.91 Ci, at cycle 4 is 2.76 Ci, and at cycle 5 is 2.65 Ci. Since the half-life of Kr-85 in quite long (268 minutes) compared to the decay time (11 minutes), the activity of Kr-85m does not alter very significantly in the process of the decay within this 11 minutes period. This makes the measurement at each cycle unaffected by the alteration of the activity due to reasonable comparison between the half-life of Kr-85m and the decay period. Again, since the activity reduction is varied between 12% at cycle 1 to 4% at cycle 5, this
condition requires the use of instrument that has small detection error in order to obtain the more accurate measurement data.

![Activity of Kr-85m vs Burnup](image.png)

**Figure 2.** The activity of Kr-85m as a function pebble fuel burnup

| No | Cycle | Burnup increment | Activity reduction |
|----|-------|------------------|-------------------|
| 1  | Cycle 1 | 0 – 16000 MWD/MTU | 12% |
| 2  | Cycle 2 | 16000 – 32000 MWD/MTU | 9% |
| 3  | Cycle 3 | 32000 – 48000 MWD/MTU | 6% |
| 4  | Cycle 4 | 48000 – 64000 MWD/MTU | 5% |
| 5  | Cycle 5 | 64000 – 80000 MWD/MTU | 4% |

The results for photon intensity from Kr-85 decay are shown in Figure 4. This photon is 0.151 MeV gamma emitted from beta decay reaction of Kr-85m. The emission probability of this gamma is 75% which is the most dominant compared to the other gamma rays emitted by Kr-85m which are 0.3%, 0.01% and 14% for gamma energy 0.129 MeV, 0.451 MeV, and 0.304 MeV, respectively. This photon intensity, which is in the order of $10^{10}$ photon per second, is considered more than sufficient to be detected since it can be categorized as a strong signal produced from a radionuclide. The instrument that will be used to detect this gamma emission should have a good resolution, which can differentiate the gamma rays emitted by Kr-85m from another gamma emission emitted by other radionuclides in the fuel pebble. The challenge is that there is another radionuclide which emits almost similar gamma energy as Kr-85 which is Sr-85m. However, the yield of Sr-85m in U235 fission reaction is much lower than Kr-85m yield which is merely $1.47 \times 10^{-13}$. The yield of Kr-85m is about $8.5 \times 10^{-5}$. It can be implied that the concentration of Sr-85m will be much lower than Kr-85m and the gamma rays with the energy of 0.151 MeV detected by the instrument can be considered mainly emitted by Kr-85m and the contribution of Sr-85m can be ignored.
Figure 3. The activity of Kr-85m performing decay in each cycle

Figure 4. Photon intensity of Kr-85m at each cycle
5. Conclusion
The activity and gamma emission of Kr-85m have been studied in order to investigate the feasibility of Kr-85m to be used as an online burnup measurement indicator in pebble bed reactor. The calculation of the activity and photon intensity of Kr-85 were performed by using ORIGEN2.1. The results show that Kr-85m activity has a good relationship with the burnup of the PBR fuel pebble. The trend of Kr-85m activity revolution is consistent with the fuel burnup increment. The activities of Kr-85m reduce for about 12%, 9%, 6%, 5%, and 4% at cycle 1, cycle 2, cycle 3, cycle 4, and cycle 5, respectively. This reduction percentage is acceptable for determining the burnup level of the fuel at each cycle. The results for gamma emission of Kr-85m show that the emission intensity is in the order of 10¹⁰ photon/second. This intensity is sufficiently high to be detected by the instrument of fuel burnup measurement. From these results, it can be concluded that Kr-85m fulfilled the criteria as an online burnup indicator in a pebble bed reactor.

Acknowledgement
The author would like to acknowledge the Head of Center for Nuclear Reactor Safety and Technology (PTKRN) BATAN for supporting this work by granting the government research fund (DIPA-PTKRN-BATAN) of the year 2017.

References
[1] Futterer M A, Fu L, Sink C et al 2014 Status of the very high temperature reactor system Progress in Nuclear Energy 77 266 – 281
[2] IAEA TECDOC 2010 High Temperature Gas-Cooled Fuels and Materials International Atomic Energy Agency
[3] Bingjing Su, Zhongxiong Zhao, Jianwei Chen et al 2006 Assessment of on-line burnup monitoring of pebble bed reactor fuel by passive neutron counting Progress in Nuclear Energy 48 686 – 702
[4] Yan Wei-Hua et al 2014 Prototype studies on the nondestructive online burnup determination for the modular pebble bed reactors Nuclear Engineering and Design 267 172 – 179
[5] Akyurek T, Tucker, L P and Usman S 2014 Review and characterization of best candidate isotopes for burnup analysis and monitoring of irradiated fuel Annals of Nuclear Energy 69 278 – 291
[6] Benkharfia H, Zidi T and Belgaid M 2016 Lumped pseudo fission products during burnup step in MCNP5 – ORIGEN coupling system Progress in Nuclear Energy 88 277 – 284
[7] I. Husnayani 2016 Calculation of radionuclide content of nuclear materials by using ORIGEN2.1 computer code Sigma Epsilon 19 20 – 25
[8] Yongwei Y, Zhengpei L, Xingqing J and Zongxin W 2002 Fuel management of the HTR-10 including the equilibrium state and the running-in phase Nuclear Engineering and Design 218 33 – 41
[9] Liu H, Du D, Han Z and Chang B 2017 Pneumatic Transportation Pattern of Fuel Pebbles in a Pebble Bed Reactor Annals of Nuclear Energy 99 434 – 443
[10] Scari M E, Costa A L and Pereira C 2016 HTR steady state and transient thermal analyses 41 7192 - 96
[11] Jianwei Chien 2004 Online Interrogation of Pebble Bed Reactor Fuel Using Passive Gamma Ray Spectroscopy PhD Dissertation University of Cincinnati