IDENTIFICATION OF IMPORTANT FISSION PRODUCT NUCLIDES FROM SPENT FUEL OF PEBBLE BED HTGR AT MAXIMUM BURN-UP

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Abstract

As Indonesia is planning to build an experimental power reactor of pebble bed HTGR type, it is important to determine the content of important fission product nuclides of its spent fuel. Identification the amount and type of the FPs is the first step toward the implementation of safeguards policy, management of the spent fuel and addressing the source term strength in case of an accident. The calculation was done using Monte Carlo method MCNP6 Code. It is intended to calculate the amount of nuclides that are important to safeguards, such as the remaining U-235 and the produced Pu-239; the amount of long-live minor actinides that are subject to spent fuel management; the amount of fission products that are important in addressing radioactive release in case of an accident.

1. INTRODUCTION

National Nuclear Energy Agency (BATAN) is planning to build experimental power reactor located in Serpong area, which is close to the existing 30 MW MTR type research reactor RSG GAS. The reactor is expected to produce power 10 MWt of pebble bed High Temperature Gas Cooled Reactor (HTGR). The fuel element refers to HTR-10.

As there are about 27,000 pebble fuels in the equilibrium core, safeguards regulatory supervision is a challenge faced by regulatory body. Due to random fuel flow pattern from entering through exiting the core, the burn up calculation might produce high uncertainty. The fuel composition especially the remaining uranium-235 and uranium-238 and produced plutonium-239 are primary initial data for safeguards inspection.

Jeong et.al [1] has developed model for fission product release from the pebble fuel under normal and accident condition. Prior to that, the fission product inventory has to be determine. This paper will estimate the fission product inventory which is used for source term calcultion, and actinide composition used for safeguards inspection. The burn up calculation employes MCNP6 code by introducing a model of infinite lattice applying periodic boundary conditions representing equilibrium core. The HTR-10 core evolution from first criticality through equilibrium core has been introduced by Y. Yang et.al [2]. Liem et.al [3] introduced an alternative fuel loading scheme for the upcoming Indonesian experimental HTGR pebble bed reactor.

This paper is intended to map out the spent pebble fuel composition at maximum burn up of 80,000 MWd/t, such as actinide composition and fission product activity. The actinide composition is important information for nuclear material accounting, on other hand fission product inventory such as Cs-137, Kr-85, I-135, I-131 and Sr-90 is crucial information for estimating fission product release and dose calculation to the public during an accident.

I-131 is an important fission product because of its volatile characteristic, and if it is released to the environment it might be inhaled or be deposited in plantations. If it is inhaled or swallowed, it could be deposited in thyroid gland and pose significant radiation dose to thyroid. Thyroid can accumulate both stable and radioactive iodine. Absorption of radioactive iodine can cause acute, chronicle and delayed effect (such as cancer). Chernobyl accident in 1986 has proved the existence of increase in significant

thyroid cancer in former Soviet Union countries during 1992-2000[4]. One of actions to protect from risk of radioactive iodine is consuming iodine potassium (KI) before receiving radioactive iodine exposure.

Kr-85 has half-life of 10.73 years, its radiological effect ranges from middle to long-term. Kr-85 emits two β- with average energy 47.5 keV and 251.4 keV, and also gamma ray of 514 keV. The daughter Kr-85 decay is Rb-85[5].

Cesium is also volatile at higher temperature than that of iodine volatility [6]. Cesium has much longer half-life than I-131, and can stay in the environment for several years after release. Cs-137 becomes long term problem because it stays in the upper soil surface.

Sr-90 is not easily released from the fuel compared to iodine and cesium, but the radionuclide may cause significant biological effect, because it behaves like calcium, accumulated in the bone when entering the body.

2. CALCULATION MODELS

As there are approximately 27,000 fuel pebbles in the equilibrium core of 10 MW and it is difficult to conduct burn up calculation to all fuel pebbles due to its number and their random geometrical arrangement, one fuel pebble is selected instead. The simplified burn up calculation approach is employed as the fuel flow in the core is random.

In general the composition of a nuclide which is the result of neutron reaction with a material in the reactor can be formulated as:

$$\frac{dN_i(t)}{dt} = \sum_{j \neq i} f_{j \to i} \lambda_j N_j(t) + \sum_{k \neq i} fact(t) \{ g_{k \to i} C_k + \gamma_{k \to i} F_k + h_{k \to i} W_k \} N_k(t) - [\lambda_i + Fact(t) \{ A_i + W_i \}] N_i(t) \quad (1)$$

where:

i,j,k	: depleted nuclide number
N	: burn up nuclide number density
λ, f	: decay constant and branch ratio
g, γ, h	: yield fraction of each transmutation
F	: Relative microscopic fission reaction rate
A	: Relative microscopic absorption reaction rate
C	: Relative microscopic capture reaction rate (=A-F)
W	: Relative microscopic (n,2n) reaction rate
$Fact$	: Normalization factor to convert relative reaction rates to absolute ones

From Eq. (1) it can be narrated that a nuclide i can be produced from decay of nuclide j, neutron capture of nuclide k, fission reaction and (n,2n) reaction. At the same time, the destruction of nuclide i come from its decay, neutron absorption producing another nuclide, and (n,2n) reaction changing itself to lighter nuclide.

Table 1 describes the material specification of pebble fuel for burn-up calculation with MCNP6 code.

TABLE 1. FUEL PEBBLE SPECIFICATIONS IN THE EQUILIBRIUM CORE [7]

Fuel Pebble	
Density of graphite in matrix and outer shell	1.73 g/cm^3
Heavy metal (uranium) loading per pebble	5.0 g
Uranium enrichment	17 %
Equivalent natural boron content of impurities in uranium	4.0 ppm
Equivalent natural boron content of impurities in graphite	1.3 ppm
Volume fraction of pebbles in the core	0.61
UO_2 density	10.4 g/cm^3
Coating layer materials (starting from kernel)	Buffer/PyC/SiC/PyC
Coating layer densities, respectively (g/cm^3)	1.1/1.9/3.18/1.9
Average number of fuel particles per pebble	8,335
Volume fraction of fuel particles in pebble fuel zone	5.0248
Maximum burn up, MWd/t	80,000

The burn up calculations was conducted using Monte Carlo method MCNP6 code. The geometrical model consists of a box filled with a fuel pebble in its centre and surrounded by 1/8 of pebble volume in such that volume ratio of fuel pebble is 0.61. To represent the fuel pebble located in the middle of the core, the box is infinitely repeated in the lateral direction, and therefore periodic boundary conditions are applied in the X and Y directions. As shown in Fig. 1, surface 1 is applied with periodic boundary condition to surface 2, and similarly surface 3 to surface 4. The bottom and the top surfaces are applied with reflective boundary condition. This geometrical model will represent that the burned fuel is exposed by neutron energy spectrum similar to the one located in the middle of the core. Applying the boundary conditions will produce k_{eff} =1.49939.

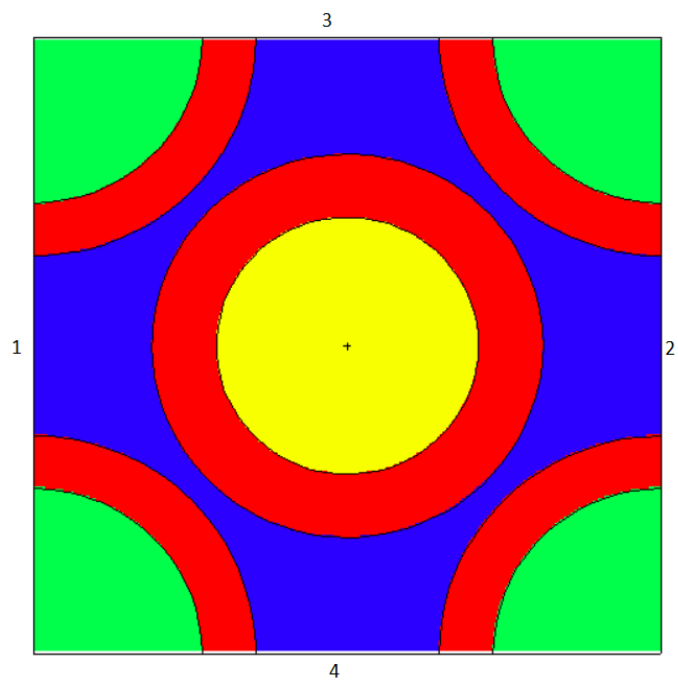


FIG. 1. MCNP6 geometrical model for burn up calculation.

Figure 1 shows the 2-D view geometrical model with MCNP6 for burn up calculation. The fuel zone in the pebble is homogenized among 8,335 coated fuel particles (CFPs) and graphite matrix. The fuel zone

is cladded with graphite material (red). Only the fuel zone of the central pebble (yellow) is burned. The evolution of fission product compositions in the neighbouring pebbles is neglected.

Tabel 2 elaborates the isotopic composition of homogenized fuel zone and graphite shell for MCNP6 material card. The fuel zone and graphite shell have density of 1.74008 g/cm3 and 1.73 g/cm3, respectively.

TABLE 2. ISOTOPIC COMPOSITION OF FUEL ZONE AND GRAPHITE SHELL

Isotope	Fuel Zone (g)	Graphite Shell (g)
U-235	8.5000E-01	-
U-238	4.1500E+00	-
Oxygen	6.7336E-01	-
B-10	1.9446E-04	1.4234E-04
B-11	8.6062E-04	6.2994E-04
Silicon	1.2913E+00	-
Carbon	9.0414E+02	6.5944E+02

The central pebble is assumed to have produced average power of 0.3704 kW (10 MW/27000). The maximum burn up of 80,000 MWd/t [8] will be reached after 1080 days operation. As the fuel operating temperature is about 1200 K [9], the nuclear data at such temperature are employed for uranium, graphite and other isotopes in the homogenized fuel zone.

3. RESULTS AND DISCUSSIONS

The burn up calculation was conducted by KCODE calculation where 20000 numbers of particles were simulated with 200 cycles and 35 of them being skipped. Only one material was depleted and six time steps were simulated. The calculation took 30.6 days with single core on windows system.

Table 3 shows the main actinide composition in a fuel pebble after achieving maximum burn up. Assuming each pebble generates 0.3704 kW on average, they will reach the maximum burn up after 1080 days operation. Of 0.85 g U-235 in the fresh fuel, only 0.1352 g U-235 which is 16 % is spent to produce energy 0.4 MWd. However, the fission energy is also generated by Pu-239 fissions which come from U-238 transmutation. Of 4.15 g U-238 in the fresh fuel, 0.012 g is transmutted, mostly into Pu-239. The pebble produced 7.673×10-3 g per pebble at maximum burn up. The actinide products at 80,000 MWd/t contain 10.15 Ci.

TABEL 3. ACTINIDE COMPOSITION OF SPENT FUEL AT MAXIMUM BURN UP OF 80,000 MWD/T

No.	Isotope	Mass (g)	Activity (Ci)
1	U-238	4.138E+00	1.391E-06
2	U-235	7.148E-01	1.545E-06
3	U-236	2.154E-02	1.393E-06
4	Pu-239	7.673E-03	4.760E-04
5	Pu-240	1.541E-03	3.496E-04
6	Pu-241	1.011E-04	1.045E-02
7	Np-237	4.687E-05	3.303E-08
8	Np-239	2.178E-05	5.051E+00
9	Pu-242	5.547E-06	2.194E-08

10	Pu-238	1.683E-06	2.883E-05
11	U-237	4.874E-07	3.977E-02
12	U-239	1.507E-07	5.051E+00
13	U-234	1.161E-07	7.219E-10
14	Np-238	7.098E-09	1.839E-03
15	Th-232	1.672E-09	1.833E-16
16	Th-234	6.010E-11	1.391E-06
	totals	4.884E+00	1.015E+01

Table 4 shows non actinides product per pebble at burn up level of 80,000 MWd/t from high to low activity. Non actinides products contain 47.82 Ci, where Xe-133 emits the highest activity of 3.098 Ci. Totally both actinides and non actinides make up 57.97 Ci at the time of shutdown.

TABLE 4. FISSION PRODUCTS COMPOSITION PER PEBBLE AT BURN UP 80,000 MWD/T

No.	Isotopes	Mass (g)	Activity(Ci)	No.	Isotopes	Mass (g)	Activity(Ci)
1	Xe-133	1.653E-05	3.098E+00	29	Kr-85	9.588E-05	3.766E-02
2	Nb-95	7.559E-05	2.974E+00	30	Eu-156	3.936E-07	2.170E-02
3	Zr-95	1.379E-04	2.962E+00	31	Cs-134	1.528E-05	1.978E-02
4	I-135	8.157E-07	2.884E+00	32	Pm-148	1.116E-07	1.835E-02
5	La-140	5.076E-06	2.823E+00	33	Sb-125	1.206E-05	1.264E-02
6	Ba-140	3.842E-05	2.812E+00	34	Cs-136	1.429E-07	1.043E-02
7	Mo-99	5.747E-06	2.761E+00	35	Ag-111	5.971E-08	9.437E-03
8	Pr-143	3.987E-05	2.685E+00	36	Pr-142	7.897E-09	9.120E-03
9	Ce-143	4.034E-06	2.681E+00	37	Sn-125	4.888E-08	5.300E-03
10	Ce-141	9.334E-05	2.661E+00	38	Eu-157	2.893E-09	3.805E-03
11	Y-91	1.069E-04	2.625E+00	39	Eu-155	7.576E-06	3.736E-03
12	Ce-144	7.940E-04	2.529E+00	40	Sm-151	1.371E-04	3.609E-03
13	Xe-135	9.932E-07	2.524E+00	41	I-130	1.070E-09	2.088E-03
14	Sr-89	7.368E-05	2.141E+00	42	Eu-154	4.149E-06	1.122E-03
15	Te-132	6.582E-06	2.000E+00	43	Sn-123	8.881E-08	7.302E-04
16	Ru-103	4.320E-05	1.396E+00	44	Sb-126	5.304E-09	4.436E-04
17	I-131	1.088E-05	1.350E+00	45	Eu-152	8.509E-07	1.502E-04
18	Nd-147	1.249E-05	1.011E+00	46	Rb-86	1.227E-09	1.001E-04
19	Pm-147	8.311E-04	7.709E-01	47	Tc-99	2.930E-03	5.016E-05
20	Pm-149	1.261E-06	4.999E-01	48	Gd-153	1.069E-08	3.779E-05
21	Ru-105	7.210E-08	4.851E-01	49	Sb-124	1.230E-09	2.152E-05
22	Rh-105	5.640E-07	4.764E-01	50	Zr-93	2.876E-03	7.232E-06
23	Cs-137	3.937E-03	3.427E-01	51	Tb-160	6.011E-10	6.788E-06
24	Y-90	6.163E-07	3.351E-01	52	Cs-135	3.643E-03	4.198E-06
25	Sr-90	2.371E-03	3.349E-01	53	Se-79	1.718E-05	2.358E-06
26	Ru-106	6.418E-05	2.129E-01	54	Sn-126	3.682E-05	1.046E-06
27	Pm-151	2.660E-07	1.945E-01	55	I-129	3.554E-04	6.279E-08
28	Sm-153	2.130E-07	9.432E-02	56	Pd-107	1.006E-04	5.177E-08

Figure 2 shows radioactivity decay after shutdown in a fuel pebble after achieving maximum burn up of 80,000 MWd/t. At the time of shutdown, the total activity of the pebble is 57.97 Ci, where non actinides contribute 83 %. Non actinide isotopes mostly come as the result of fission reactions. One hour after shutdown the pebble activity decays by 7.8 % of its origin. After 1 day and 100 days, the total activities remain 45.07 Ci (75.9 %) and 8.25 Ci (13.9 %), respectively.

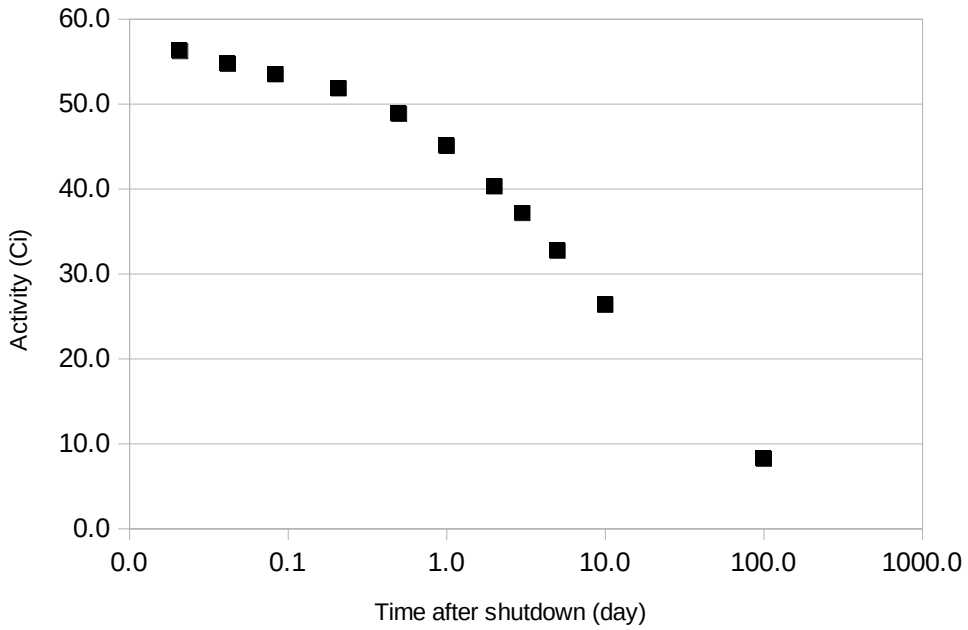
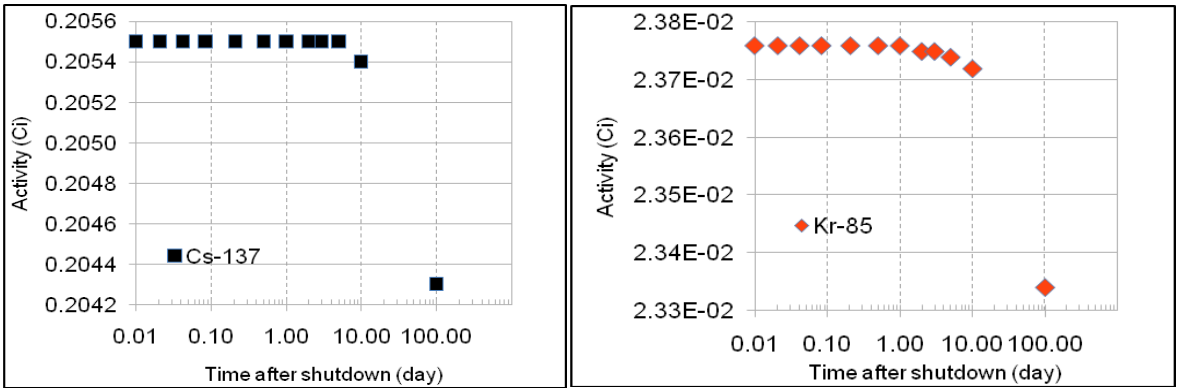


FIG. 2. Pebble activity after shutdown.

Figure 3 show the evolution of important fission products, such as Cs-137, Kr-85, I-135, I-131 and Sr-90 during 100 days after shutdown. Eventhough there is no production term from neutron reaction, the change of fission product concentration comes from their decay and decay of other isotopes producing those isotopes.



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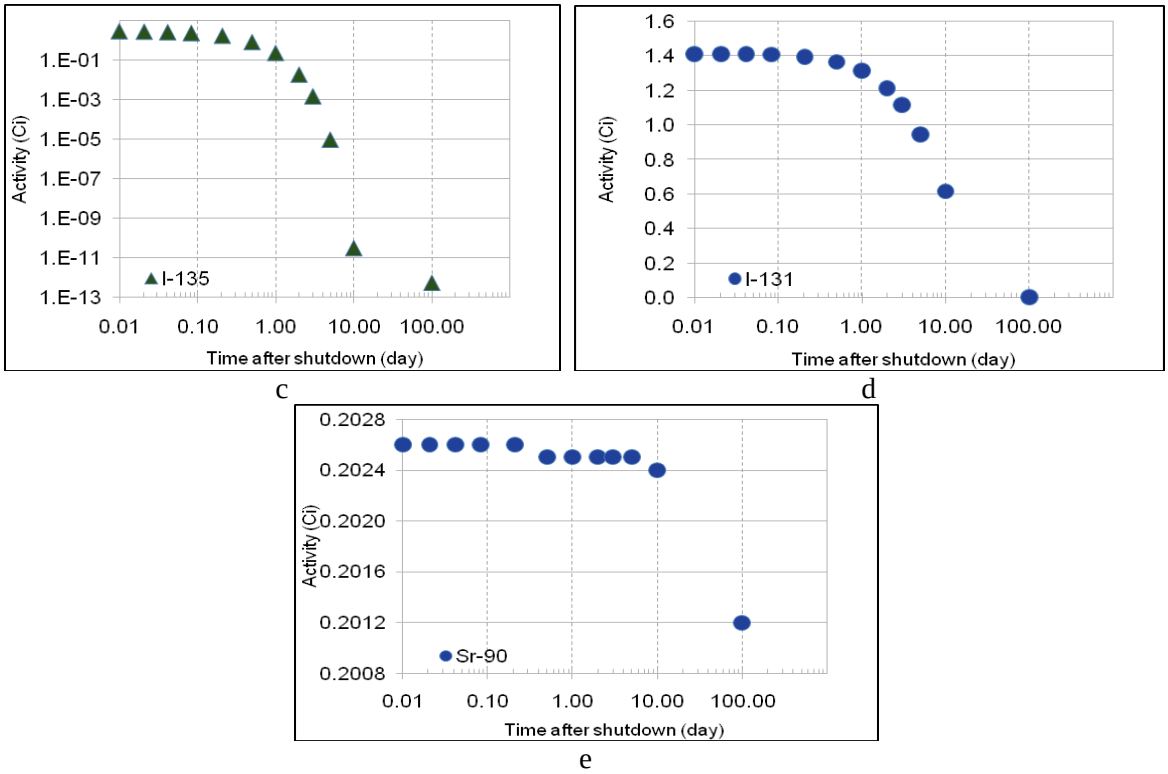

FIG. 3. The evolution of important fission product 100 days after shutdown.

4. CONCLUSSIONS

Burn up calculations of pebble fuel have been conducted using MCNP6 code employing infinite square lattice representing a pebble fuel in the middle of the core exposed by average neutron energy spectrum of the core. After achieving burn up of 80,000 MWd/t, only 0.1352 g U-235 (16 %) is spent to produce energy 0.4 MWd, and 7.673×10-3 g of Pu-239 as the result of U-238 transmutation. The fuel produced radioactivity of 57.97 Ci, where 83% of them comes from non actinide products.

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