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The Analysis of Neutron and Gamma Sources in Spent PWR Fuels

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1. Introduction

The spent nuclear fuel is burned during the planned cycle in the plant and then generates elements such as actinide series elements, fission products, and plutonium with a long half-life. An 'interim storage' step is needed to manage the high radioactivity and heat emitted by nuclides until permanent-disposal or reprocessing of spent nuclear fuel is legislated by government. In the case of Korea, there is no space to dispose of high-level radioactive waste after use, so there is a need for a period of time using interim storage. Therefore, the intensity of neutrons and gamma-ray must be determined to ensure the integrity of spent nuclear fuel during interim storage. In particular, the most important thing in spent nuclear fuel is burnup evaluation, estimation of the source term of neutrons and gamma-ray is regarded as a reference measurement of the burnup evaluation. In this study, an analysis of spent nuclear fuel was conducted by setting up a virtual fuel burnup case based on CE16x16 fuel to check the total amount and spectrum of neutron, gamma radiation produced. The correlation between BU (burnup), IE (enrichment), and CT (cooling time) will be identified through spent nuclear fuel burnup calculation. In addition, the composition of nuclide inventory, actinide and fission products can be identified.

2. Methods and Results

In this study, an analysis of spent fuel will be conducted by setting up a virtual fuel burnup case based on CE16x16 fuel to determine the amount and spectrum of neutron and gamma source term produced. The nuclide library of CE16x16 fuel basically used the template provided in SCALE 6.1

2.1 Nuclear Spent Fuel Characteristics Data in Korea

According to KHNP's SF burnup data [2], for 19,277 bundles of spent nuclear fuel generated by the end of 2019's at 20 domestic PWR nuclear power plants in operation. It can be seen that the distribution of spent nuclear fuel with a enrichment of 3.0 to 4.5 wt% and burnup of 30,000 to 50,000 MWD/MTU occurs the most.

Table 1. Nuclear Spent Fuel Generation in Korea (2019)

Burnup (MWD/MTU)	Generation (bundle)	Ratio (%)	Cumulative generation (bundle)
0~10,000	6	0.03 %	6
10,000~20,000	1,300	6.74 %	1,306
20,000~30,000	1,527	7.92 %	2,833
30,000~40,000	**4,726**	**24.52 %**	7,559
40,000~45,000	**6,432**	**33.37%**	13,991
45,000~50,000	**4,170**	**21.63 %**	18,161
50,000~60,000	1,116	5.79 %	19,277
total	19,277	100 %	-

Based on the correlation between Burnup and Enrichment, the collected spent fuel data can be confirmed through the graph below.

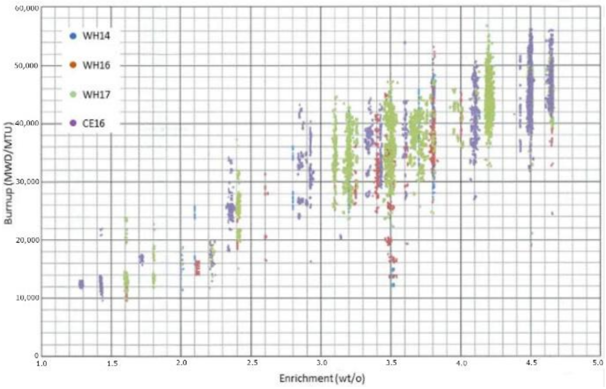

Fig. 1. Nuclear Spent Fuel Generation in Korea (2019)

2.2 Standard Case

In order to specify a standard case that can represent data of figure 1, a linear relational expression that can represent the data is obtained and shown in the graph.

$$BU = 9\,IE + 6 \quad (1)$$

IE: Enrichment (%)
BU: Burnup (GWd/MTU)

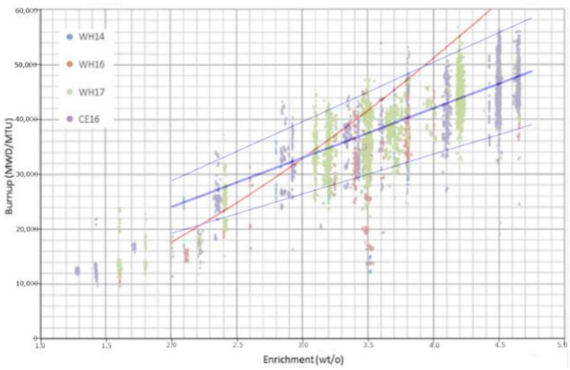

Fig. 2. Nuclear Spent Fuel Generation trend line:
Korea Standard Case (blue line) and Swedish Case (red line)

Based on the Standard case(blue bold trend line) of spent nuclear fuel generated in Korea, the trend of overseas PWR assembly generation (red bold trend line) was additionally confirmed.(Fig. 2 below) When substituting the relational expression of Sweden's PWR spent nuclear fuel assembly [1], it was confirmed that the trend was slightly out of line with the degree of domestic generation. Therefore, in order to reflect the trend of overseas cases as a reference, the upper and lower trends, which are plus-minus 20% of domestic standard cases, were added. (blue trend line)

Here, five points of a specific nuclear fuel case (with a specific BU and IE) placed on the linear relational equation (1) were selected to be compared with other cases that were set up. Data following relational equation (1) was selected at regular intervals within an area of 3.1wt% to 4.65wt% where data was significantly distributed. Therefore, 3.1, 3.5, 3.8, 4.2, 4.65 wt% of enrichment point was selected. (Table 2 below)

Table 2. Nuclear Spent Fuel Generation in Korea:

Burn up (GWd/MTU)	33.9	37.5	40.2	43.8	47.85
Enrichment (%)	3.1	3.5	3.8	4.2	4.65

Standard Burnup Case (2019)

Burnup history was set on the trend line, that is, the burnup corresponding to the five points of the Standard case, which was irradiated by the power plant for 16 months and decayed for 50 days. The spectrum and generation of neutron and gamma sources were calculated using the ORIGEN ARP code through the following plant operation history.

2.3 Case Ⅰ: IE Effect Case(Less and Excess)

The following should be identified in IE effect. After fixing 40200 MWd/MTU, which is 3.8 wt% burnup in the domestic standard case, neutrons and gamma sources were calculated by dividing the less case with a low enrichment of nuclear fuel to 3.1 wt% and an excess case with a high enrichment of 4.65 wt%. Therefore, it was observed whether there was a significant difference in value by setting the enrichment differently under the same burnup condition.

Table 3. IE Effect Case (Less/STD/Excess)

BU (GWd/MTU)	33.9	37.5	**40.2**	43.8	47.85
IE (%)	**3.1 (Less case)**	3.5	**3.8 (STD)**	4.2	**4.65 (Excess case)**

2.4 Case Ⅱ: Power(Cycle) Effect(Less and Excess)

The following should be identified in power(cycle) effect. Among standard cases, 3.8 wt% of IE (enrichment) and 40200 MWd/MTU of BU (burnup) are set as comparison criteria. The standard case was set to 6 months of irradiation and 50 days of decay. For the less case, the operating days in the standard case was reduced, and the burnup history was set to 12 months (360 days) and 50 days (decay). The excess case was set by increasing the operating days to 18 months (540 days) and 60 days of decay. Neutrons and gamma sources were calculated by dividing into less case and excess case in the same way as the IE effect of 2.4. It was observed whether there was a significant difference in values by setting the number of burnup history differently under the same fuel conditions.

Table 4. Power (Cycle) Effect Case

-	Less	STD	Excess
Irradation	12M (360days)	16M (540days)	18M (540days)
Decay	50days	50days	60days

2.5 Case Ⅲ: Impurity Effect

The Impurity case identifies the tendency of neutrons and gamma sources for IE 3.8 wt% case of standard case. The non-actinide impurity composition estimated to exist in light water reactors was additionally set by referring to ORNL's technical report, ORIGEN TM-6051. Through comparison with the standard case without impurities, it was confirmed how much the values of neutrons and gamma source would change.

3.1 Standard Case

In the standard case, the burnup was set as shown in Table 2 by holding five points of the Korea standard trend line. Based on 1 ton of uranium, it was confirmed how many neutrons and gamma sources were generated according to the five burnup points. In order to confirm the overall tendency, the log-scale graph is displayed in the upper right corner, and the main graph is set to a linear scale to make the difference easier to see. (Fig. 3, 4 below)

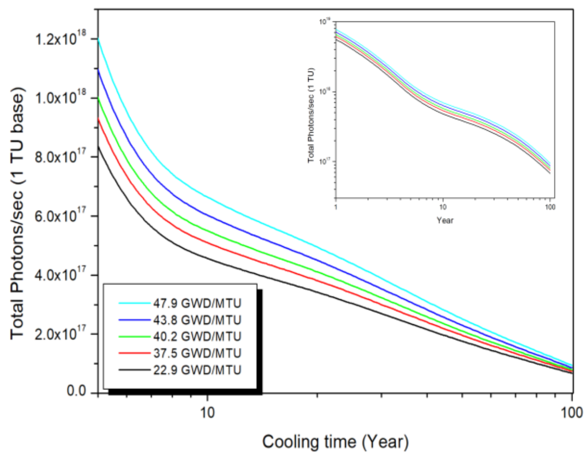

Fig. 3. Total neutron production with respect to Burnup, Enrichment and Cooling time (1TU Uranium)

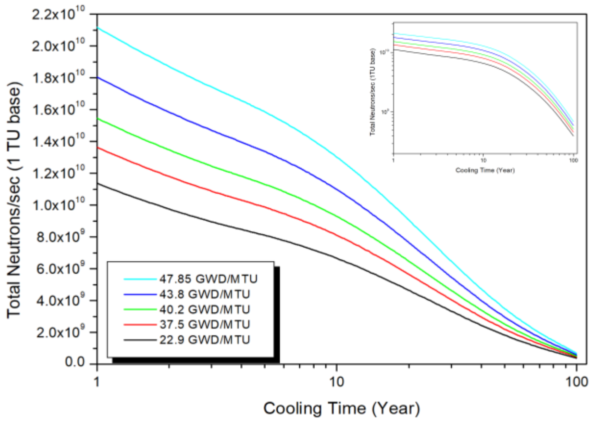

Fig. 4. Total photon production with respect to Burnup, Enrichment and Cooling time (1TU Uranium)

Ideal results were derived according to the correlation between general burnup and initial enrichment, and it was confirmed that neutrons and gamma nuclides tend to decrease as the burnup and enrichment decrease.
In addition, it was confirmed that when the spent nuclear fuel was cooled from 1 to 100 years, the amount of neutron source decreased by about 4.76 % and about 8.3% on a gamma source.

Next, the changes in the spectra of neutrons and gamma according to the cooling time were confirmed by focusing on 40.2GWd/MTU burnup, which is the midpoint in the standard case. The cooling time was set to five variables of 5, 10, 20, 50, and 100 years. Starting with 5 years, the minimum year for fuel withdrawal, 10 years and 20 years were selected. Also, the result of 50 years was confirmed in consideration of the maximum cooling time (40yr basis on 2019's data) discharged from Kori unit 1, the first reactor operated in Korea. Finally, the spectrum during 100-year cooling were compared.

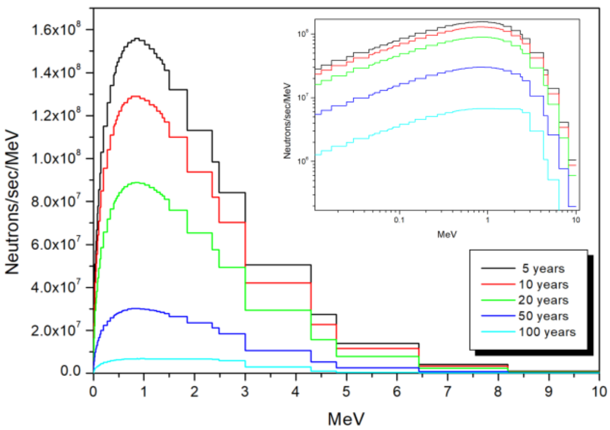

Fig. 5. Neutron Spectrum with respect to Cooling time (BU= 40.2 GWd/MTU, IE= 3.8 %)

Figures 5 and 6 respectively show that the neutron and gamma spectra rapidly decrease as the cooling time increases. It can be seen that the neutron spectrum gradually decreases to almost the same shape. (Fig.5 above) After 100 years of cooling, the neutron spectrum was reduced to 6 orders of 10.

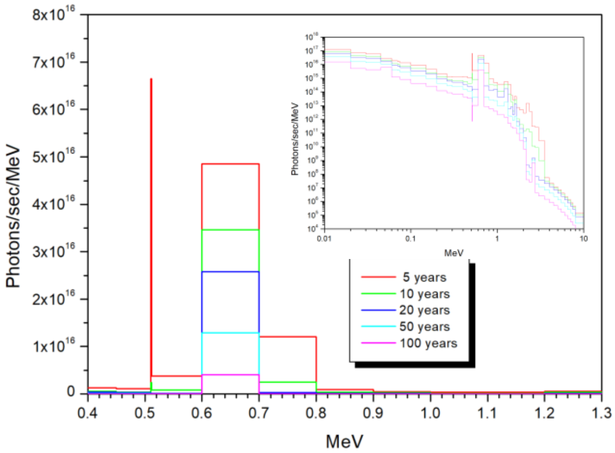

Fig. 6. Gamma radiation Spectrum with respect to Cooling time (BU= 40.2 GWd/MTU, IE= 3.8 %)

In the case of gamma spectrum, there is a noticeable single emission peak. When the energy area is enlarged, it appears to be 0.512 MeV, which is mainly verified by the data with a cooling period of 5 years.
It can be seen that when the cooling period is not long to 5 years, a stationary positron that have lost kinetic energy collides with an electron and emits annihilation radiation divided into two 0.511 MeV energy.

3.2 Case Ⅰ: IE Effect Case(Less and Excess)

The following is a comparison between a low enrichment fuel of 3.1 wt% and a high enrichment fuel of 4.65 wt% in a fixed state of 40.2 GWd/MTU, which is a burnup of the standard case.

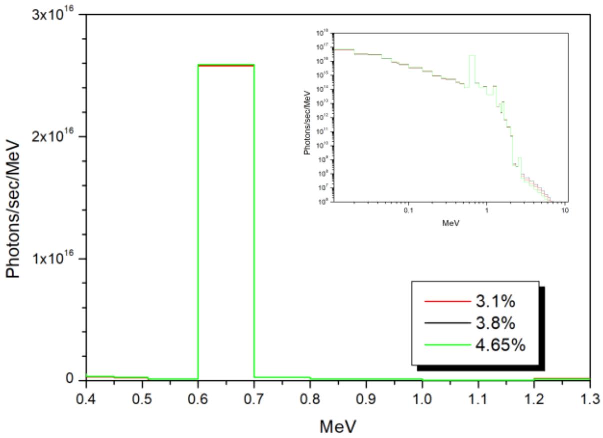

Fig. 7. Gamma Radiation Spectrum with respect to Initial Enrichment (BU= 40.2 GWd/MTU, CT= 20 yr)

The difference in gamma spectrum according to enrichment does not appear to be significant. However, it was confirmed that photon tends to occur in the order of 3.8, 3.1, and 4.65 wt% at energy of 2 MeV or higher. Spectrum peaks are found between 0.6 ~ 0.7 MeV.

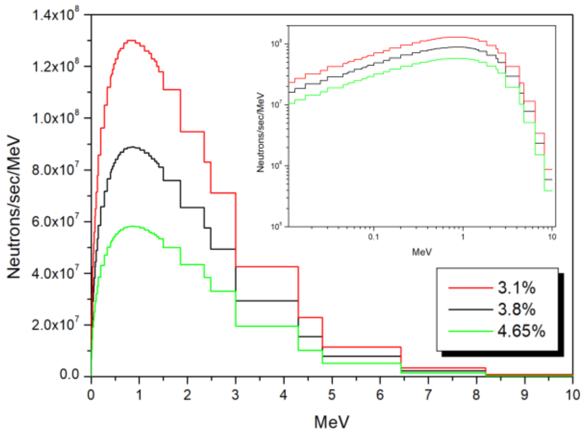

Fig. 8. Neutron Spectrum with respect to Operation Period (BU= 40.2 GWd/MTU, CT= 20 yr)

In the case of the neutral spectrum, the patterns of the spectrum differ depending on the enrichment. The higher fuel of enrichment is burned, the less neutrons are produced. The reason for this phenomenon is that when fuel is burned at the same burnup, the amount of actinium produced in high enrichments of fuel will be less. After cycle 3 down in the output file, the generated nuclide and nuclide concentration (curies) were checked, as the amount of Pu increased, less neutron sources were generated. Therefore, it can be seen that the decrease in the minor actinide series, including Pu, resulted in a decrease in neutrons.

3.3 Case Ⅱ: Power(Cycle) Effect

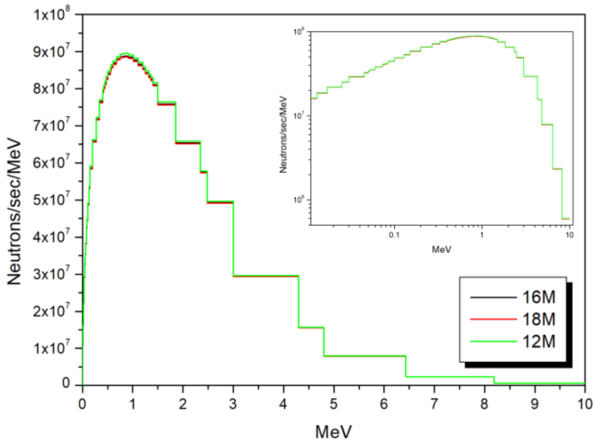

Fig. 9. Neutron Spectrum with respect to Operation Period (BU= 40.2 GWd/MTU, CT= 20 yr)

In the third case, fuel with the same BU and IE as in the standard case was set, but the operating years of the NPP were increased(18 month) and decreased(12 month), and the spectrum tendency was identified. The results of the neutron spectrum according to the operation period do not show much difference, but the shorter the operation period, the smaller the number of neutrons emitted. (Fig.9 above) This is because the irradiation increases because the power density per cycle increases for a short operation period.

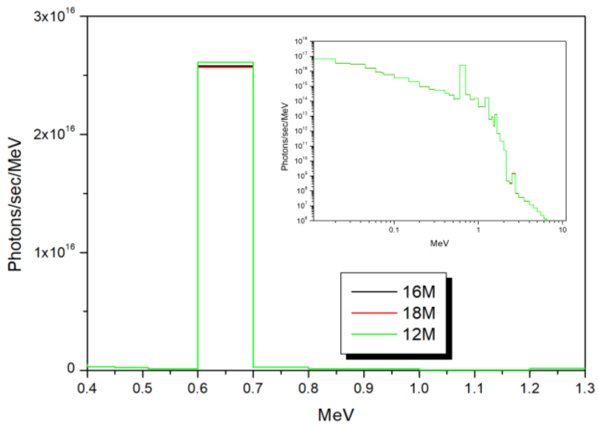

Fig. 10. Gamma Radiation Spectrum with respect to Operation Period (BU= 40.2 GWd/MTU, CT= 20 yr)

Similarly, for gamma-ray spectra in figure. 10, the spectrum is high for a short period of operation. In addition, the shorter the operating period, the less time the nuclide will decay, resulting in more photons.

3.4 Case Ⅲ: Impurity Effect

In the fourth case, referring to the data from ORNL, impurities were added. When compared with the standard case, the difference in amount was extremely small, so there was no meaning for the comparison.

3. Conclusions

In this study, based on the standard case, a burnup history was set according to enrichment, operating period, and presence of impurity. Through this, the correlation of the tendencies shown by the spectra of neutron and gamma sources to confirm fuel integrity during interim storage of spent nuclear fuel was identified. In future studies, the trend of values will be confirmed and compared by applying burnup history and nuclear fuel parameters of other power plants by expanding from CE's results.

ACKNOWLEDGEMENT

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