Study the Effect of Some Parameters on [Cs-134/Cs-137] Activity Ratio for Nuclear Spent Fuel Using ORIGEN-ARP Code

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Abstract: Safeguard state system is engaged in Non Destructive Assay (NDA) Research and Development (R&D) work on spent fuel elements (SFE). One of the traditional methods for the verification of SFE's is the so-called gamma spectrometry, relying on gamma measurements for the assessment of burnup SFE. For better understanding of the principles behind Cs-134/Cs-137 activity ratio, significant efforts were done in terms of both Monte Carlo modeling and depletion & evolution code calculations. However, this study focuses on the impact of some irradiation history parameters on the Cs-134/Cs-137 activity ratio in SFE, where ORIGEN-ARP code was used to determine such activity ratio. The cases considered in this study are some parameters of Low Enriched Uranium (LEU) of 17×17 PWR fuel with an initial enrichment of 4.5%. The burnup ranged between 10 and 60 GWd/tu, as well as 18 values of cooling time, from 0 up to 30 years. The varied parameters were the average power level, the cooling time between cycles and the duration of an irradiation cycle. Conclusions about the role of the considered irradiation history parameters on the Cs-134/Cs-137 activity ratio were drawn and the obtained values were discussed.

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1. Introduction

It is important that a state safeguard system should verify the reactor burnup (BU) and reactor power history. Gamma-ray spectroscopy is a nondestructive method that can be used to measure the fission products of irradiated fuel elements [1]. Gamma measurement is a good tool for such verification. In the process of selecting the measuring device for verifying an irradiated fuel, there are some factors should be considered. These factors include the fuel type, percentage of BU and cooling time (CT). High purity germanium spectrometers are used, when such measurement have to be performed. Also, CdTe and CdZnTe probes have been extensively used particularly for safeguard purposes [2]. BU could be measured indirectly through the direct measurement of some correlated fuel parameters. This could be achieved via detecting the radiation emissions from radionuclides built up during irradiation such as Cs-134, Cs-137 and/or gamma energies [3, 4]. A lot of work has been done to verify the BU by either the direct measurement of Cs-137, or the ratio Cs-134/Cs-137, from their gamma energies [1-13]. ORIGEN-ARP (a part of the SCALE 6 package) [5], represents an important tool to calculate some quantities that can help in the preparation for the verification process.

However, this work aims to investigate the impact of some parameters, such as the average power (AP), cooling time between two complete irradiation cycles (CTIC) and the duration of an irradiation cycle (DIC) on the activity ratio of Cs-134/Cs-137.

2. Structure of the work

Three groups of simulations were performed and investigated using ORIGEN-ARP. Six BU values (from 10 to 60 in steps of 10 GWd/tu) were used in each group of these simulations for a Westinghouse 17x17 Power Water Reactor (PWR), with UO_2 fuel of an initial enrichment (IE) of 4.5%. Eighteen values of CT from discharge up to 30 year were considered. It can be referred to references [3, 6], for detailed description of the parameters that were kept fixed for all simulations. The varied parameters were the AP, DIC and CTIC. The number of irradiation cycles and the duration of the final irradiation cycle were determined by the final BU value according to the selection of AP and DIC values.

The first group of simulations were performed to study the impact of the AP on the Cs-134/Cs-137 activity ratio, where three average power values (30, 40, and 50 MW/tu) were used with 360 days as DIC and 30 days of CTIC [7, 8]. The activity ratio for an AP of 40 MW/tu was used as a reference to compare the activity ratio obtained with other AP values.

The second group of simulations was performed to study the impact of DIC on the Cs-134/Cs-137 activity ratio. Three values (270, 360 and 420 days) we selected as DIC while the AP was set as 40 MW/tU and the CTIC was set as 30 days for all simulations. The activity ratio of 360 days irradiation cycle was used as a reference to compare the obtained results for different irradiation cycles.

The third group of simulations was performed to study the effect of changing the CTIC on the Cs-134/Cs-137 activity ratio. Three CTIC values (30, 60 and 90 days) were used and the AP was set as 40 MW/tu for all simulations. CTIC of 30 days was used as a reference to compare the total neutron emissions calculated with different CTIC.

3. Results and Discussion

A large amount of data was generated in the obtained output files (54 output files) for each simulation. These data include the isotopic composition of each nuclide both during irradiation and cooling as well as the mass concentration in grams for both Cs-134 and Cs-137. Several scripts that run in a bash shell environment were developed for handling the generated files and to extract the relevant information. The half-life time and specific activity of Cesium isotopes (134 & 137) that was used for the extraction of the Activity of Cesium isotopes per Curie, are presented in Table (1) [14].

Table 1. Specific Activities of Cesium Isotopes

Isotope	Half Life time	Specific Activity Ci/gm
Cs-134	2.1 yr	1300
Cs-137	30 yr	88

The first group of simulations shows that, the impact of the AP on the Cs-134/ Cs-137 Activity Ratio was not significant at relatively low BU values, since the irradiation cycle is almost one or two cycles only. So the decay of Cs-134 and Cs-137 is not a considerable factor in the comparison at different AP levels. Table (2) shows that at BU value of 10 GWd/tu, almost no impact has observed on the Cs-134/Cs-137 activity ratio for different AP values.

Table 2. The Activity Ratio of (Cs-134/Cs-137) at different AP values and BU=10 GWd/tu

Cooling Time (days)	Activity ratio at AP 30 MW/tu	Activity ratio at AP 40 MW/tu	Activity ratio at AP 50 MW/tu	Activity ratio at AP 30 MW/tu, normalized to Activity ratio AP 40 MW/tu	Activity ratio at AP 40 MW/tu normalized to Activity ratio AP 40 MW/tu
0	0.41495	0.4171	0.41526	0.99485	0.99558
1	0.41462	0.41677	0.41496	0.99484	0.99565
3	0.41393	0.41604	0.41424	0.99492	0.99566
10	0.41146	0.41358	0.41178	0.99487	0.99564
30	0.40445	0.40652	0.40475	0.99492	0.99565
100	0.3809	0.38286	0.38119	0.99487	0.99564
300	0.32089	0.32255	0.32114	0.99486	0.99564
365	0.30349	0.30505	0.30373	0.99489	0.99565
730	0.22192	0.22305	0.22208	0.9949	0.99564
1095	0.16226	0.1631	0.16239	0.99488	0.99567
1460	0.11864	0.11926	0.11873	0.99485	0.9956
1825	0.08675	0.0872	0.08682	0.99487	0.99563
2190	0.06343	0.06376	0.06348	0.99491	0.99562
2555	0.04638	0.04662	0.04642	0.99484	0.99565
2920	0.03391	0.03409	0.03394	0.99488	0.99565
3285	0.0248	0.02493	0.02482	0.99485	0.99562
3650	0.01813	0.01823	0.01815	0.99486	0.99566
7300	7.92E-04	7.96E-04	7.93E-04	0.99484	0.99563
10950	3.46E-05	3.48E-05	3.46E-05	0.99487	0.99565

For relatively higher BU values, the activity ratio is affected at lower values of AP and there is a deviation in the ratio almost up to 6.5%. This effect decreases with increasing the AP value to 2.5% at 50 MW/tu. This was explained to be due to the half-life time of Cs-134 which is 2.1 year, and at low AP values more irradiation cycles are used, so the build-up of Cs-134 is affected where it starts to decay within the irradiation cycles, since Cs-137 has no significant effect with changing the AP. Figs. (1, 2) show the impact of the AP on the activity of Cs-134 and Cs-137 at different BU and AP values which was found in complete agreement with the results obtained by Thomas [15], where he showed that the decay of Cs-134 is the factor that affects the ratio Cs-134/Cs-137 and consequently the determination of BU from this ratio needs a correction for the decay time in such calculations.

The second group of simulations investigates the impact of DIC on the Cs-134/ Cs-137 Activity Ratio. At low BU values (10 and 20 GWd/tu), it is clear that no changes are observed, since the same days of irradiation have spent for all simulations. For relatively high BU values (30 and 40 GWd/t_u), the impact of DIC on the Activity Ratio of Cs-134/ Cs-137 starts from 8% and increased up to 42%, due to the huge changes in the Activity of Cs-134. Table (3) shows that at BU 30 GWd/tu there is a high impact on the ratio Cs-134/Cs-137 for different DIC values which was found in agreement with ref. [15].

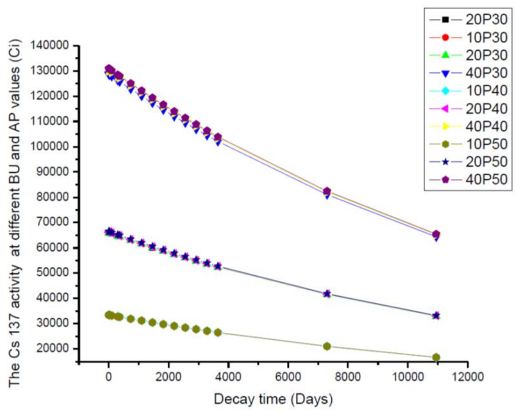

Figure 1. The Activity of Cs-137 in Ci at different BU & AP values

At relatively high BU values (50 & 60 GWd/t_u) the ratio is affected dramatically at DIC 270 days, since the activity of Cs-134 drops almost to zero at the beginning of discharge. But for the other DIC values there is almost no significant change, since the irradiation cycle is relatively large, so the number of complete cycles is reduced and Cs-134 build-up grows faster than its decay. Figure (3) shows the impact of DIC on the Cs-134/Cs-137 activity ratio at BU= 50 GWd/tu.

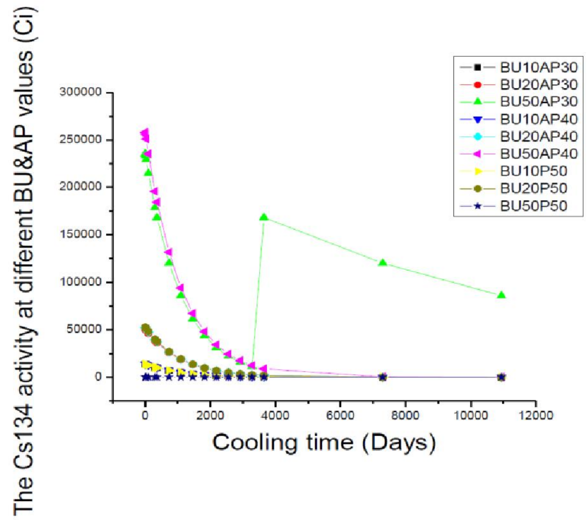

Figure 2. The Activity of Cs-134 in Ci at different BU & AP values

Although the irradiation cycle is changed, the Activity of Cs-137 remains almost constant, due to its relatively high half-life time (30 year), and the activity ratio is affected through Cs-134. Figs. (4, 5) show the impact of DIC on both Cs-134 and Cs-137 activities.

Table 3. The Cs-134/Cs-137 Activity Ratio at different DIC values and BU= 30 GWd/tu

Cooling Time (days)	**Activity ratio at DIC 270**	**Activity ratio at DIC 360**	**Activity ratio at DIC 410**	**Activity ratio at DIC 270 normalized to Activity ratio DIC 360**	**Activity ratio at DIC 410 normalized to Activity ratio DIC 360**
0	1.10834	1.09312	1.11731	1.01392	1.02213
1	1.10738	1.09121	1.11644	1.01481	1.02312
3	1.10554	1.0843	1.11449	1.01959	1.02785
10	1.09893	1.065	1.10783	1.03186	1.04022
30	1.08024	0.9998	1.08898	1.08046	1.0892
100	1.01731	0.83537	1.02563	1.21779	1.22775
300	0.85707	0.79687	0.86406	1.07553	1.08431
365	0.81058	0.5717	0.81718	1.41783	1.42938
730	0.59271	0.41804	0.59751	1.41783	1.42932
1095	0.43339	0.30566	0.43688	1.41788	1.4293
1460	0.31687	0.2235	0.31944	1.41781	1.4293
1825	0.23169	0.16342	0.23359	1.41783	1.42944
2190	0.16941	0.11949	0.1708	1.41779	1.4294
2555	0.12387	0.08737	0.12489	1.41779	1.42936
2920	0.09058	0.06389	0.09131	1.41779	1.42934
3285	0.06623	0.04671	0.06677	1.41781	1.42935
3650	0.04843	0.0478	0.04882	1.013	1.02127
7300	0.00212	0.00209	0.00213	1.01303	1.02129
10950	9.24094E-5	9.12218E-5	9.31621E-5	1.01302	1.02127

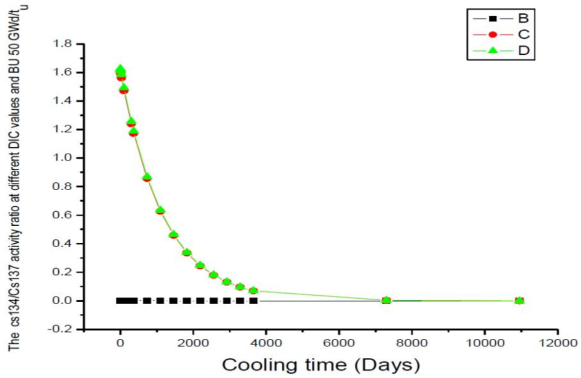

Figure 3. The Cs-134/Cs-137 Activity ratio at different DIC values and Bu= 50 GWd/tu

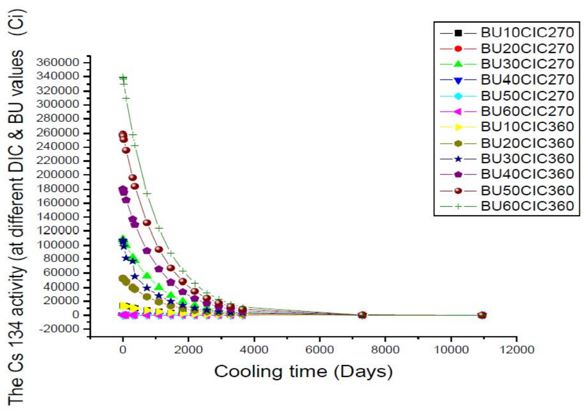

Figure 4. The Activity of Cs-134 in Ci at different BU & DIC values

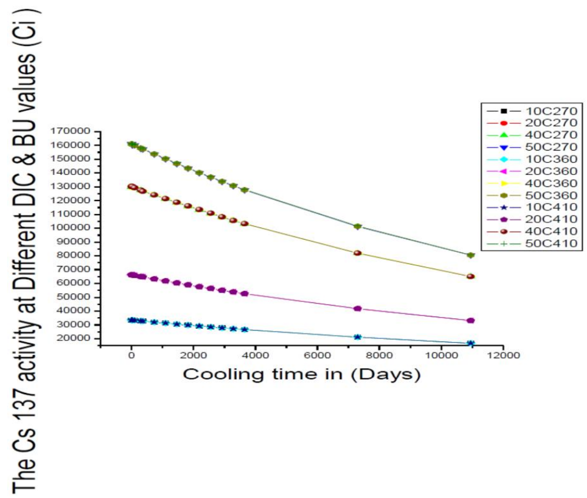

Figure 5. The Activity of Cs-137 in Ci at different BU & DIC values

The third group of simulations shows the impact of CTIC on Cs-134/Cs-137 activity ratio. At low BU values (10 & 20 GWd/tu), almost no change observed. That is, for BU=10 GWd/tu, no change can be observed because it is the same days of irradiation for all simulations. For relatively high BU (30 and 40 GWd/tu) values, the impact of DIC on the Cs-134/Cs-137 activity ratio starts from 3% and increased up to 36% due to the huge change in the activity of Cs-134. Table (4) shows that at BU= 30 GWd/tu, there is high impact on the ratio Cs-134/CS-137 for different CTIC values which is in agreement with ref. [15].

Table 4. The Cs-134/Cs-137 activity ratio at different CTIC values and BU= 30 GWd/t_u

Cooling Time (days)	**Activity ratio at CTIC 60**	**Activity ratio at CTIC 30**	**Activity ratio at CTIC 90**	**Activity ratio at CTIC 60 normalized to Activity ratio at CTIC 30**	**Activity ratio at CTIC 90 normalized to Activity ratio atCTIC 30**
0	1.06607	1.09312	1.03907	0.97525	0.95055
1	1.06524	1.09121	1.03817	0.9762	0.95139
3	1.06337	1.0843	1.03645	0.9807	0.95588
10	1.05701	1.065	1.03026	0.9925	0.96738
30	1.03904	0.9998	1.01274	1.03925	1.01295
100	0.97861	0.83537	0.95377	1.17147	1.14173
300	0.82442	0.79687	0.80351	1.03456	1.00833
365	0.77971	0.5717	0.75996	1.36384	1.32928
730	0.57012	0.41804	0.55566	1.3638	1.3292
1095	0.41686	0.30566	0.40631	1.36379	1.32928
1460	0.30481	0.2235	0.29709	1.36382	1.32927
1825	0.22287	0.16342	0.21722	1.36383	1.32924
2190	0.16296	0.11949	0.15883	1.36377	1.32919
2555	0.11916	0.08737	0.11613	1.36383	1.32919
2920	0.08713	0.06389	0.08492	1.3638	1.32918
3285	0.06371	0.04671	0.06209	1.36382	1.32921
3650	0.04658	0.0478	0.0454	0.97445	0.94971
7300	0.00203	0.00209	0.00198	0.97443	0.9497
10950	8.88906E-5	9.12218E-5	8.66328E-5	0.97445	0.94969

For high BU values the change is slightly noticeable and increased from 2% to 4% as the Cs-134 build-up grows faster than its decay and almost no change in the activity of Cs-137. Figs. (6 & 7) show the activity of Cs-134 at different CTIC values and BU= 50 GWd/tu and the activity of Cs-137 at different CTIC and different BU values respectively.

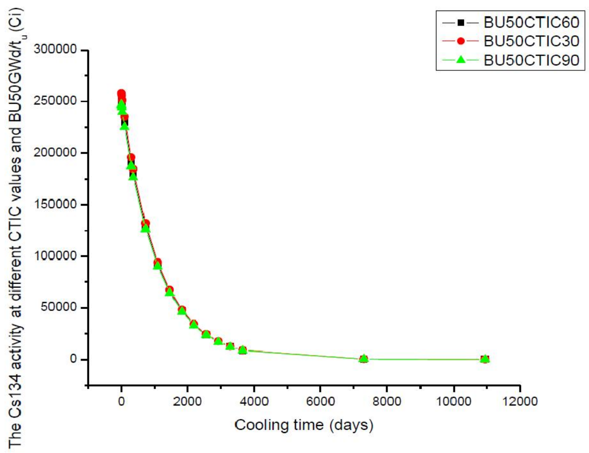

Figure 6. The Cs-134 Activity in Ci at different CTIC and BU= 50 GWd/tu

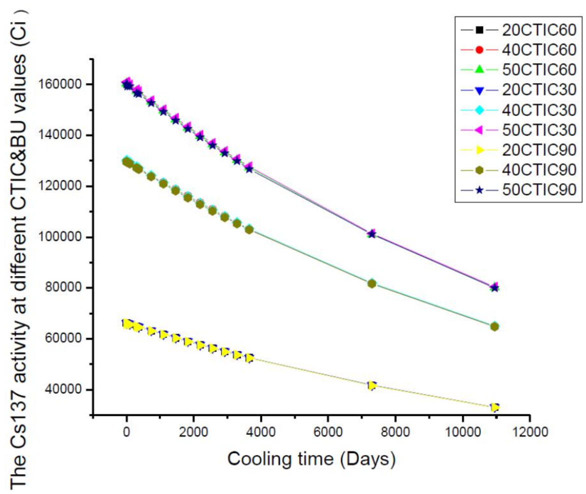

Figure 7. The Cs-137 Activity in Ci at different BU & CTIC values

4. Conclusion

Gamma emission from SFE with different irradiation histories was studied through the activity ratio Cs-134/Cs-137 by using the ORIGEN-ARP code. The considered case is, LEU 17×17 PWR fuel with an initial enrichment of 4.5%. TheBU ranged between 10 and 60 GWd/tu and 18 values of cooling time, from 0 up to 30 years, were also considered. That is, the varied parameters were the AP levels, CTIC and the DIC.

The analysis of the obtained data revealed that the activity ratio of Cs-134/Cs-137 is of little sensitivity to the AP, but of large sensitivity for both the CTIC and the DIC at relatively high BU values i.e. (30 & 40 GWd/tu). The BU values below 30 GWd/tu, were found to be almost of no effect on the activity ratio Cs-134/Cs-137. It was found also that, by varying the considered irradiation history parameters, Cs-134 production changed up to 40%, while the variation in Cs-137 production was limited to less than 2% in most cases. So when analyzing NDA data for Cs-134, the irradiation history parameters should be taken into consideration to correct the obtained data.

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