# Assessment of Plutonium Isotopic Ratio Change for PWR Spent Fuel Relative to Average Power Level Using ORIGEN-ARP Code

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**Abstract:** A study was performed to determine the ratios between some Plutonium isotopes in spent fuel of PWR. These ratios could be used for safeguard applications in a nuclear reactor as forensic information, to determine the origin of an unknown nuclear waste and to verify the activities declared by the operator. Some parameters of the reactor could affect the ratio of plutonium isotopes. One of these parameters is the average power level of the reactor. In this work ORIGEN-ARP from SCALE 6.1 code was used for the assessment of the effect of the change in the average power level on the Plutonium isotopic ratio. The analysis of the obtained results revealed that the ratio of plutonium isotopes is not much affected by the average power level for burnup values above 20 GWd/tu.
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**Keywords:** Nuclear Safeguard Applications; Spent Fuel elements; ORIGEN-ARP Code; Plutonium Isotopic Ratio

## 1. Introduction

The fission products and transuranic production in a PWR depend on some features and physical parameters of the reactor. For example, the type of the reactor fuel, moderator, and overall design. The isotopic profiles of some actinides could be used to determine some of these features. Also the analysis of some fission products may reveal the history of the fuel [1]. From the actinides that present in the fission products, plutonium and americium elements are most effective than other nuclides from the radioecology and radiation safety points of view. Plutonium is represented by many isotopes but the contribution of the isotopes ($^{238}$Pu, $^{239}$Pu, $^{240}$Pu and $^{241}$Pu) is higher than other isotopes to the total Plutonium pollution [2]. The predicted accuracy with the isotopic data could be utilized to form conclusions about history. Many methods, codes and measuring techniques were used in previous work (ICP-MS, MCNPx, ORIGEN-ARP and HPGe), for nuclear safeguards application to predict the behavior of such isotopes [1-3]. The build-up of plutonium isotopes depend on the interplay of neutron reactions causing capture or fission. This build-up changes with the neutron spectrum, which is determined by details about the reactor and fuel design [3]. The importance of Pu isotopic ratios is due to the fact that some factors could be predicted from the determination of such isotopes such as, the correlation between the main reactor types, that has been found by the total $^{238}$Pu content as well as the $^{242}$Pu/$^{240}$Pu ratio. It is known that, the higher uranium enrichment in the fuel affects directly the $^{238}$Pu abundance. Also, the softer the plutonium spectrum, the higher is the ratio of $^{242}$Pu/$^{240}$Pu. So, the level of enrichment can be obtained by knowing the amount of $^{238}$ Pu [4]. ORIGEN-ARP, (part of the SCALE 6.1) package [5], is an important tool to calculate some quantities that would help in the verification process. Examples of such quantities are the total neutron emission and the relative isotopic contribution to the total neutron emission. Previous work was carried out by Rossa et al. [6-8] and Trellue et al [9], to develop spent fuel libraries to be used for the investigation of Non-Destructive Assay (NDA) methods applied to Spent Fuel Elements (SFE).

However, this work aims to study the effect of changing the reactor average power (AP) level on the plutonium isotopic ratio.

## 2. Structure of the work

A group of simulations were performed and investigated using ORIGEN-ARP code. Six burnup (BU) values (from 10 to 60 GWd/tu in steps of 10 GWd/tu) were used in such groups of simulation for a Westinghouse 17x17 PWR, with $UO_2$ fuel of uranium initial enrichment (UIE) of 4.5%. Twenty values of cooling time (CT) from discharge up to 100 year were considered. It can be referred to references [6, 7], for detailed description of the parameters that were kept fixed for all simulations. The varied parameter was the Average Power (AP). The number of irradiation cycles and the duration of the final irradiation cycle were determined by the final BU value according to the AP and DIC chosen. The group of simulations was performed to study the impact of AP on Pu isotopic ratio. Three AP values (30, 40, and 50 MW/$t_u$) were used with 360 days as irradiation cycle and 30 days of cooling time interval between two cycles. All Pu isotopes ($^{238}$Pu, $^{239}$Pu, $^{240}$Pu and $^{241}$Pu) were normalized to $^{239}$Pu as the most prevalent Plutonium Isotope [10].

## 3. Results and Discussion

Large data were obtained during these simulations including the total neutron emission, total gamma emission and isotopic concentration per gram. Eighteen output files were generated and many scripts and programs were used to analyze such files and to extract the relevant information. The following results were obtained for these groups of simulations.

Since $^{239}$Pu is the most prevalent plutonium isotope, so studying the buildup of such isotope with different BU values and different cooling time is important. Fig. (1) Illustrates the relation between the isotope $^{239}$Pu total mass in grams and the cooling time (CT) at different BU values. It is obvious that with increasing the BU value the amount of $^{239}$Pu mass increased, until at higher BU values a slight change is recognized only between (40 and 60 GWd/tu). Therefore, the effect of AP change on the buildup of $^{239}$Pu at the same BU value should be interpreted. Figures (2, 3 and 4) illustrate that at lower AP values (30 MW/tu), the buildup of $^{239}$Pu is slightly changed and no change at other AP values.

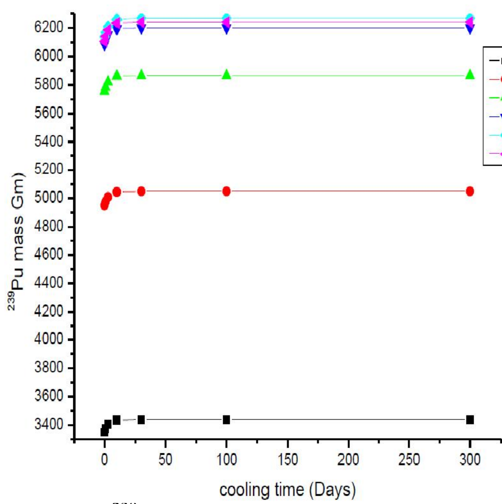


Figure 1. $^{239}$Pu mass content against cooling time at different BU values

The $^{238}$Pu/ $^{239}$Pu Ratio represents an important factor in verifying the initial enrichment [4], so the effect of changing the average power on this ratio was considered. The results of such ratios were normalized to that of AP 40 MW/tu which is the international standard of operating PWR [8].

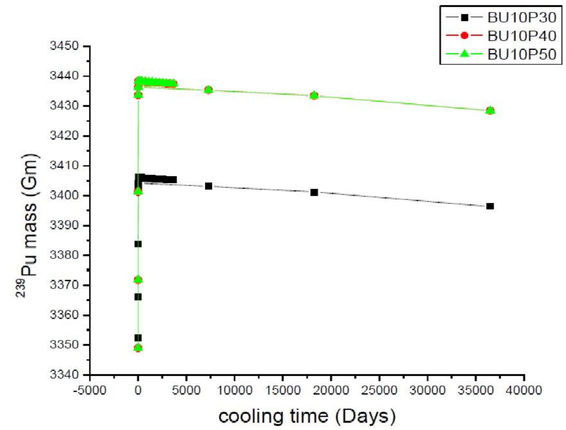


Figure 2. $^{239}$Pu mass content against CT at different AP values for BU=10 MWd/tu

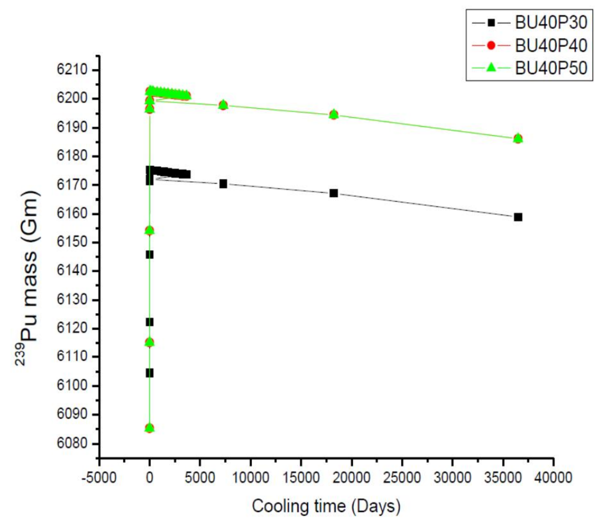


Figure 3. $^{239}$Pu mass content against CT at different AP values for BU=40 MWd/tu

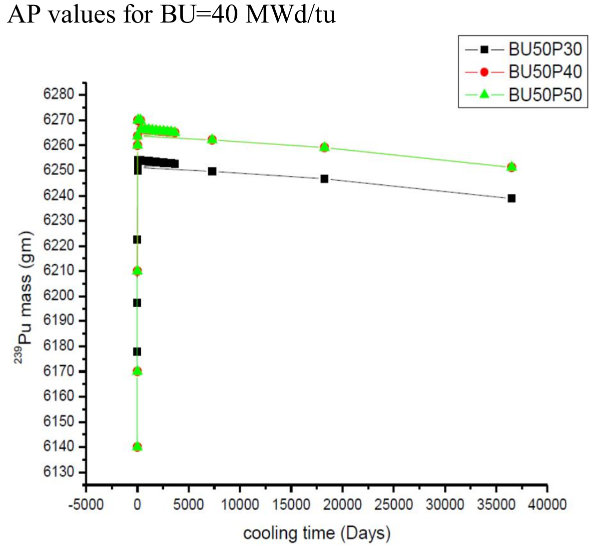


Figure 4. $^{239}$Pu mass content against CT at different AP values for BU 50=MWd/tu

Figures (5, 6) show the difference of the $^{238}Pu/^{239}Pu$ Ratio at different AP and at lower BU values (10, 20 MWd/tu) where it decreased with increasing BU, from around 3% to 2.5% for lower AP values and from 6% to 3% for higher AP values compared to the standard one of AP = 40 MW/tu.

Figures (7, 8) show the difference in the $^{238}Pu/^{239}Pu$ Ratio at different AP and at higher BU values (40, 50 MWd/tu), which is almost around 4-5% for lower AP and around 3-4% for higher AP values. As for the $^{238}Pu/^{239}Pu$ ratio the impact is noticeable at both low and high BU values that are due to the production of $^{238}Pu$ from $^{237}Np$ neutron capture [10]. Since the total neutron emission is affected by changing the AP during 3 years of cooling time [8] so the production of $^{238}Pu$ is affected, and the Table of results is shown in Appendix (1).

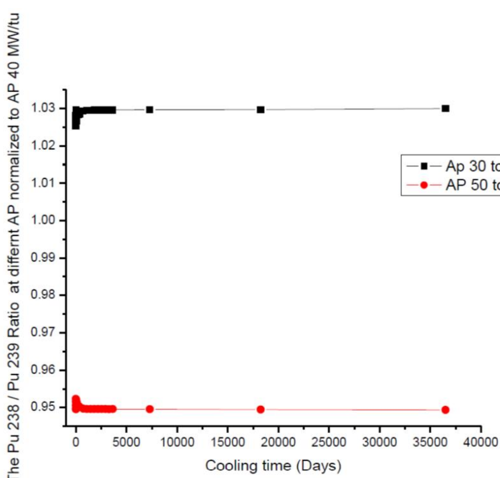


Figure 5. The ratio of $^{238}Pu/^{239}Pu$ against CT at different AP values for BU=10 MWd/tu

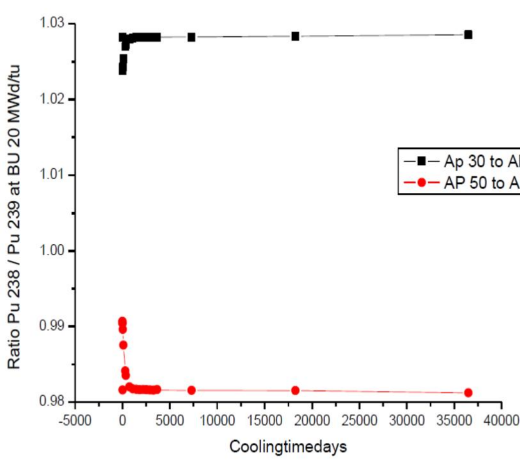


Figure 6. The ratio of $^{238}Pu/^{239}Pu$ against CTat different AP values for BU=20 MWd/tu

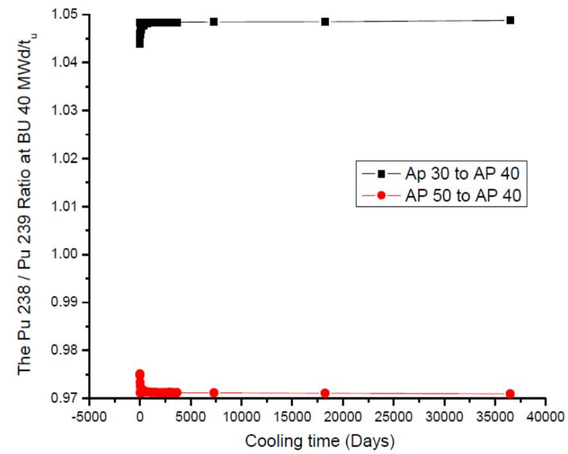


Figure 7. The ratio of $^{238}Pu/^{239}Pu$ against CT at different AP values for BU=40 MWd/tu

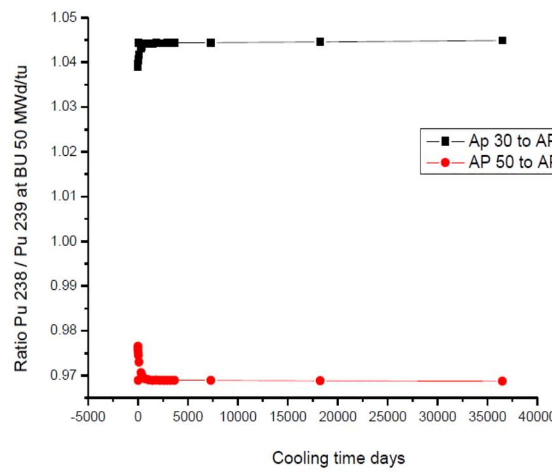


Figure 8. The ratio of $^{238}Pu/^{239}Pu$ against CT at different AP values for BU=50 MWd/tu

The $^{240}Pu/^{239}Pu$ Ratio represents also an important factor to verify the BU values declared by the operator [10]. So, the effect of changing the AP was considered and the results of such ratio were normalized to that of AP=40 MW/tu which is the international standard operation of PWR [8].

Figures (9, 10) show that there is almost no significant change in the $^{240}Pu/^{239}Pu$ ratio at different AP values and at the same BU value. For the $^{240}Pu/^{239}Pu$ ratio, since $^{240}Pu$ comes directly from $^{239}Pu$ [11] and as described before that $^{239}Pu$ is slightly affected by AP change so the $^{240}Pu/^{239}Pu$ ratio has no slightly change and the Table of results is shown in Appendix (1).

The $^{241}Pu/^{239}Pu$ ratio represents an important factor also in the forensics of a nuclear activity [3, 10]. So, the effect of changing the AP on this ratio was considered and the results of such ratios were normalized to that of AP=40 MW/tu which is the international standard operation of PWR [8].

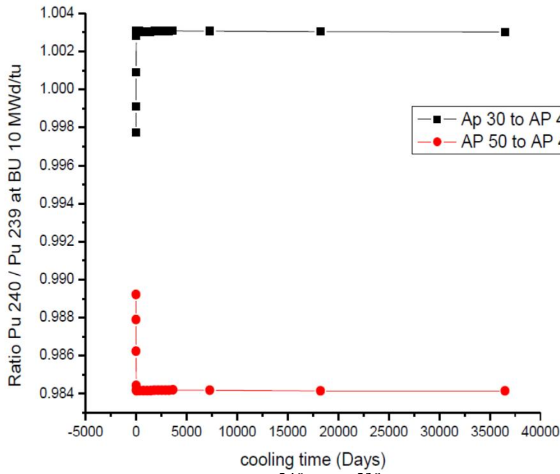


Figure 9. The ratio of $^{240}Pu$/ $^{239}Pu$ against CT at different AP values for BU=10 MWd/tu

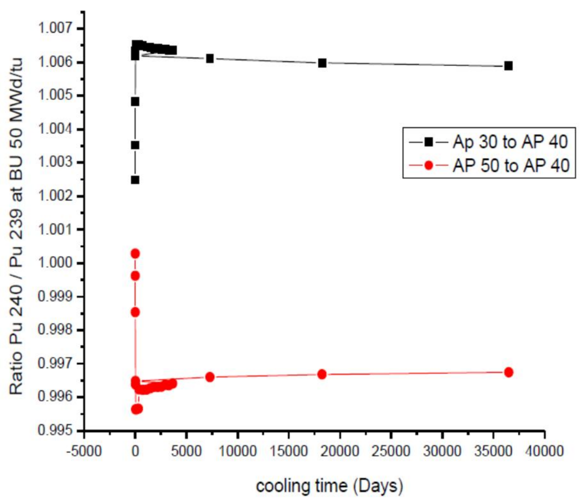


Figure 10. The ratio of $^{240}Pu$/ $^{239}Pu$ against CT at different AP values for BU=50 MWd/tu

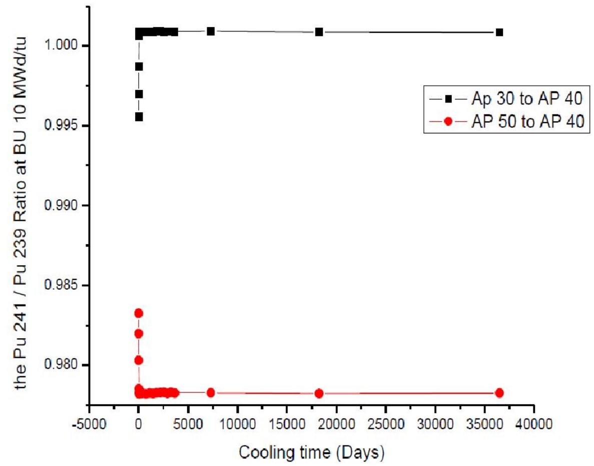


Figure 11. The ratio of $^{241}Pu$/ $^{239}Pu$ against CT at different AP values for BU=10 MWd/tu

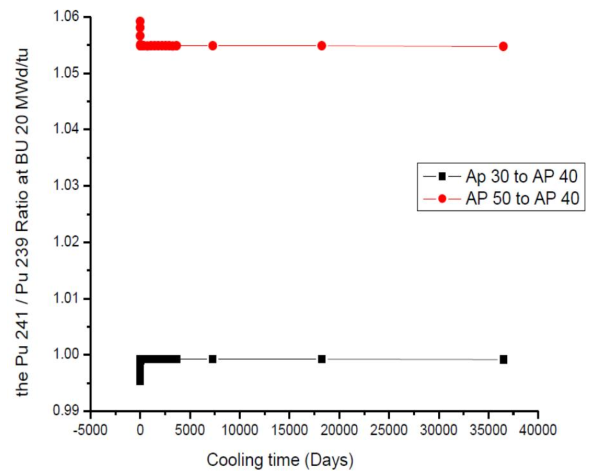


Figure 12. The 241Pu/ 239Pu ratio against CT at different AP values for BU=20 MWd/tu

Figures (11, 12) show that for low BU value there is almost no significant change in the Ratio $^{241}Pu$/ $^{239}Pu$, at low AP value (around 3%) and at BU=10 MWd/tu, while at higher AP value and it increases to 5% at BU=20 MWd/tu.

Figures (13, 14) show that for high BU values there are almost small change in the $^{241}Pu$/ $^{239}Pu$ ratio at low and high AP values (from 1.5% to 2%).

As for the $^{241}Pu$/ $^{239}Pu$ ratio the impact is noticeable at low and high burnup values. This is due to the production of $^{241}Pu$ from $^{240}Pu$ by neutron capture [11]. Since the total neutron emission is affected by changing the AP during 3 years of cooling time at low burnup values [8], so the production of $^{241}Pu$ is affected, and the Table of results is shown in Appendix (1).

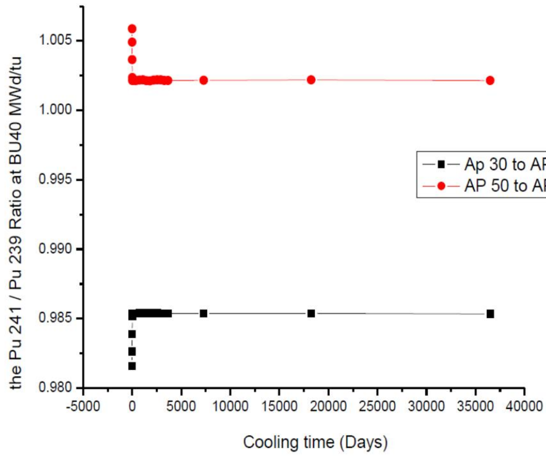


Figure 13. The $^{241}Pu/^{239}Pu$ ratio against CT at different AP values for BU=40 MWd/tu

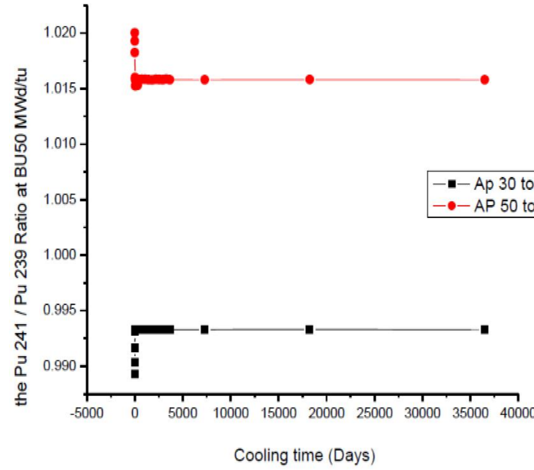


Figure 14. The $^{241}Pu/^{239}Pu$ ratio against CT at different AP values for BU=50 MWd/tu

APPENDIX 1. The ratio of $^{238}Pu/^{239}Pu$ at BU=10 GWd/tu,

| Decay time | AP 30 | AP 40 | AP 50 | Ap30/AP40 | Ap50/AP40 |
|---|---|---|---|---|---|
| 0 | 0.0019 | 0.00185 | 0.00175 | 1.02815 | 0.94961 |
| 1 | 0.0019 | 0.00185 | 0.00176 | 1.02717 | 0.95055 |
| 3 | 0.00191 | 0.00186 | 0.00177 | 1.02608 | 0.9516 |
| 10 | 0.00192 | 0.00187 | 0.00178 | 1.02536 | 0.95232 |
| 30 | 0.00192 | 0.00187 | 0.00178 | 1.02562 | 0.95209 |
| 100 | 0.00193 | 0.00188 | 0.00178 | 1.02663 | 0.95144 |
| 300 | 0.00193 | 0.00188 | 0.00179 | 1.02831 | 0.9504 |
| 365 | 0.00193 | 0.00188 | 0.00179 | 1.02858 | 0.9502 |
| 730 | 0.00193 | 0.00187 | 0.00178 | 1.02932 | 0.94973 |
| 1095 | 0.00191 | 0.00186 | 0.00176 | 1.02947 | 0.94962 |
| 1460 | 0.0019 | 0.00184 | 0.00175 | 1.0295 | 0.9496 |
| 1825 | 0.00188 | 0.00183 | 0.00174 | 1.02955 | 0.94962 |
| 2190 | 0.00187 | 0.00182 | 0.00172 | 1.02954 | 0.94961 |
| 2555 | 0.00185 | 0.0018 | 0.00171 | 1.02956 | 0.94961 |
| 2920 | 0.00184 | 0.00179 | 0.0017 | 1.02955 | 0.94961 |
| 3285 | 0.00183 | 0.00177 | 0.00168 | 1.02955 | 0.94959 |
| 3650 | 0.00181 | 0.00176 | 0.00167 | 1.02956 | 0.94961 |
| 20 | 0.00167 | 0.00163 | 0.00154 | 1.02959 | 0.94957 |
| 7300 | 0.00155 | 0.0015 | 0.00143 | 1.02964 | 0.94955 |
| 18250 | 0.00132 | 0.00128 | 0.00122 | 1.02968 | 0.94947 |
| 36500 | 8.92972E-4 | 8.67017E-4 | 8.23124E-4 | 1.02994 | 0.94938 |

APPENDIX 1. The ratio of $^{238}$Pu/ $^{239}$Pu at BU=20 GWd/tu,

| Decay time | AP 30 | AP 40 | AP 50 | Ap30/AP40 | Ap50/AP40 |
|---|---|---|---|---|---|
| 0 | 0.0072 | 0.00703 | 0.00696 | 1.02409 | 0.9904 |
| 1 | 0.0072 | 0.00703 | 0.00697 | 1.02393 | 0.99059 |
| 3 | 0.00721 | 0.00704 | 0.00698 | 1.02377 | 0.99068 |
| 10 | 0.00722 | 0.00705 | 0.00698 | 1.02393 | 0.99042 |
| 30 | 0.00725 | 0.00708 | 0.007 | 1.02431 | 0.98961 |
| 100 | 0.00733 | 0.00715 | 0.00706 | 1.02536 | 0.98753 |
| 300 | 0.00746 | 0.00726 | 0.00715 | 1.02696 | 0.98411 |
| 365 | 0.00747 | 0.00728 | 0.00716 | 1.02728 | 0.98353 |
| 730 | 0.00749 | 0.00728 | 0.00715 | 1.02797 | 0.98203 |
| 1095 | 0.00744 | 0.00724 | 0.00711 | 1.02813 | 0.98175 |
| 1460 | 0.00739 | 0.00718 | 0.00705 | 1.02819 | 0.98168 |
| 1825 | 0.00733 | 0.00713 | 0.007 | 1.02818 | 0.98164 |
| 2190 | 0.00727 | 0.00707 | 0.00694 | 1.02819 | 0.98165 |
| 2555 | 0.00722 | 0.00702 | 0.00689 | 1.02819 | 0.98164 |
| 2920 | 0.00716 | 0.00696 | 0.00684 | 1.02819 | 0.98162 |
| 3285 | 0.0071 | 0.00691 | 0.00678 | 1.02818 | 0.98159 |
| 3650 | 0.00705 | 0.00685 | 0.00673 | 1.02821 | 0.98166 |
| 20 | 0.00652 | 0.00634 | 0.00622 | 1.02824 | 0.98161 |
| 7300 | 0.00602 | 0.00586 | 0.00575 | 1.02826 | 0.98156 |
| 18250 | 0.00515 | 0.00501 | 0.00492 | 1.02837 | 0.98152 |
| 36500 | 0.00348 | 0.00338 | 0.00332 | 1.02856 | 0.98123 |

APPENDIX 1. The ratio of $^{238}$Pu/ $^{239}$Pu at BU=30 GWd/tu,

| Decay time | AP 30 | AP 40 | AP 50 | Ap30/AP40 | Ap50/AP40 |
|---|---|---|---|---|---|
| 0 | 0.01725 | 0.01681 | 0.01642 | 1.02625 | 0.97717 |
| 1 | 0.01724 | 0.0168 | 0.01642 | 1.02635 | 0.97715 |
| 3 | 0.01724 | 0.0168 | 0.01641 | 1.0267 | 0.97708 |
| 10 | 0.01726 | 0.0168 | 0.01641 | 1.02736 | 0.97691 |
| 30 | 0.01735 | 0.01687 | 0.01648 | 1.02849 | 0.97692 |
| 100 | 0.0176 | 0.01707 | 0.01668 | 1.03128 | 0.97716 |
| 300 | 0.018 | 0.01737 | 0.01698 | 1.03595 | 0.97758 |
| 365 | 0.01806 | 0.01742 | 0.01703 | 1.03676 | 0.97766 |
| 730 | 0.01812 | 0.01745 | 0.01706 | 1.03873 | 0.97791 |
| 1095 | 0.01803 | 0.01735 | 0.01696 | 1.03915 | 0.97784 |
| 1460 | 0.0179 | 0.01722 | 0.01684 | 1.03934 | 0.97791 |
| 1825 | 0.01776 | 0.01709 | 0.01671 | 1.03935 | 0.97787 |
| 2190 | 0.01762 | 0.01695 | 0.01658 | 1.0393 | 0.97786 |
| 2555 | 0.01748 | 0.01682 | 0.01645 | 1.03929 | 0.97787 |
| 2920 | 0.01735 | 0.01669 | 0.01632 | 1.03934 | 0.97787 |
| 3285 | 0.01721 | 0.01656 | 0.01607 | 1.03933 | 0.97019 |
| 3650 | 0.01708 | 0.01643 | 0.01607 | 1.03932 | 0.97782 |
| 20 | 0.01579 | 0.01519 | 0.01485 | 1.03939 | 0.97779 |
| 7300 | 0.0146 | 0.01404 | 0.01373 | 1.03946 | 0.97774 |
| 18250 | 0.01248 | 0.012 | 0.01173 | 1.03957 | 0.97765 |
| 36500 | 0.00843 | 0.00811 | 0.00792 | 1.03991 | 0.97735 |

APPENDIX 1. The ratio of $^{238}Pu/ ^{239}Pu$ at BU= 40 GWd/tu,

| Decay time | AP 30 | AP 40 | AP 50 | Ap30/AP40 | Ap50/AP40 |
|---|---|---|---|---|---|
| 0 | 0.0331 | 0.03185 | 0.03111 | 1.03896 | 0.9765 |
| 1 | 0.03307 | 0.03182 | 0.03107 | 1.03932 | 0.97626 |
| 3 | 0.03304 | 0.03178 | 0.03101 | 1.03971 | 0.97574 |
| 10 | 0.03305 | 0.03176 | 0.03097 | 1.04036 | 0.97506 |
| 30 | 0.03321 | 0.03191 | 0.0311 | 1.0407 | 0.97446 |
| 100 | 0.03372 | 0.03237 | 0.03149 | 1.04172 | 0.97296 |
| 300 | 0.03449 | 0.03306 | 0.03209 | 1.04322 | 0.9706 |
| 365 | 0.03461 | 0.03317 | 0.03218 | 1.04348 | 0.97023 |
| 730 | 0.03475 | 0.03328 | 0.03225 | 1.04412 | 0.96922 |
| 1095 | 0.03456 | 0.0331 | 0.03207 | 1.04426 | 0.96901 |
| 1460 | 0.03431 | 0.03285 | 0.03183 | 1.04426 | 0.9689 |
| 1825 | 0.03404 | 0.0326 | 0.03159 | 1.04432 | 0.96893 |
| 2190 | 0.03378 | 0.03235 | 0.03134 | 1.04431 | 0.96891 |
| 2555 | 0.03351 | 0.03209 | 0.03109 | 1.04429 | 0.9689 |
| 2920 | 0.03325 | 0.03184 | 0.03085 | 1.04433 | 0.96892 |
| 3285 | 0.03299 | 0.03159 | 0.03061 | 1.04435 | 0.9689 |
| 3650 | 0.03273 | 0.03134 | 0.03037 | 1.04434 | 0.96888 |
| 20 | 0.03026 | 0.02898 | 0.02807 | 1.04441 | 0.96888 |
| 7300 | 0.02798 | 0.02679 | 0.02595 | 1.04443 | 0.96888 |
| 18250 | 0.02391 | 0.02289 | 0.02218 | 1.04458 | 0.96882 |
| 36500 | 0.01615 | 0.01546 | 0.01497 | 1.0449 | 0.9687 |

APPENDIX 1. The ratio of $^{238}Pu/ ^{239}Pu$ at BU=50 GWd/tu,

| Decay time | AP 30 | AP 40 | AP 50 | Ap30/AP40 | Ap50/AP40 |
|---|---|---|---|---|---|
| 0 | 0.05463 | 0.05233 | 0.05103 | 1.04399 | 0.97511 |
| 1 | 0.05456 | 0.05224 | 0.05094 | 1.04444 | 0.97503 |
| 3 | 0.05448 | 0.05213 | 0.05081 | 1.04507 | 0.97478 |
| 10 | 0.05444 | 0.05205 | 0.05066 | 1.04593 | 0.97335 |
| 30 | 0.05469 | 0.05227 | 0.05084 | 1.04626 | 0.97252 |
| 100 | 0.05547 | 0.05299 | 0.05151 | 1.04683 | 0.97207 |
| 300 | 0.05664 | 0.05406 | 0.05251 | 1.04771 | 0.97125 |
| 365 | 0.05682 | 0.05422 | 0.05269 | 1.04789 | 0.97169 |
| 730 | 0.057 | 0.05438 | 0.05282 | 1.04824 | 0.97134 |
| 1095 | 0.05669 | 0.05408 | 0.05252 | 1.04832 | 0.97127 |
| 1460 | 0.05627 | 0.05368 | 0.05214 | 1.04833 | 0.97126 |
| 1825 | 0.05584 | 0.05326 | 0.05173 | 1.04833 | 0.97124 |
| 2190 | 0.0554 | 0.05285 | 0.05133 | 1.04833 | 0.97122 |
| 2555 | 0.05497 | 0.05243 | 0.05092 | 1.04833 | 0.97121 |
| 2920 | 0.05454 | 0.05202 | 0.05053 | 1.04837 | 0.97127 |
| 3285 | 0.05411 | 0.05161 | 0.05013 | 1.04838 | 0.97124 |
| 3650 | 0.05369 | 0.05121 | 0.04974 | 1.04837 | 0.97124 |
| 20 | 0.04963 | 0.04734 | 0.04597 | 1.04839 | 0.97119 |
| 7300 | 0.04588 | 0.04376 | 0.0425 | 1.04848 | 0.97117 |
| 18250 | 0.03921 | 0.03739 | 0.03631 | 1.0485 | 0.9711 |
| 36500 | 0.02647 | 0.02524 | 0.02451 | 1.04878 | 0.97096 |

APPENDIX 1. The ratio of $^{238}Pu$/ $^{239}Pu$ at BU= 60 GWd/tu¸

| Decay time | AP 30 | AP 40 | AP 50 | Ap30/AP40 | Ap50/AP40 |
|---|---|---|---|---|---|
| 0 | 0.08056 | 0.07739 | 0.07486 | 1.04101 | 0.96738 |
| 1 | 0.08042 | 0.07721 | 0.07464 | 1.04164 | 0.96682 |
| 3 | 0.08025 | 0.07697 | 0.07437 | 1.04251 | 0.9661 |
| 10 | 0.08014 | 0.07679 | 0.07412 | 1.04362 | 0.96527 |
| 30 | 0.08045 | 0.07705 | 0.07437 | 1.04411 | 0.9652 |
| 100 | 0.08147 | 0.07796 | 0.07526 | 1.04504 | 0.96541 |
| 300 | 0.08299 | 0.0793 | 0.07658 | 1.04659 | 0.96576 |
| 365 | 0.08322 | 0.07949 | 0.07678 | 1.04683 | 0.96581 |
| 730 | 0.0834 | 0.07962 | 0.07691 | 1.04751 | 0.96596 |
| 1095 | 0.08292 | 0.07915 | 0.07646 | 1.04764 | 0.96598 |
| 1460 | 0.08231 | 0.07856 | 0.07589 | 1.04766 | 0.96599 |
| 1825 | 0.08167 | 0.07795 | 0.0753 | 1.04771 | 0.966 |
| 2190 | 0.08103 | 0.07734 | 0.07471 | 1.0477 | 0.96599 |
| 2555 | 0.0804 | 0.07674 | 0.07413 | 1.04769 | 0.96599 |
| 2920 | 0.07977 | 0.07614 | 0.07355 | 1.0477 | 0.96598 |
| 3285 | 0.07914 | 0.07554 | 0.07297 | 1.04771 | 0.96598 |
| 3650 | 0.07852 | 0.07495 | 0.0724 | 1.04772 | 0.966 |
| 20 | 0.07258 | 0.06927 | 0.06692 | 1.04773 | 0.96597 |
| 7300 | 0.06709 | 0.06403 | 0.06185 | 1.04777 | 0.96594 |
| 18250 | 0.05733 | 0.05471 | 0.05284 | 1.04781 | 0.96591 |
| 36500 | 0.03869 | 0.03692 | 0.03566 | 1.048 | 0.96581 |

**4. Conclusion**

The plutonium isotopic ratio for Spent Fuel Elements with different irradiation histories was studied by using ORIGEN-ARP code. The case considered is LEU 17×17 PWR fuel with an initial enrichment of 4.5%. The BU ranged between 10 and 60 GWd/tu and 20 values of cooling time, from 0 up to 100 years, were also considered. The varied parameter is the average power level.

The analysis of the obtained data revealed that plutonium isotopic ratio is of little sensitivity to the average power for BU values above 20 GWd/tu. It was found that, by varying the considered irradiation history parameters, the total neutron emission is affected. Consequently, the isotopes that depend on neutron capture in their production are also affected.

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