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Fingerprinting commercial nuclear reactor forensics using the ORIGEN-S simulation code

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The ability to determine the origin of a specific spent-fuel sample from a commercial nuclear reactor was studied using the Origen-S simulation code by calculating the plutonium and uranium isotopic concentration data for a range of nuclear power reactors. This range of reactors is based on a typical Westinghouse PWR fuel assembly with a fuel type of W17 X 17, having individual operating histories. Isotopic ratios of plutonium in nuclear reactors during the fuel-cycle period provide information on how the plutonium grows into the fuel as a function of burnup, as well as its attractiveness to proliferators. Using the results from the calculation of uranium and plutonium isotopic ratios, the origin of each spent-fuel assembly for a particular reactor can be predicted and documented for a future nuclear forensics reference database.

Keywords: Nuclear forensics; ORIGEN-S, PWR and Pu isotopic ratio.

1. Introduction

Traditional nuclear forensic techniques rely on precise and sophisticated measurement of the isotopic, chemical, and morphological signatures of a sample of spent fuel to ascertain its process history, specifically its origin and transportation route (see Refs. 1–5 for more details). When these measurement techniques are applied to nuclear reactor fuel, they are conducted in two stages:

- Fresh fuel after sintering and fabrication but prior to loading into the core.
- After the spent fuel has been unloaded and allowed to cool (thermally and radioactively) after being irradiated during multiple fuel cycles, all of which are associated with a new rearrangement within the core.

Due to the harsh environment surrounding spent nuclear fuel, physical and chemical measurement of the fuel pose significant logistical and safety challenges; therefore they cannot be performed until several years after discharge once fission product species (e.g., ^{137}Cs and ^{90}Sr) and minor actinides have appreciably decayed. Accurate simulation techniques were developed to overcome those challenges. These techniques employ Bateman equations and destructive radiochemical and non-destructive assay measurements to validate the resulting models.

Commercial nuclear reactors of the same type are presumed to have similar nuclear parameters, such as the amount of fuel destroyed over time (i.e., burnup), neutron transport, moderator-coolant materials, and thermal hydraulics. However, the spent fuel of each nuclear reactor requires different forensic techniques to determine its origin. The operational parameters unique to a specific reactor can be used to differentiate spent-fuel samples from different types of reactors or even between individual reactors of the same design. Therefore, measurement and calculation of the plutonium and uranium isotopic ratios of a specific reactor can produce a unique "fingerprint" of that reactor. This can be accomplished by employing a standalone ORIGEN-S simulation code. The code is implemented via a computational software tool available from the Radiation Safety Information Computational Center of the Oak Ridge National Laboratory (ORNL), USA. This method will be used to compile a database for all commercial nuclear reactors under investigation.

Nuclear forensics requires a combination of technical data and specialized skills and knowledge to retrieve, analyze, and interpret the data. Research and development of nuclear forensic techniques has focused on both destructive and non-destructive assay measurements. Nicolaou[6] conducted ORIGEN-S simulations of different reactor types (such as PWR, BWR, VVER, CANDU, and Magnox) by applying visualization and sample-classification techniques. In the most recent revision of the Origen-S code, a substantial amount of work went into benchmarking this code, specifically the cross-section libraries, which improved the computational tool's accuracy. Robel[7] used the updated version of ORIGEN-S, which enabled realistic variation in fresh-fuel composition, as well as classification of actual unknown spent fuels using Partial Least Square Discriminant Analysis (PLSDA). The goal of this study was to illustrate the distinguishing factors among nuclear reactors of the same type by investigating the differences between their unique uranium and plutonium isotopic ratios.

2. Materials and Method

In this work, the ORIGEN-ARP code system[8] was used to calculate the plutonium and uranium isotope compositions of the reactor spent fuel during the period of cycle up to the end of irradiation (EOI). The research focused on Pressurized Water Reactors (PWRs) presently in operation within the United States, with emphasis on output electrical power between 900 and 1200 MWe. This reactor type uses boric acid as burnable absorber and runs a complete cycle without unplanned outage and with no control-rod activity. Table 1 lists the characteristics of the commercial nuclear reactors

that were investigated. It includes the corresponding electrical power output, fuel enrichment, burnup value, fuel cycle, and power history. The goal of this investigation was to model the fuel assembly of typical Generation II Westinghouse PWRs that fall within the above-mentioned range to produce isotopic plutonium compositions for benchmark comparison. The burnups of the fuel samples analyzed ranged from 33 to 40 GWd/MtU, with the fuel enrichment distributed from 2.8 to 4.1 wt% of ^{235}U. The output from ORIGEN-ARP simulations is representative of the fuel assembly as a whole; it includes information on actinides, fission products, and neutron flux. It is important to note that an assumption about the fuel assemblies was made based on the uniform distribution of flux throughout the reactor core without considering a particular region. This assumption could result in uncertainties in the data obtained.

Table 1. Overview of the characteristics of commercial nuclear reactors.

Reactor	Electrical Output (MWe)	Fuel Enrichment (%)	Burn Up (GWd/MTU)	Fuel Cycle (months)	Power History (%)
Comanche Peak 1	1209	4.0	40	18	91
North Anna 1	903	2.8	39	18	78
Indian Point 2	1022	4.1	36	24	98
Seabrook Station 1	1080	3.57	33	12	77

3. Results and Discussion

Uranium and plutonium isotopic data were obtained from the reactors listed in Table 1. The atomic concentrations of uranium and plutonium isotopes calculated using ORIGEN-ARP code are shown in Figs. 1 and 2, respectively. This provides information on the enrichment of "unknown" spent fuel. ^{239}Pu has the highest atomic concentration among the other isotopes present, while ^{238}Pu gives the lowest concentration in all cases, as expected. Although all four cases considered belong to the same type of reactor, there is a unique difference in the concentration ratios of ^{235}U/^{236}U at the EOI.

Isotopic ratios of plutonium ($^{239/240}$Pu, $^{239/241}$Pu, and $^{239/242}$Pu) are shown in Fig. 3. This provides information on the neutron flux in each case. The isotopic ratios of plutonium depicted in Fig. 3 provide information about the neutron flux (n/cm^2-sec) attained by the following individual reactors at the EOI: Comanche Peak (8.86×10^{13}), Indian Point (5.68×10^{13}), North Anna (1.13×10^{14}), and Seabrook (1.15×10^{14}). Another important dataset is the depletion of ^{241}Pu due to β^- decay to ^{241}Am ($T_{1/2} = 14.9$ y) in each case. The tendency of the neutron flux of different reactors to diverge as burnup progresses suggests that this behavior can serve as a marker to determine the type of reactor in which a given sample of spent nuclear fuel was irradiated. The alpha activity ratio of ^{238}Pu/(^{239}Pu + ^{240}Pu) shown in Fig. 4 gives values around 2.0 at the end of the irradiation period in each case.

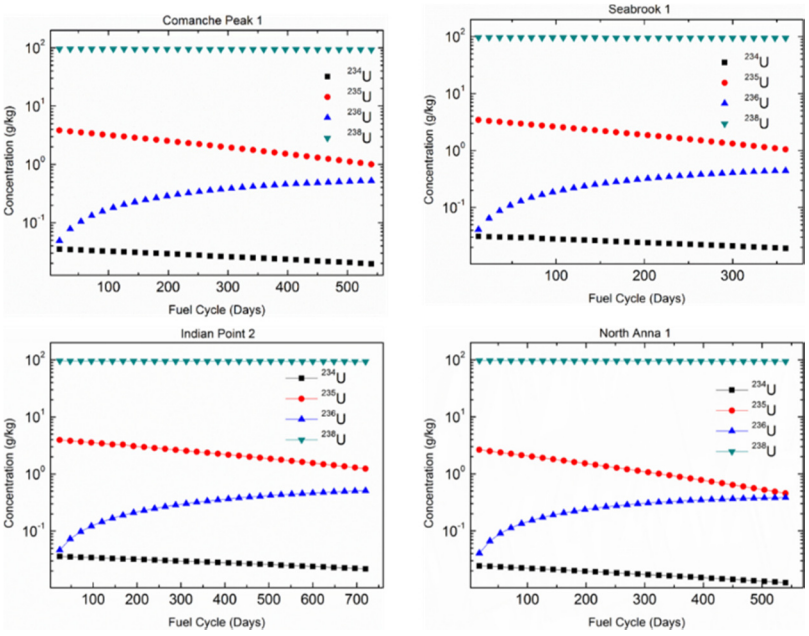

Fig. 1. Concentration of uranium isotopic content during the fuel-cycle period and EOI.

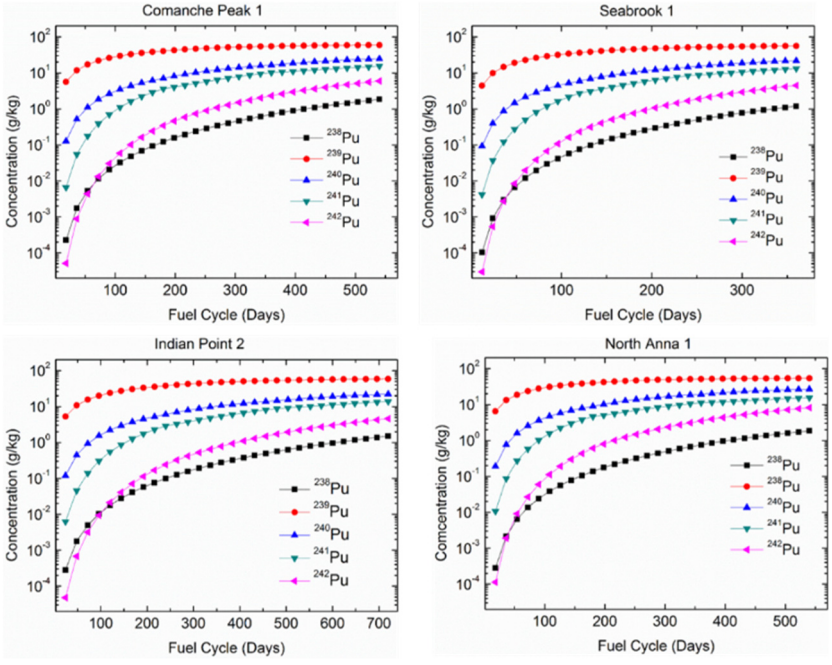

Fig. 2. Concentration of plutonium isotopic content during the fuel-cycle period and EOI.

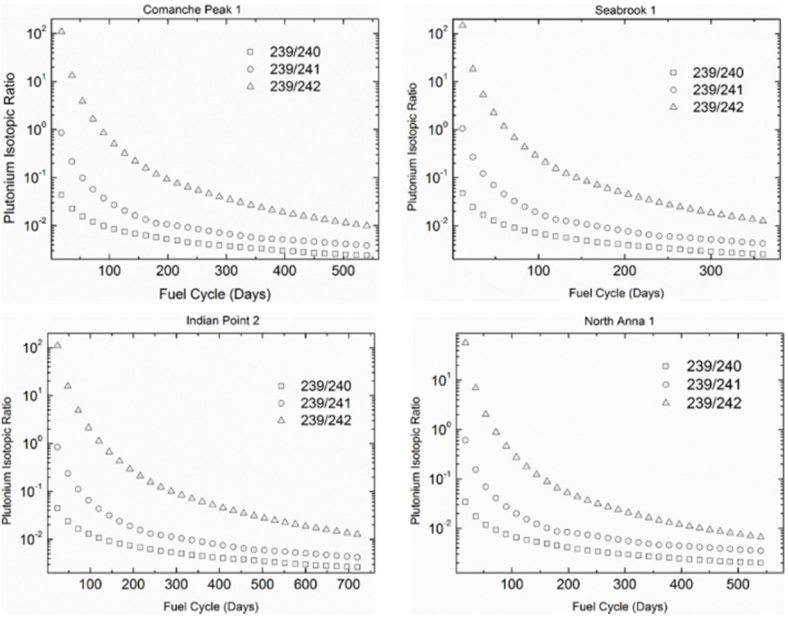

Fig. 3. Isotopic ratio of plutonium in nuclear reactors during the fuel-cycle period and EOI.

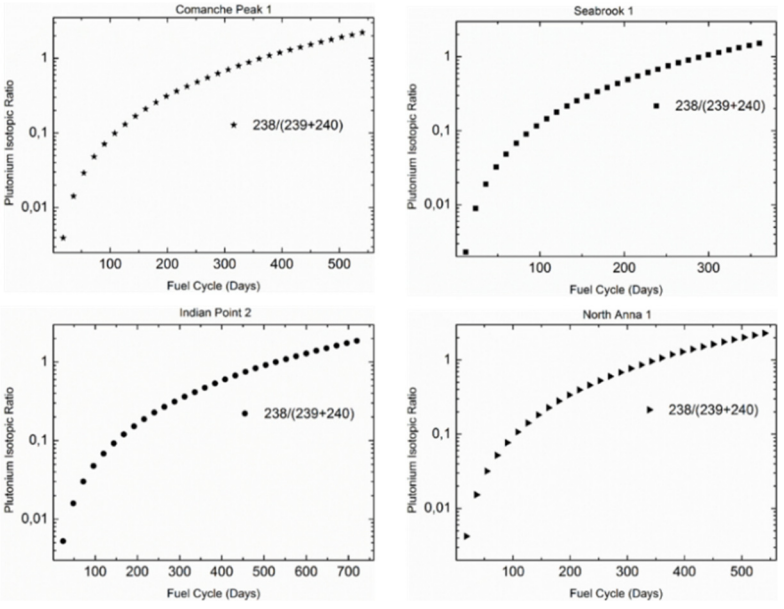

Fig. 4. Isotopic ratio of plutonium in nuclear reactors during the fuel-cycle period and EOI.

Plutonium isotopic ratios shown in Fig. 4 ($^{238/238+240}$Pu) illustrate how the plutonium grows in the fuel as a function of burnup, as well as its attractiveness to proliferators. The data indicate that the best time to divert ^{239}Pu is early-on in the fuel cycle, before the "poisonous" plutonium isotopes have time to accumulate. However, the build-up of ^{238}Pu originating from α-decay of ^{242}Cm ($T_{1/2} = 163$ d) should not be underestimated in each case.

4. Conclusions

It can be deduced that ^{239}Pu has the highest atomic concentration among the other isotopes present, while ^{238}Pu gives the lowest concentration in all cases considered. In addition, the isotopic ratios of plutonium (239/240, 239/241, and 239/242) provided information on when to divert ^{239}Pu from the reactor if the necessary applicable methods are available. Although an appreciable uncertainty can be obtained from the results, this needs to be validated and benchmarked with the experimental method's results. Future work on other types of reactors has been considered in order to complete the database.

Acknowledgments

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