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A composite position independent monitor of reactor fuel irradiation using Pu, Cs, and Ba isotope ratios

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ABSTRACT

When post-irradiation materials from the nuclear fuel cycle are released to the environment, certain isotopes of actinides and fission products carry signatures of irradiation history that can potentially aid a nuclear forensic investigation into the material's provenance. In this study, combinations of Pu, Cs, and Ba isotope ratios that produce position (in the reactor core) independent monitors of irradiation history in spent light water reactor fuel are identified and explored. These position independent monitors (PIMs) are modeled for various irradiation scenarios using automated depletion codes as well as ordinary differential equation solutions to approximate nuclear physics models. Experimental validation was performed using irradiated low enriched uranium oxide fuel from a light water reactor, which was sampled at 8 axial positions from a single rod. Plutonium, barium and cesium were chemically separated and isotope ratio measurements of the separated solutions were made by quadrupole and multi-collector inductively coupled mass spectrometry (Cs and Pu, respectively) and thermal ionization mass spectrometry (Ba). The effect of axial variations in neutron fluence and energy spectrum are evident in the measured isotope ratios. Two versions of a combined Pu and Cs based PIM are developed. A linear PIM model, which can be used to solve for irradiation time is found to work well for natural U fuel with < 10% ^{240}Pu and known or short cooling times. A non-linear PIM model, which cannot be solved explicitly for irradiation time without additional information, can nonetheless still group samples by irradiation history, including high burnup LEU fuel with unknown cooling time. ^{137}Ba/^{138}Ba is also observed to act as a position independent monitor; it is nearly single valued across the sampled fuel rod, indicating that samples sharing an irradiation history (same irradiation time and cooling time) in a reactor despite experiencing different neutron fluxes will have a common ^{137}Ba/^{138}Ba ratio. Modeling of this Ba PIM shows it increases monotonically with irradiation and cooling time, and a confirmatory first order analytical solution is also presented.

1. Introduction

Plutonium and fission products can be released into the environment from nuclear facilities (e.g., from an accidental or controlled release) or nefarious acts involving nuclear materials (e.g., nuclear terrorism or nuclear smuggling). Nuclear forensic analysis can be applied to these scenarios to uncover information about the provenance of the nuclear materials involved. In the case of releases from nuclear facilities, isotopic signatures in environmental samples may reveal information about the source term and transport of material (Donohue, 1998; Yamamoto et al., 2014). In the case of nuclear terrorism or smuggling, isotopic signatures can help investigators answer questions like “where might this material have been produced?” Anthropogenic Pu, Cs, and Ba are all produced in relatively high abundance in uranium-based nuclear reactor fuel (Brown et al., 2018). In this study, we will show that they all have isotopes that are sensitive in various ways to the irradiation environment in the fuel. Samples taken from multiple fuel pellets at different axial positions along a fuel rod with a known

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irradiation history were analyzed by mass spectrometry to quantify the Pu, Cs, and Ba isotopic composition. From these data, we identified previously unreported monitors of irradiation time that are relatively insensitive to the range of neutron fluxes typical of a thermal reactor operating in the tens of Megawatts power range. Using these monitors, we can independently show that the fuel pellets used for this study shared a common irradiation history despite experiencing a range of burnup of approximately a factor of two resulting from variations in the flux at different positions within the reactor core.

1.1. Intra-element isotope ratios and fission products as reactor fuel monitors

Isotope monitors in spent reactor fuel have been used extensively for safety, safeguards, and fuel performance purposes (Paoletti Gualandi et al., 1976), but their application to nuclear forensics has been more limited. Plutonium isotope ratios have been used as signatures of neutron energy spectrum and burnup (Moody et al., 2014). Inter-element ratios have been shown to be diagnostic (Scott, 2005), but are difficult to measure accurately due to differences in matrix dependent elemental abundance sensitivities. Noble gas intra-element isotope ratios in emissions can be diagnostic (Charleton et al., 2000), but their mobility limits applications and they do not persist after dissolution of spent fuel. The most widely used monitors are radioactive burnup monitors which are easily measured by gamma ray spectroscopy (e.g., ^{137}Cs). Stable isotopes such as ^{148}Nd offer a more precise and time invariant burnup monitor, but require the use of isotope dilution mass spectrometry (IDMS) instead of decay counting for isotope abundance quantification. In both cases, burnup is determined by measuring the absolute quantity of the monitor isotope, and hence both techniques assume that the material being analyzed retains its original chemical proportions. For nuclear forensic applications, where chemical alteration of the material often cannot be ruled out, intra-element isotope ratio monitors (e.g. ^{240}Pu/^{239}Pu) have the advantage of being highly resistant to fractionation.

1.2. Plutonium isotope system

Pu isotopes are of particular interest to nuclear forensics for a number of reasons. First, since Pu is not naturally occurring, there are very low backgrounds for Pu, aside from traces in the environment that remain from atmospheric weapons testing and reactor accidents. Pu isotope ratios are strongly correlated with burnup, but some Pu isotopes are also sensitive to the neutron energy spectrum. Last but not least, they are an important metric of the quality of the Pu produced in the fuel. A first order approximation of the ^{240}Pu/^{239}Pu as a function of fluence is

$$\frac{N_{P0}}{N_{P9}}(t) \approx \frac{\phi \sigma_c^{P9}}{2} \quad (1)$$

Where

N_{P0} is the atom density of ^{240}Pu;
N_{P9} is the atom density of ^{239}Pu;
σ_c^{P9} is the microscopic cross section for thermal neutron capture on ^{239}Pu;
ϕ is the neutron flux; and
t is irradiation time.

A derivation of Equation along with a comparison to SCALE code depletion model (Bowman, 2011) results is found in Appendix A.

1.3. Cesium isotopes as spent fuel monitors

1.3.1. ^{133}Cs and ^{137}Cs as burnup monitors

Many fission product concentrations are correlated with burnup. ^{137}Cs concentration is known as one of the best burnup monitors due to: 1) a relatively high fission yield at mass 137; 2) a long half-life relative to irradiation time; 3) the difference in chain yield as the fuel transitions from U fission to Pu fission is small (about 1–4 percent) (Brown et al., 2018); 4) ^{137}Cs concentration can readily be measured by gamma spectrometry; and 5) ^{137}Cs has a very low thermal neutron capture cross-section of about 0.1 b. ^{133}Cs also grows into reactor fuel relatively linearly with burnup and is a stable isotope. However, utilization of ^{133}Cs as a burnup monitor is complicated by that facts that 1) it cannot be measured by non-destructive techniques -it requires mass spectrometry analysis; 2) it has a potentially significant 30 b thermal neutron capture cross-section; 3) it is naturally occurring in the environment; 4) there is some flux sensitivity in the ^{133}Xe precursor, which has a 5.243 d half-life and 200 b thermal neutron capture cross section; and 5) there is greater potential for significant isotopic fractionation due to thermal gradient driven diffusion of Xe and I precursors in the fuel[13]. Fresh reactor fuel usually has a low concentration of Cs impurities –on the order of parts per billion. For spent PWR fuel initially enriched to 3%, after 30 GWd/MTU of irradiation, fissiogenic Cs concentration in spent fuel is about 0.6 wt %, or about two thirds of the Pu concentration.

The formation of ^{133}Cs is primarily through the decay of ^{133}Xe as the mass 133 chain fission products rapidly decay to stable ^{133}Cs. Ignoring the slight flux sensitivities of ^{133}Xe and ^{133}Cs, we approximate to first order the concentration of ^{133}Cs as a function of the mass 133 chain fission yield alone. Likewise, ignoring the decay of ^{137}Cs, we approximate to first order the concentration of ^{137}Cs as forming from fission alone. This gives:

$$\dot{N}_{C3(7)} = \gamma_{133(7)} \phi \Sigma_f^{fuel} \# \quad (2)$$

Assuming a constant flux, integrating over time yields:

$$N_{C3(7)}(t) = \gamma_{133(7)} \phi \Sigma_f^{fuel} t \# \quad (3)$$

Where

$\dot{N}_{C3(7)}$ is the rate of change of the number density of ^{133}Cs or ^{137}Cs in the fuel
$\gamma_{133(7)}$ is the fission yield of ^{133}Cs or ^{137}Cs in the fuel
Σ_f^{fuel} represents the macroscopic cross section of the sum of the main fissionable isotopes in the fuel (^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu)
$N_{C3(7)}$ is the number density of ^{133}Cs or ^{137}Cs in the fuel.

1.3.2. ^{135}Cs as a flux monitor

Unlike the other Cs isotopes, the production of ^{135}Cs is highly sensitive to thermal flux (Fig. 1), since it is formed from decay of ^{135}Xe (9.1 h). This latter nuclide has an extraordinary 2.6 Mb cross section for thermal neutron capture, making it the dominant fission product poison during reactor operation.

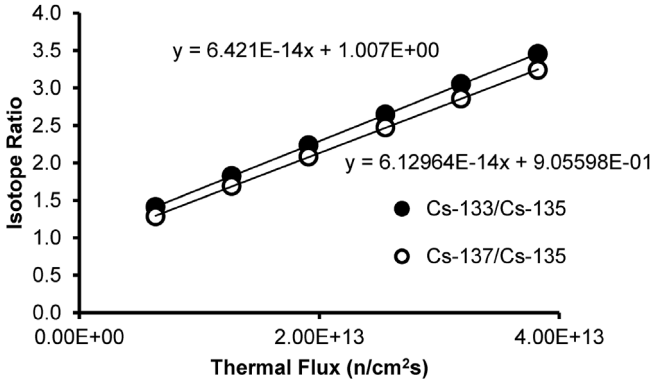

Fig. 1. Cs isotope ratios plotted against thermal flux from BR3 model in ORIGEN-ARP, shortly after shutdown. The slope of the ^{133}Cs/^{135}Cs is slightly higher due to higher fission chain yield of mass 133 than mass 137.