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Selected Isotopes for Optimized Fuel Assembly Tags

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October 2008

Pacific Northwest
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Summary

In support of our ongoing signatures project, we present information on three isotopes selected for possible use in optimized tags that could be applied to fuel assemblies to provide an objective measure of burnup.

1. Important factors for an optimized tag are compatibility with the reactor environment (corrosion resistance), low radioactive activation, at least two stable isotopes, a moderate neutron absorption cross section, which gives significant changes in isotope ratios over typical fuel assembly irradiation levels, and ease of measurement in the secondary ionization mass spectrometer.
2. From the candidate isotopes presented in the third FY 2008 quarterly report, the most promising appear to be titanium, hafnium, and platinum. The other candidate isotopes (iron and tungsten) exhibited inadequate corrosion resistance or had neutron capture cross sections either too high or too low for the burnup range of interest.

This report also presents preliminary mechanical design parameters. The most promising mechanical configuration considered so far is a wire or thin ribbon of metal, placed either inside an unused guide or instrumentation tube, or in a groove, machined specifically to accommodate the metal tag, on the outside of these tubes.

Acronyms and Abbreviations

BWR	boiling water reactor
Hf	hafnium
IRM	isotope ratio method
PNNL	Pacific Northwest National Laboratory
Pt	platinum
PWR	pressurized water reactor
SIMS	secondary ionization mass spectrometry
Ti	titanium
VVER	Russian version of a PWR; Russian abbreviation

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Introduction

This report describes the selection process for three isotopes that could be applied to optimized tags that can be applied to standard fuel assemblies used in light water reactors. This report describes work performed by the authors at Pacific Northwest National Laboratory for NA-22 as part of research to identify specific signatures that can be developed to support counter-proliferation technologies.

Isotope ratio methods have been used to estimate total lifetime fluence at research reactors by measuring impurities in aluminum core supports (Cliff et al. 2005) and by measuring impurities in graphite from graphite moderated reactors (Reid et al. 2001; Gesh 2004). One technique for measuring the relative abundance of two isotopes is the secondary ionization mass spectrometer (SIMS). It is particularly well adapted for atomic masses around 50 (Gerlach et al. 2006).

The isotope ratio method (IRM) estimates the energy production in a fission reactor by measuring isotope ratios in non-fuel reactor components. The isotope ratios in these components then can be related directly to the cumulative energy production with standard reactor calculations. Gerlach et al. (2006) discussed using impurities in the Zircaloy components of fuel assemblies, including measurements of samples taken from the fuel assembly channels of commercial boiling water reactors (BWRs)(Gerlach et al. 2007).

Measuring the change in the concentration of a specific isotope is possible; it is difficult to correlate to energy production because the initial concentration of that element may not be known. However, if the ratio of two isotopes of the same element can be measured, the energy production then can be determined without knowledge of the absolute concentration of that impurity because the initial natural ratio is known. This is the fundamental principle underlying the IRM. Extremely sensitive mass-spectrometric methods currently are available that allow accurate measurements of the isotope ratios in a tag.

Rather than rely on trace impurities, the optimized tag approach involves attaching a small tag or chip made of specific isotopes that have desirable nuclear, chemical, and ionization characteristics. One necessary nuclear characteristic is having at least two stable isotopes for isotope ratio measurements. Optimal isotopes have a thermal neutron capture crosssection low enough that they are not practically depleted during anticipated fuel assembly irradiation, have good corrosion resistance and chemical compatibility with the fuel assembly Zircaloy and coolant, and have atomic weights significantly higher or lower than major interference species in the secondary ionization mass spectrometry (SIMS) machine.

Isotope ratio techniques have been demonstrated on titanium and hafnium impurities in commercial Zircaloy. Given enough time and a full-scale SIMS machine, they easily can identify any assembly that was “short-cycled” to produce plutonium as opposed to a full cycle for energy production. However, the optimized tags should enable shorter measurement times and/or the use of miniaturized SIMS machines for analysis at the reactor site.

Candidate Elements and Isotopes and Selection of Optimal Tag Materials

In Table 1, the candidate elements and isotopes considered in the study are listed. Candidates selected had at least two stable isotopes and produced significant amounts of radioactive isotopes.

Table 1. Candidate Elements and Isotopes for Optimized Tags

Element	Stable Isotopes	Best Measurable Ratio(s)	R(0)/R(60 GWd/MT)[a]	Corrosion Resistance
Iron	54, 56, 57, 58	57/56	0.586	Poor
Hafnium	174, 176, 177, 178, 179, 180	178/176	0.172	Good
Osmium	184, 186, 187, 188, 189, 190, 192	190/192	0.713	Poor
Platinum	190, 192, 194, 195, 196, 198	196/195	0.368	Good
Titanium	46, 47, 48, 49, 50	49/48	0.583	Good
Tungsten	180, 182, 183, 184, 186	184/182	0.032	Fair

(a) Smaller numbers here indicate a greater change in the isotope ratio. Assuming there are no difficulties measuring these ratios, the smaller number implies a better indicator.

Review of the listed candidates for adequate corrosion resistance removed iron, osmium, and tungsten from consideration. Tungsten does, however, have the greatest change in isotope ratios and might be used in a tag if the tag were enclosed inside a clad tube. For the time being, we consider hafnium (Hf), titanium (Ti), and platinum (Pt) to be the most practical choices. Figures 1 through 7 show isotope ratio variations with burnup, using four different coolant densities. The highest density, 0.74g/cc, is typical of pressurized water reactors (PWRs), Russian PWRs (VVERs), and BWRs low in the core, where void fraction is essentially zero.

The variation of the $^{49}Ti/^{48}Ti$ ratio with burnup is shown in Figure 1; it has been presented in previous technical reports.

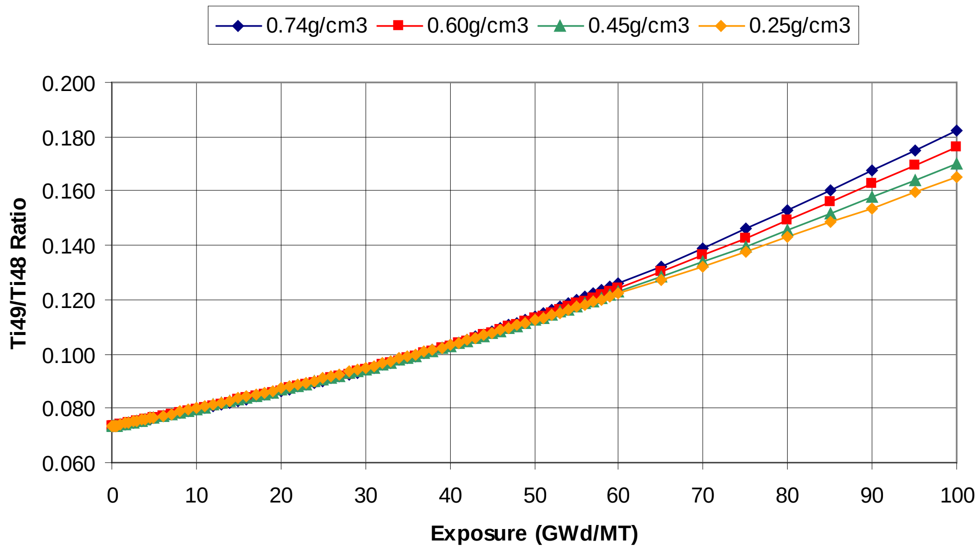

Figure 1. Calculated $^{49}Ti/^{48}Ti$ Ratios as a Function of Burnup

Hafnium has several natural stable isotopes and provides at least five potentially useful isotope ratios. Figures 2 through 6 show variation in specific hafnium isotope ratios over the normal commercial burnup of a fuel assembly.

As seen in Figure 2, the ratio of ^{174}Hf to ^{176}Hf declines steadily over the normal commercial operation of a fuel assembly. Note that the ratio itself is small because only 3% of natural hafnium is ^{174}Hf; it decreases in abundance with irradiation. This ratio is not difficult to measure in the laboratory but may not be the first choice to measure with the portable SIMS machine at the reactor site.

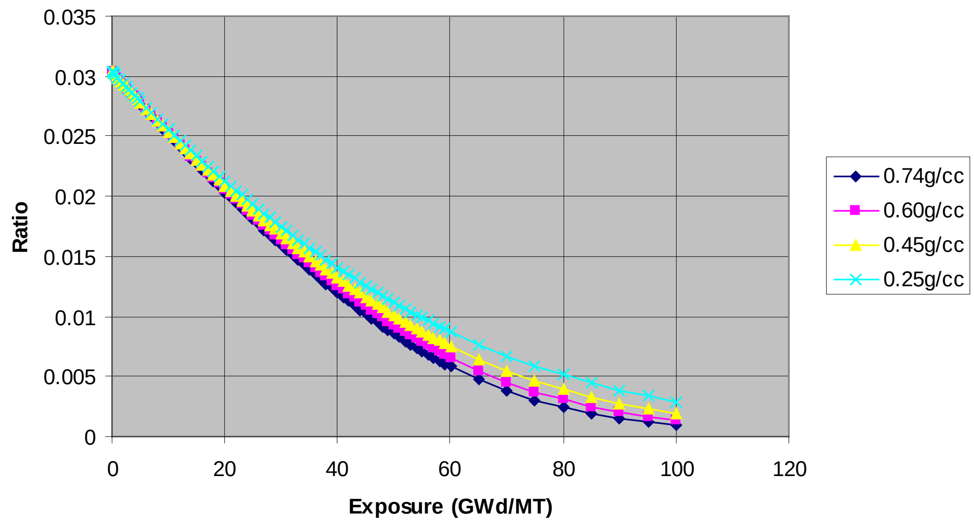

Figure 2. Calculated $^{174}Hf/^{176}Hf$ Ratios as a Function of Burnup

The ratio of ^{177}Hf to ^{176}Hf declines sharply with increasing burnup; it is most useful below about 20 GWd/MT.

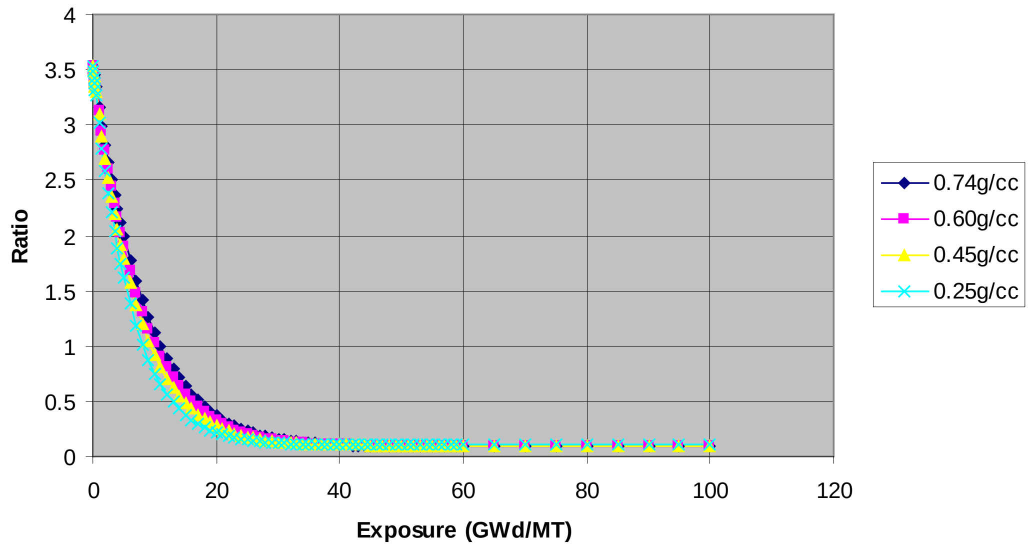

Figure 3. Calculated ^{177}Hf ^{176}Hf Ratios as a Function of Burnup

The ratio of ^{178}Hf to ^{176}Hf initially peaks around 10 GWd/MT, then declines over the normal commercial operation of a fuel assembly, as shown in Figure 4. This particular isotope ratio has a large difference between about 10 GWd/MT and the normal full-cycle operation of about 60 GWD/MT.

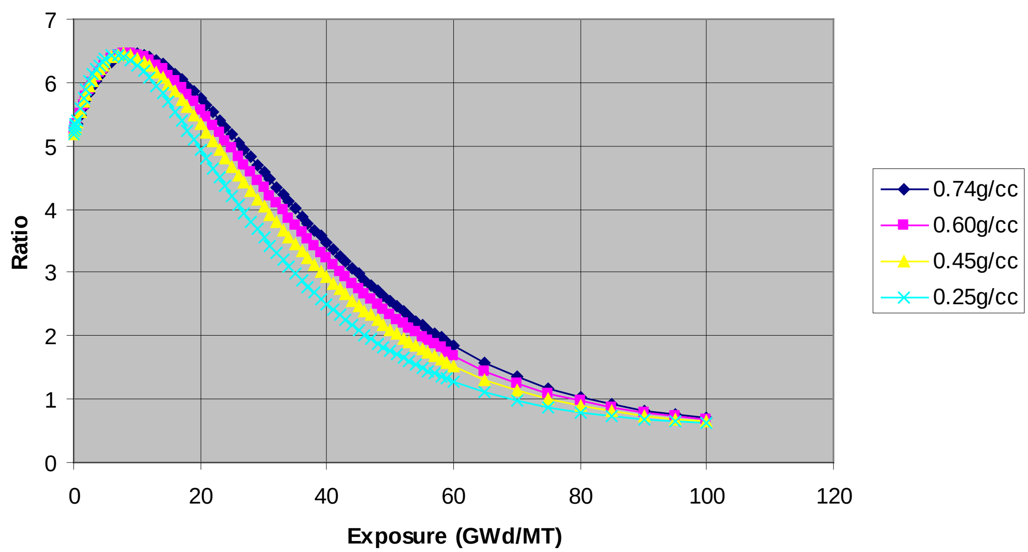

Figure 4. Calculated ^{178}Hf/^{176}Hf Ratios as a Function of Burnup

The ratio of ^{179}Hf to ^{176}Hf increases steadily during normal commercial operation of a fuel assembly (Figure 5). This isotope ratio also varies strongly between about 10 GWd/MT and normal commercial end of assembly life.

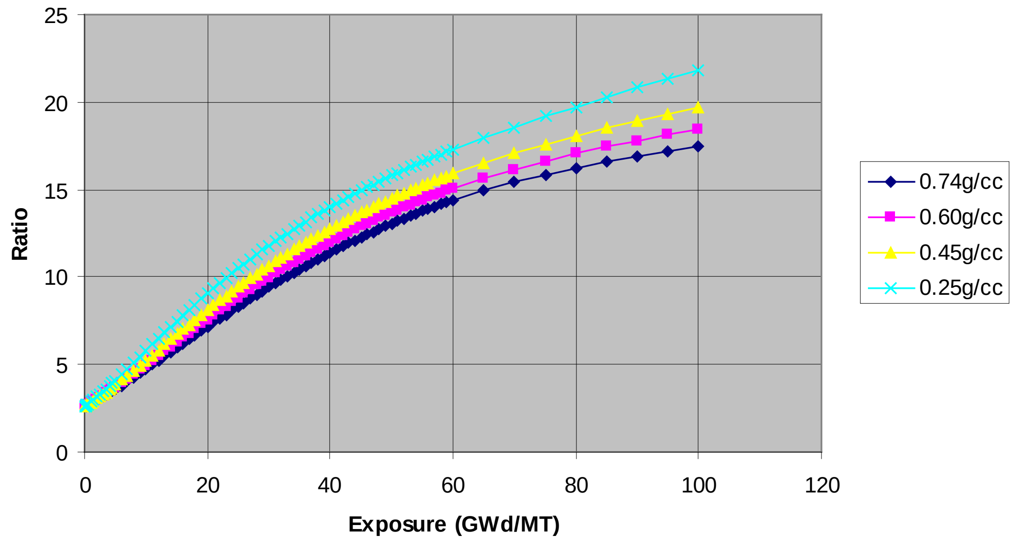

Figure 5. Calculated ^{179}Hf/^{176}Hf Ratios as a Function of Burnup

Figure 6 depicts the gradual increase in the ratio of ^{180}Hf to ^{176}Hfover the normal commercial operation of a fuel assembly. This ratio changes more with lower moderator density, such as would be seen in a BWR.

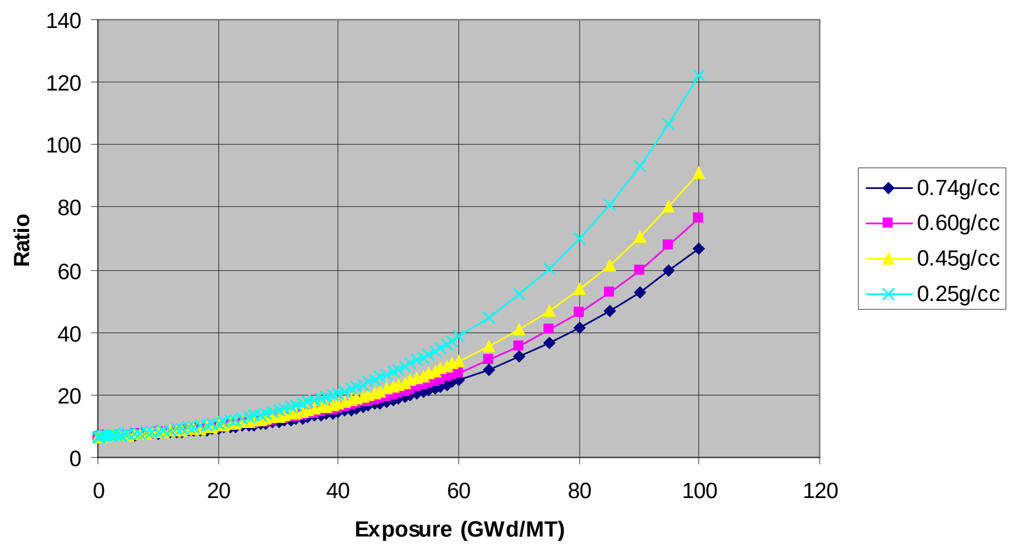

Figure 6. Calculated ^{180}Hf/^{176}Hf Ratios as a Function of Burnup

The ratio of ^{196}Pt to ^{195}Pt increases steadily over the normal commercial operation of a fuel assembly, as shown in Figure 7.

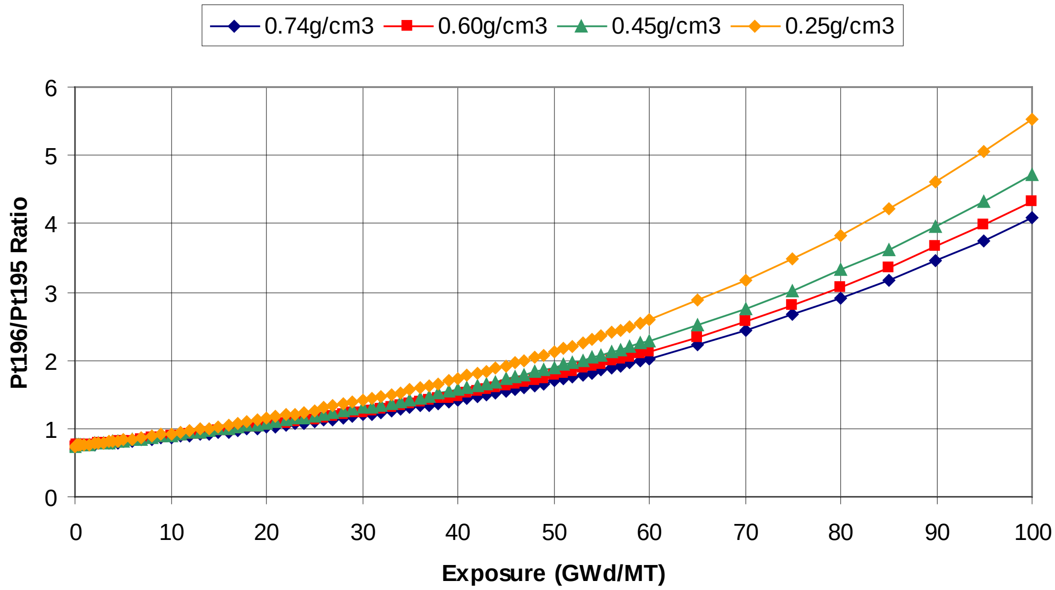

Figure 7. Calculated $^{196}Pt/^{195}Pt$ Ratios as a Function of Burnup

To date, the most practical form of a tag for use with typical light water reactor fuel assemblies appears to be either a wire or ribbon of metal. The wire or ribbon could be placed either in an unused guide or instrumentation tube or in a groove, machined specifically to accommodate the metal tag, on the outside of these tubes.

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