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Passive Neutron Resonance Absorption Densitometry of Thermal Reactor Spent-fuel

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Abstract

The Monte-Carlo particle transport code MCNP[1] was used to calculate the energy spectrum of neutrons near the surface of Pressure Water Reactor (PWR) assemblies, as a function of burnup. The calculated energy spectra are approximately inversely proportional to the energy, over the calculated energy range up to 1000 eV. The dominant resonance structures are flux depressions seen at the ^{238}U absorption resonances from 5 eV to 300 eV, the 0.3 eV ^{239}Pu resonance, and the 1 eV ^{240}Pu resonance. A simple Excel spreadsheet was used to estimate the signal that could be observed in ^{239}Pu and ^{235}U fission foils placed close to the PWR assembly surfaces and between various neutron filters made of natural Cd and Gd. Arrangements with two fission foils were considered. The first was placed behind a Gd filter that removed most of the thermal flux below ~0.1 eV. The second fission foil was placed behind a filter that removed most of the neutrons below ~0.5 eV. A signal that can be related to the plutonium content of a PWR assembly can be obtained by dividing the total neutron output by the difference in the observed fission rates in the first and second fission foils. The present model calculations predict that from low burnup to 40 GWd/MT, the above-mentioned ratio would vary by ~25%. The ratio change will be ~35% larger with plutonium foils then with uranium foils, and the counting statistics will be better with plutonium foils. To check for the sensitivity of the proposed ratio to elements other than plutonium, fission fragments and non- uranium and plutonium actinides were removed from the model calculations. These calculations showed that only ~25% of the ratio change was due to Am, Np, and/or the fission products.

Introduction

Spent nuclear fuel contains most of the world's weapons-useable fissile material. At present, the quantity of plutonium contained within the worldwide stockpile of spent fuel is ~1400 tonnes and is growing at ~70 tonnes/year [2]. This quantity of plutonium is already 5-6 times the worldwide military inventory [2]. The critical mass of reactor grade plutonium is only ~25% higher than that of weapons grade, and nuclear explosive devices can be designed that use reactor grade plutonium [3]. The IAEA has estimated that a motivated state could convert a few spent fuel assemblies into weapons-useable plutonium components in 1 to 3 months [4]. Safeguarding the plutonium contained within spent reactor-fuel is increasingly, becoming one of the dominant safeguards issues of the 21st century. In international safeguards, spent fuel is not characterized as self-protecting because non-nuclear-weapons states must be considered as possible proliferators. At present there are no assay methods that directly measure the plutonium content of non-

breeder-reactor spent fuel. This is because the gamma-ray doses are dominated by emissions from fission products and neutron doses are dominated by emissions from Cm isotopes. The fact that Cm dominates the neutron emission from light water reactor spent-fuel is a due to the fact that the nuclear reactions are dominantly being induced by thermal and near-thermal neutrons. This dominance of the slow moving neutrons is favorable to multiple neutron captures on ^{238}U creating significant quantities of Cm. This is not the case for fast breeder reactors where the neutron emission can be dominantly from the Pu isotopes. Measurements of fission product gamma-rays and Cm neutron counting rates are generally used in conjunction with operator declarations and computer codes to infer the plutonium content of spent fuel. These types of methods are open to several diversion scenarios.

The level of safeguards presently applied to the plutonium content of the world's nuclear reactor spent fuel would be significantly improved if a cost-effective underwater technique is developed to directly quantify the plutonium. In this paper we consider a technique that relies on the fact that one of the dominant neutron absorption mechanisms in spent fuel is neutron capture on ^{239}Pu, and that the neutron energy dependence of this capture reaction is very different from other capture reactions. The energy distribution of neutrons at the surface of spent fuel will contain flux depressions in the giant resonance region of ^{239}Pu from 0.1 to 0.5 eV.

Review articles on the non-destructive assay of spent nuclear fuels [5,6] have discussed the possibility of using neutron resonance absorption to assay the plutonium content of spent nuclear-fuel assemblies. It has been shown that neutron flux depressions can be used to assay many of the actinide isotopes contained within spent fuel. However, neutron resonance absorption has generally been dismissed because most authors have only considered detecting flux depressions using time-of-flight techniques with a strong pulsed neutron source. The technique presented here will dramatically simplify the neutron resonance absorption assay technique by using the neutrons generated by the spent fuel and measuring the ^{239}Pu flux depressions at the surface of assemblies using either ^{239}Pu or ^{235}U fission foils with filters that cut off the neutron energy spectra just below and above the ^{239}Pu giant resonance at ~0.3 eV.

MCNP Calculations

Figure 1 shows a cross sectional view of our MCNP model of a PWR assembly. The green circles show the fuel pins and the purple background represents the water. The modeled assembly has a standard PWR 17×17 grid, with 25 water channels, and 264 fuel pins. The modeled pins have a diameter of 0.95 cm with a Zr clad thickness of 0.057 cm. The UO_2 pallet diameter is 0.836 cm with a density of 10.3 g/cm^3. The initial ^{235}U enrichment was assumed to be 3.02%. All assemblies with finite burnup were assumed to have a cooling time of 3 years. The production and burnup of both fission fragments and actinides was determined using the code RADDB. Table 1 shows the assumed atomic fractions for the fuel at three different burnups. Only those isotopes with either non-negligent atomic fractions, or a non-negligent product of their atomic fraction and absorption cross-section near 0.3 eV are shown. For example, even though ^{242m}Am has a

neutron absorption cross section at 0.2 eV that is approximately a factor of two larger than the ^{239}Pu neutron absorption at 0.3 eV, the ^{242m}Am atomic fraction is more than 2000 times lower than ^{239}Pu, at an assembly burnup of 40 GWd/MT. Given the small amount of ^{242m}Am present it was not included in our MCNP calculations.

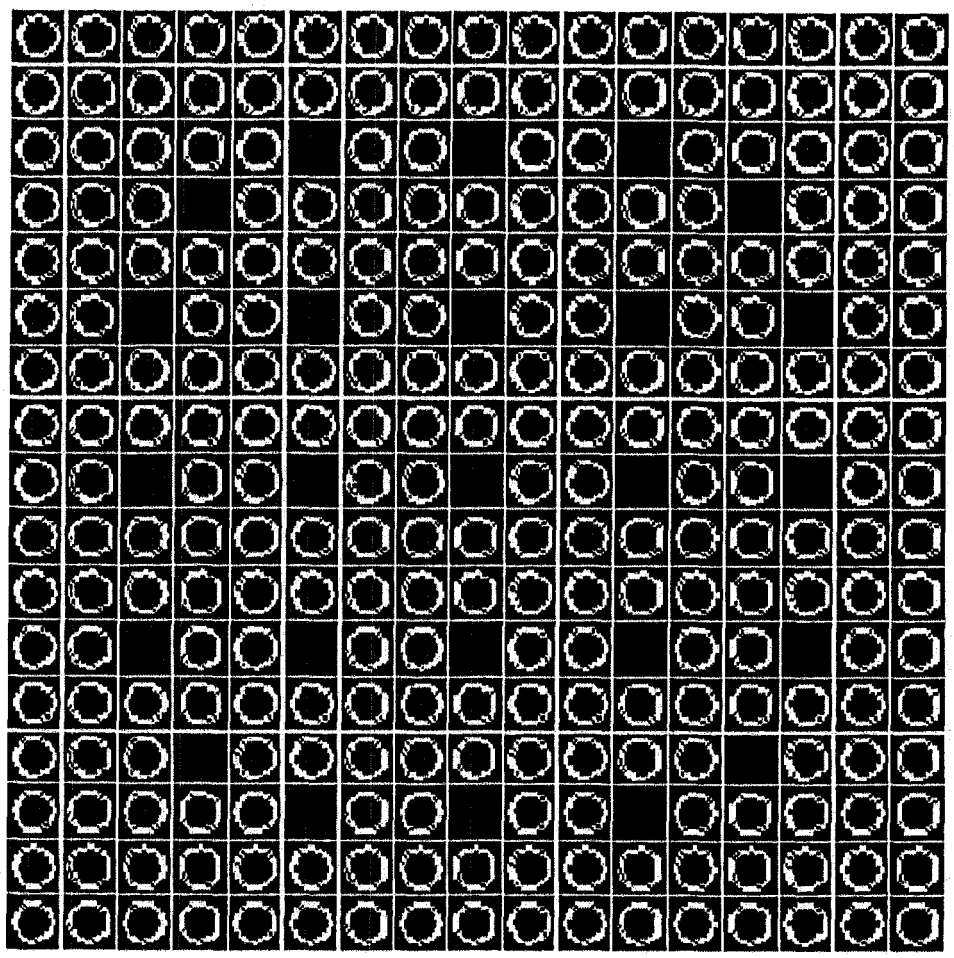

Figure 1. Cross sectional view of our MCNP model of a PWR assembly.

Figure 2 shows the calculated neutron energy spectra near the surface of a fresh PWR assembly, and near the surface of assemblies with burnups of 20 GWd/MT and 40 GWd/MT. The flux tally surfaces used were 1.6 mm from the outside edge of the outer row of pins. A fresh assembly does not contain any significant spontaneous fission neutron sources and thus one would not be able to measure neutrons at the surface of fresh assemblies. The fresh assembly calculations are to represent assemblies with a low but non-zero burnup. The flux depressions due to ^{238}U, ^{239}Pu, and ^{240}Pu are clearly seen in the 20 and 40 GWd/MT calculations. The only obvious flux depression seen from a fission product is the flux depression at ~16 eV due to absorption in ^{131}Xe.

Two additional neutron spectra were calculated. To quantitatively determine what fraction of the flux depressions near ~0.3 eV is due to fission fragments and non-plutonium/uranium actinides, calculations were performed at 40 GWd/MT with all the fission fragments and all Am and Np isotopes removed. To determine the effect of

additional water between the pins and the detection system, calculations were performed at 40 GWd/MT with an additional 4 mm of water between the outer row of fuel pins and the flux tally surfaces.

Table 1. Atomic fractions for PWR fuel at several burnups.

Isotope	Half life (y)	Fresh fuel	20 GWd/MT	40 GWd/MT
^{16}O	stable	0.66667	0.66667	0.66667
^{95}Mo	stable	0.00000	0.00047	0.00082
^{99}Tc	2×10^{5}	0.00000	0.00040	0.00074
^{101}Ru	stable	0.00000	0.00037	0.00075
^{105}Pd	stable	0.00000	0.00015	0.00040
^{131}Xe	stable	0.00000	0.00018	0.00029
^{133}Cs	stable	0.00000	0.00044	0.00079
^{135}Cs	2.3×10^{6}	0.00000	0.00012	0.00027
^{141}Pr	stable	0.00000	0.00039	0.00076
^{143}Nd	stable	0.00000	0.00032	0.00050
^{145}Nd	stable	0.00000	0.00023	0.00043
^{147}Sm	stable	0.00000	0.00006	0.00008
^{150}Sm	stable	0.00000	0.00010	0.00020
^{152}Sm	stable	0.00000	0.00005	0.00008
^{153}Eu	52.1	0.00000	0.00003	0.00008
^{235}U	7.0×10^{8}	0.01007	0.00449	0.00194
^{236}U	2.3×10^{7}	0.00000	0.00100	0.00135
^{238}U	4.5×10^{9}	0.32326	0.32212	0.31958
^{237}Np	2.1×10^{6}	0.00000	0.00008	0.00022
^{238}Pu	87.7	0.00000	0.00001	0.00010
^{239}Pu	2.4×10^{4}	0.00000	0.00152	0.00221
^{240}Pu	6537	0.00000	0.00044	0.00091
^{241}Pu	14.7	0.00000	0.00022	0.00048
^{242}Pu	3.8×10^{5}	0.00000	0.00005	0.00024
^{241}Am	432	0.00000	0.00001	0.00010

Excel Spreadsheet Calculations

To perform calculations in a timely and cost effective manner, the MCNP calculated neutron energy spectra were propagated through various filters and fission foils using a simple Excel spreadsheet. This spreadsheet assumes that all neutrons are traveling perpendicular to the filters and foils. Neutron absorption is the only mechanism considered in changing the neutron flux. This should not unduly affect the relative absorption and fission signals in the filters and foils, but the results presented here should be confirmed later with full MCNP calculations. To reduce the importance of the thermal region (<0.1 eV) a natural Gd foil was placed before the first fission foil. Gd was chosen to filter out the neutrons below ~0.1 eV because it has a high capture cross-section at thermal energies and the capture cross section is changing rapidly near 0.1 eV. An approximately 20 mg/cm^2 natural Gd filter can be used to significantly absorb neutrons

below ~0.1 eV while transmitting most of the neutrons above 0.1 eV. Many spent-fuel ponds contain boron in the pool water to absorb thermal neutrons. The thickness of the Gd filter could be reduced to compensate for any boron in the water.

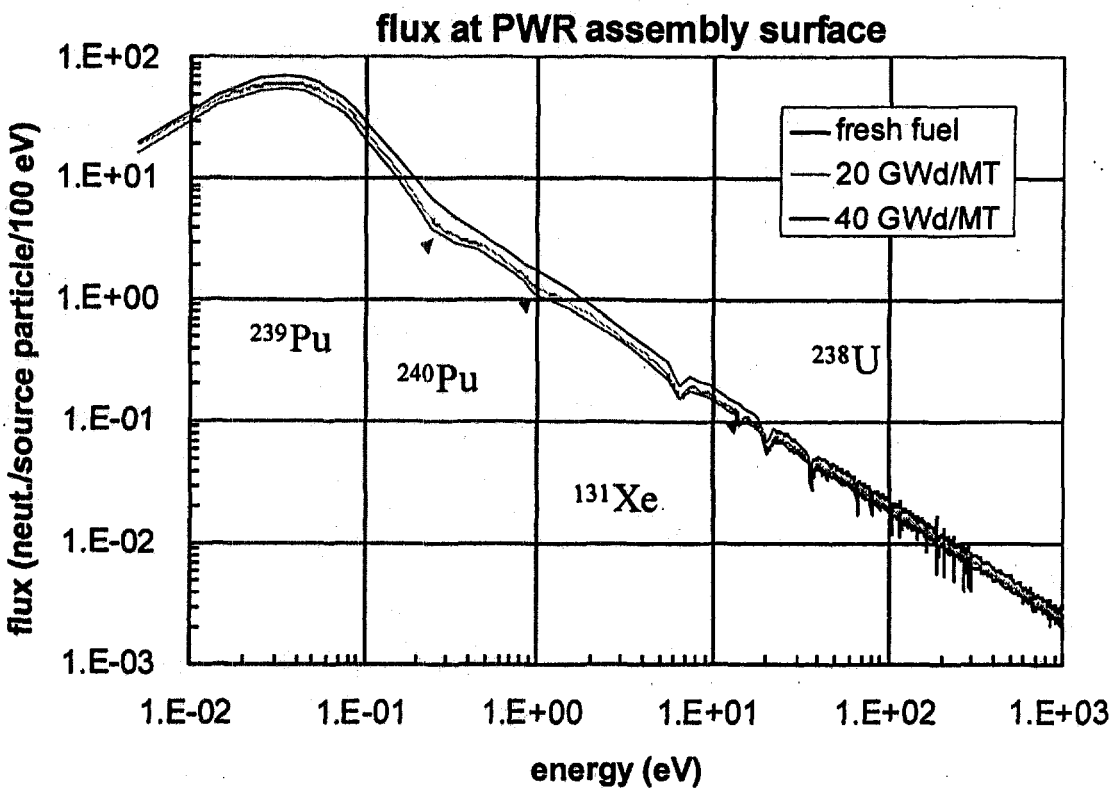

Figure 2. Calculated neutron energy spectra near the surface of a fresh PWR assembly, and near the surface of assemblies with burnups of 20 GWd/MT and 40 GWd/MT.

A natural Cd filter was placed behind the first fission foil, and a second fission foil was placed behind this Cd filter. Cd was chosen as the second filter because its absorption cross-section is rapidly decreasing as the neutron energy increases above ~0.5 eV. An approximately 3 mm thick Cd filter can be used to block most of the neutrons below ~0.5 eV, while allowing most the neutrons above 0.5 eV to pass unhindered. Given the above described arrangement of Gd and Cd filters and two fission foils, the first fission foil will be sensitive to the neutron flux above ~0.1 eV and the second foil will be sensitive to the flux above ~0.5 eV. The difference between the fission rate observed in these two foils will thus be a measure of the neutron flux in the region around the ^{239}Pu resonance centered at ~0.3 eV. Figure 3 shows the neutron energy spectrum at the surface of a 40 GWd/MT PWR assembly after transversing a 21-mg/cm^2 natural Gd filter and a 3-mm natural Cd filter.

Only ~1/5 of the spontaneous Cm neutrons generated in the PWR assembly are slowed to an energy less than 1000 eV at the surface of the assembly. This could create a problem for a poorly designed instrument because 80% of the neutrons have the potential to pass through the instrument, slow down in the surrounding water into the less-than 1000 eV energy region and return through the fission foils inducing an additional signal not

modeled here. These returning neutrons could create a signal in the fission foils that is comparable to the signal generated by the neutrons when they first passed through the foils. This problem can be mitigated by the careful placement of neutron filters near the instrument water interface. The filters/foils and additional absorbers surrounding the instrument would be positioned to minimize the neutrons returning from the surrounding water. The instrument would be designed so that any returning neutrons would give the same signal in both fission foils. The above discussed difference method would thus remove the signals for the returning neutrons.

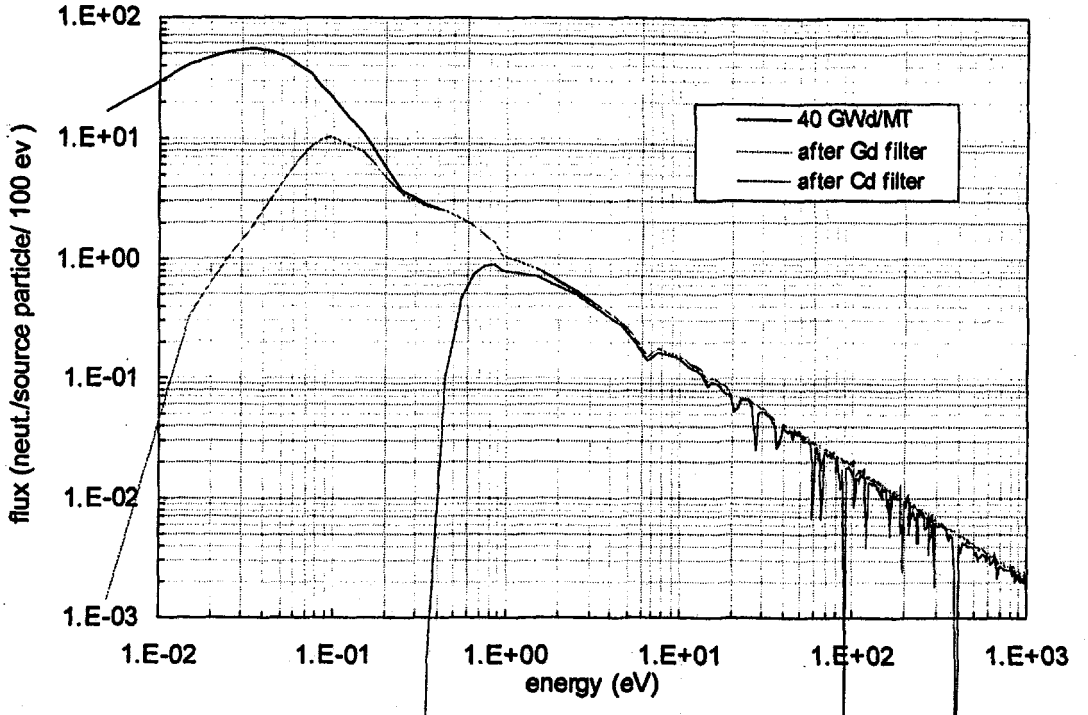

Figure 3. Calculated neutron energy spectra near the surface of a 40 GWd/MT PWR assembly, and the same flux after passing through a 21 mg/cm^2 natural Gd filter and a 3 mm natural Cd filter.

The purpose of the present paper is to examine the possibility that the ratio of the total neutron output divided by the difference in the signal from the two fission foils could be used to measure the plutonium content of PWR spent-fuel assemblies. This ratio will be a minimum at low burnups because the low plutonium content at low burnup translates into no flux depression in the 0.1 eV to 0.5 eV energy region. At high burnups this ratio will be higher because the relative difference in the signals from the two fission foils will be decreased by the flux depression around ~0.3 eV generated by the absorption of neutrons by the ^{239}Pu in the spent-fuel.

The best fission foil for the measurement of the ^{239}Pu flux depression at 0.3 eV would be a ^{239}Pu fission foil because it naturally has a strong fission resonance at the exact location of the absorption resonance producing the flux depression at 0.3 eV. However, because of potential manufacturing and shipping problems that would arise with ^{239}Pu fission foils, calculations are presented here for both ^{239}Pu and ^{235}U fission foils. Figure 4 shows the ratio of the total neutron output divided by the difference between the observed fission

rates in the two fission foils as a function of the total Pu content of PWR assemblies. The solid blue line shows the result with ^{239}Pu fission foils and the dashed red line shows the result with ^{235}U fission foils. The symbols show the calculations for 0, 20, and 40 GWd/MT. As expected the ^{239}Pu foils show the greatest sensitivity to the plutonium content of the assemblies. The ratio of the total neutrons to the difference in the signal from the two foils has been normalized to one for fresh fuel. The dependence of the ratio on the plutonium content (or burnup) of PWR assemblies is 35% greater with ^{239}Pu fission foils. The real advantage to using ^{239}Pu foils will be even greater than this because the rate in the first ^{239}Pu foils with be more than a factor of two larger than the rate in a similar thickness ^{235}U foil. Perhaps more importantly, the first ^{239}Pu foil will give a signal much larger than the second foil due to the strong 0.3 eV resonance in ^{239}Pu, while the ^{235}U foils will give more comparable signals in both foils. For these reasons the ^{239}Pu foils will return smaller counting statistics errors in a fixed measurement time.

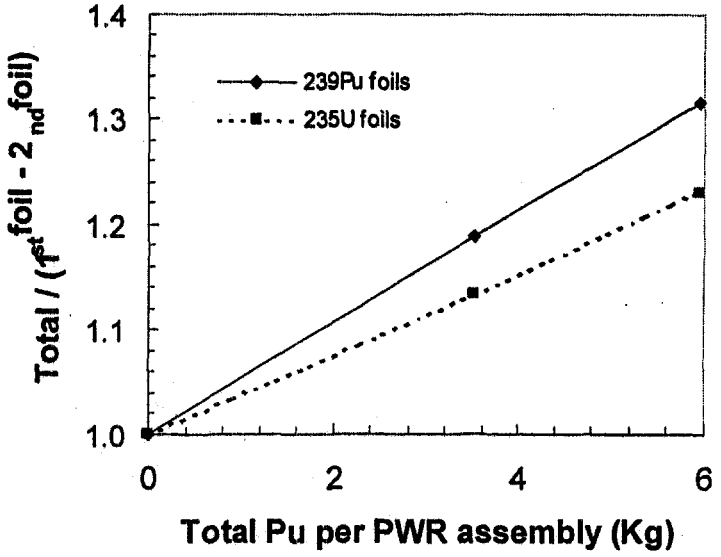

Figure 4. The ratio of the total neutron output divided by the difference between the observed fission rates in the two fission foils as a function of the total Pu content of PWR assemblies.

When all Am and Np, and all fission fragments are removed from the 40 GWd/MT calculations the ratio of the total neutron output divided by the difference in the signal from the two foils decreases by 21% and 28% for the ^{239}Pu and ^{235}U foils cases, respectively. As expected the ^{239}Pu foils give a signal more dominated by the plutonium in the assembly. These calculations show that the proposed neutron resonance absorption signal is dominantly due to the plutonium in the spent-fuel. Additional calculations should be performed to determine which specific isotopes are generating the additional 21% and 28% signals in the ^{239}Pu and ^{235}U foils. The only removed isotope with a half-life of less than several hundred years is ^{153}Eu with a half-life of 52.1 years. This fission fragment will not create a cooling time dependence of the neutron resonance absorption signal because it has no resonance structure near 0.3 eV. The neutron resonance absorption signal that is not directly due to plutonium does not create a significant problem because these non-plutonium isotopes will be co-located with the plutonium and are responsible for only ~1/4 of the signal.

One possible source of a cooling-time dependence of the neutron resonance absorption signal is the 14.7-year half-life of ^{241}Pu. ^{241}Pu has a resonance at ~0.3 eV that has approximately half the strength of the corresponding ^{239}Pu resonance. At high burnups ^{241}Pu can makeup ~12% of the total plutonium mass. Ignoring, for the moment, that ^{241}Pu decays into ^{241}Am, we estimate that the removal of half of the ^{241}Pu over an ~15 year period would cause a reduction of the resonance absorption signal of ~3%. However, ^{241}Pu decays into ^{241}Am which has an absorption resonance at ~0.3 eV which has a strength comparable to the corresponding ^{239}Pu resonance. If one takes into account the decay of ^{241}Pu into ^{241}Am, the resonance absorption signal proposed here may, in fact, increase by a few percent in the first 15 years of cooling time. Calculations of the proposed neutron resonance absorption signal should be performed as a function of cooling time to confirm that any such dependence is less than a few percent per decade.

When the distance from the filters/foils to the outer row of fuel pins is increased by an additional 4 mm the proposed neutron resonance absorption signature for the plutonium content a PWR assemblies decreases by ~1/3. This is expected because the mean free path of neutrons in water is ~7 mm and with an additional 4 mm of water between the fuel pins and the detector, a significant fraction of the neutrons will change their energy by scattering off the hydrogen. These calculations indicate that any instrument using the neutron resonance absorption signature proposed in this paper, will have to be placed in contact with PWR assemblies and some method of verifying this contact will have to be included in the measurement procedure.

Conclusions and the Path Forward

The calculations presented here give additional support to the idea that the flux depression near 0.3 eV can be used to determine the plutonium content of PWR spent-fuel. The ratio of the total neutron output divided by the difference between the signals observed in fission foils behind Gd and Cd filters show a significant dependence on the plutonium content of PWR assemblies. This ratio varies by ~15% as the plutonium content varies from 2 kg to 6 kg. To enable the proposed technique to work, the neutron resonance absorption instrument will need to be reliably placed in contact with the outer row of fuel pins. In addition to this requirement, the fission rates in the two fission foils and the total neutron output would need to be measured with uncertainties less then 2%. For high burnups the neutron source term is so strong that counting statistics will not be the limiting factor when attempting to reduce the measurement uncertainties. Additional thought needs to be given to systematic errors associated with uncertainties in the placement of instruments relative to assemblies while several meters underwater. For low burnup assemblies the limiting factor will be the counting statistics. Calculations should be performed to determine the absolute counting rates. The burnup below which the proposed technique could not be used can be determined given these calculations, a maximum allowable measurement time, and a maximum allowable plutonium uncertainty.

The calculations presented here have confirmed our expectation that higher quality measurements will be obtained with ^{239}Pu fission foils. However, our calculations indicate that a useful signal will be obtainable with less costly and more readily available ^{235}U foils. We thus recommend that any future experimental work and/or possible field tests should focus on ^{235}U foils. If the technique was found to be successful and was accepted for use by safeguard agencies at a future date, it would be a simple matter to perform a cost analysis (dollars per assemblies safeguarded) at that time to determine if one should switch to more costly ^{239}Pu foils.

Additional MCNP calculations need to be performed to model the passage of neutrons through the filters and the signals generated in the fission foils to confirm the simple Excel spreadsheet calculations performed here. These more complete calculations should address the affects of neutrons scattering in the water beyond the instrument and returning to the fission foils.

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