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MONITORING SPENT OR REPROCESSED NUCLEAR FUEL USING FAST NEUTRONS

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ABSTRACT

Attempting to assay the Pu fraction in spent or reprocessed fuel by counting spontaneous fission neutrons immediately runs into the problem that the neutron signal from the spent fuel is dominated by the spontaneous fission neutrons from the ^{242}Cm , ^{244}Cm and ^{240}Pu isotopes. We have found that this problem can be overcome by using fast neutron correlations to measure the number of induced fissions. When the spent fuel is placed inside a polyethylene moderator blanket and lead shield, the number of neutron induced fissions in ^{239}Pu , ^{241}Pu and ^{235}U increases dramatically and this increase can be measured with an array of fast neutron counters. In the case of spent fuel, potential difficulties arise because of the need to shield the detectors from the very high gamma ray flux. However the gamma ray flux can be exponentially attenuated by using a lead shield about 1 mean free path thick for fast neutrons, which would still allow a fast neutron signal sufficient to allow one to determine the total amount of ^{239}Pu , ^{241}Pu and ^{235}U .

INTRODUCTION

A key fact about fissile material is that a sufficient quantity of the material can produce chains of fissions, including some very long chains. These fission chains will give rise to distinctive time correlations in a neutron detector placed near the fissile material. These correlations are measurable and can be analyzed to infer attributes of the fissile material including fissile isotope masses, assembly neutron multiplication, time scale for fission chain evolution, neutron escape times, and moderation time scales. It turns out that the correlation signal is very robust with respect to background and to depletion of neutrons due to absorbing material.

Our contribution to this field is the development of methods for using neutron and γ -ray correlations measured with fast neutron counters in order to obtain a detailed understanding of fissile assemblies containing plutonium (Pu) and highly enriched uranium (HEU). We are currently working with neutron detectors consisting of arrays of tens of segmented liquid scintillator cells capable of detecting fast neutrons and γ -rays with a time resolution of a few nanoseconds and about 10% efficiency for neutron energies above 1.25 MeV. This efficiency makes this kind of array a good candidate to replace ^3He tubes, which suffer from a diminishing supply of ^3He . With this kind of time resolution, arrays of liquid scintillators can directly see the neutrons and gamma rays emitted in fission chains. We have been able to show that this allows us to obtain much more detailed information about fissile assemblies than can be obtained using ^3He detectors, which are primarily useful for detecting slow neutrons. Indeed observation of fission neutrons with ^3He detectors typically requires surrounding the ^3He tube with a moderating material in order to slow the fast fission neutron down to thermal or epithermal energies. This need to thermalize fission neutrons introduces a moderation time scale into the neutron correlations which in many situations of interest obscures the fission chain evolution time scales which we have found useful for analyzing the composition and structure of fissile assemblies.

Our analysis of fissile assemblies using fast neutron counting is informed by a general theory of the space-time development of neutron correlations in a fissile assembly due originally to Feynman and

Schrödinger, and our work is driven by continuous refinements of this theory. We have produced results with many applications, from simple detection of Pu and HEU to characterization of the mass, composition, size, geometry, shielding, and other properties of fissile assemblies. These capabilities have allowed us to solve a wide range of problems in the fields of emergency response, non-proliferation, reactor safeguards, treaty verification, distinguishing threat from non-threat when searching for SNM, etc.

HISTORICAL BACKGROUND

The basic theory of neutron fluctuations and time correlations in near critical assemblies of fissionable materials was developed during WWII by R. Feynman and E. Schrödinger. The most important prediction is that a modest amount of multiplication leads to long fission chains and highly non-Poisson neutron count distributions. Feynman and de Hoffmann (1944). developed a formula for relating the excess neutron intensity fluctuation relative to Poisson distribution

$$Y = \frac{\langle c^2 - \bar{c}^2 \rangle}{\bar{c}} - 1$$

in a homogeneous sub-critical assembly to the neutron multiplication M and neutron lifetime $1/\alpha$ inside the assembly. In particular, the “Feynman correlation function” $Y_{2F} \equiv Y/2$ is given by

$$Y_{2F} = \varepsilon \left(M - \frac{M-1}{\bar{\nu}} \right) \left(\frac{\nu_{2s}}{\bar{\nu}_s} + (M-1) \frac{\nu_{2i}}{\bar{\nu}_i} \right) \left[1 - \frac{1 - e^{-\alpha t}}{\alpha t} \right], \quad (1)$$

where ε is the detection efficiency and ν and ν_2 are the average numbers of neutrons and neutron pairs in a single fission [1]. The indices s and i refer to spontaneous and induced fissions. This formula agreed with measurements of Y for the “Water Boiler Reactor” carried out at Los Alamos in 1944 by E. Segre.

If the detection efficiency ε is unknown, then Eq. (1) must be supplemented with additional information in order to determine the multiplication. One possibility is to use 3-neutron correlations [2]. In the decades following WW II determining the multiplication of sub-critical assemblies using ^3He neutron detectors to measure the 2- and 3-neutron correlations became a cottage industry at Los Alamos. However, because ^3He neutron detectors can only see thermal neutrons it is difficult to see individual fission chains or distinguish multiple time scales in a non-homogeneous assembly. These deficiencies have been overcome with development of neutron detectors that can directly detect fission neutrons with nanosecond timing.

FAST NEUTRON COUNTING

In 2008, W. Stöfl et al built a liquid scintillator array that uses liquid xylene and has the ability to count individual MeV neutrons and γ -rays with nanosecond timing resolution as described in [3]. A number of experiments have been carried out with this array and its descendants that demonstrate its usefulness for directly seeing the multiple neutrons emitted in a single fission chain.

One type of problem where fast neutron counting has turned out to be particularly useful is the analysis of non-homogeneous fissile material assemblies, for example a fissile material assembly surrounded by a layer of moderating material. The neutron multiplication M of a fissile assembly surrounded by an external moderator will be larger than that of the “bare” fissile assembly because neutrons that are thermalized in the external moderator can be reflected back into the fissile assembly, producing

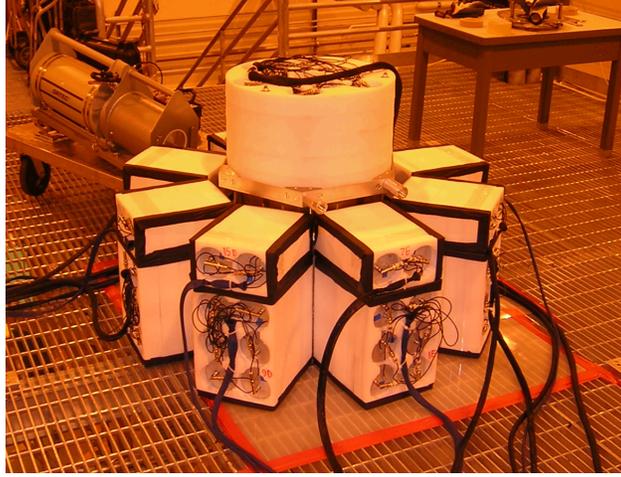


Figure 1. Liquid scintillator array with fast electronic readout used in LLNL experiments

additional fission chains. If the assembly contains isotopes with a large thermal neutron cross-section, e.g. ^{235}U or ^{239}Pu , the increase in the multiplication can be significant. The Feynman 2-neutron correlation function including these “restart” fissions is given by:

$$Y_{2F} = \varepsilon \left(M - \frac{M-1}{\bar{\nu}} \right) \left[\frac{\nu_{2s}}{\bar{\nu}_s} + (M_0 - 1) \frac{\nu_{2f}}{\bar{\nu}_f} + (M - M_0) \frac{\nu_{2th}}{\bar{\nu}_{th}} \right] \left[\left(\frac{M_0}{M} \right)^2 \left(1 - \frac{1 - e^{-\alpha_1 t}}{\alpha_1 t} \right) + \left(1 - \frac{M_0^2}{M^2} \right) \left(1 - \frac{1 - e^{-\alpha_2 t}}{\alpha_2 t} \right) \right] \quad (2)$$

i.e. Y_{2F} is the sum of 2 terms of the same form as the Feynman-de Hoffmann formula (1), except that the induced fission term is now the sum of contributions from fast and slow neutrons. The indices f and th refer to fast and thermal induced fissions. This formula works particularly well in cases where the fast time scale $1/\alpha_1$ and restart time scale $1/\alpha_2$ are very different. This happens to be the case, for example, when a homogeneous fissile material is surrounded by a distinct thin moderating shell.

ASSAYING THE PLUTONIUM IN MOX FUEL

One example of a situation where one might hope that the formula (2) is useful is a nuclear fuel assembly surrounded by a shell of moderating material such as polyethylene. We have obtained evidence that this expectation is correct by carrying out Monte Carlo calculations using MCNP for simulated fuel assemblies surrounded by a cylindrical array of liquid scintillators.

The geometry used in the calculations is shown in Fig. 2. We assumed that the nuclear fuel (indicated in red) has a square cross-section of side 14 cm and is 1 m long. The polyethylene moderator (indicated in blue) was assumed to be a cylindrical shell 5 cm thick over the length of the assembly. The cylindrical detector array was 1.4 m long and contained 192 10-cm diameter, 7.5-cm deep xylene cells. The actinide compositions used in our simulations are shown in Table 1. In all cases of MOX fuel, the observing time was 20 minutes, while in all cases of spent fuel, it was 5 minutes.

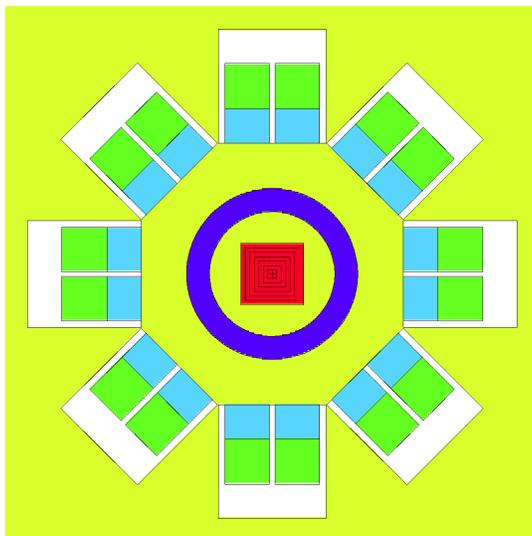


Figure 2. Geometry used in Monte Carlo calculations.

We obtained the 2-neutron correlation function Y_{2F} by analyzing the statistics of the fast neutrons recorded in the cylindrical stack of liquid scintillators. Our strategy is to use the simulated 2-neutron and 3-neutron correlations for time intervals less than a microsecond to determine the rate of generation of spontaneous neutrons and the “bare” multiplication M_0 . In Fig. 3 we show the results of a Monte Carlo calculation of the Feynman correlation function Y_{2F} for 125 kg of MOX fuel with no CH_2 layer.

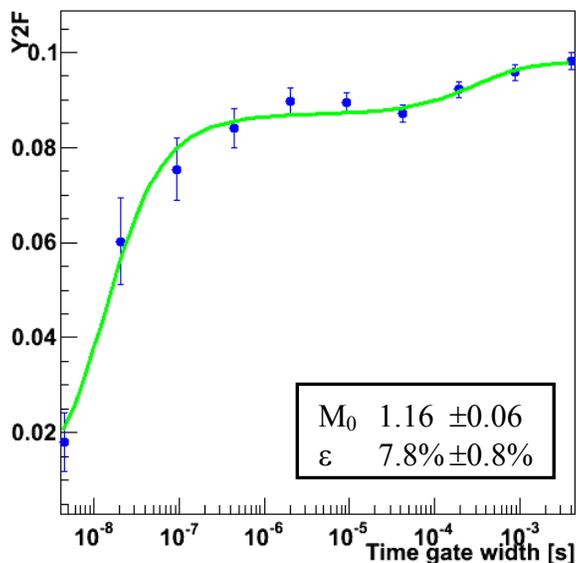


Figure 3. Monte Carlo Y_{2F} for 125 kg MOX fuel with no surrounding CH_2 moderator

Our simulated MOX fuel contained no ^{242}Cm or ^{244}Cm , so the predominant source of spontaneous fission neutrons is ^{240}Pu . The values for the detection efficiency and “bare” multiplication we obtained from our Monte Carlo calculations of Y_{2F} (shown in Fig. 3) and Y_{3F} for the 125 kg of MOX fuel with no external CH_2 moderator are $\varepsilon = 7.8\%$ and $M_0 = 1.16$. Minit was used to fit the independent Y_{2F} data

points using Eq. (2), and to determine all the standard deviations shown in the figures. The presence of the hydrogen-rich xylene detector cells close to the fuel acts as a neutron moderator and generates a small second shoulder at a longer time scale typical of neutron thermalization. This second shoulder is related to neutrons “bouncing off” the detector array thermalized and fissioning ^{235}U and ^{239}Pu on their return journey. The source strength of spontaneous fission neutrons we infer from the observed count rate, together with the inferred values of M_0 and ϵ , is completely consistent with the spontaneous fission rate expected from the 0.6 kg of ^{240}Pu in 125 kg of MOX fuel. One should note that in MOX fuel, (α, n) processes account for approximately 10% of the total neutron source strength. This small contribution was not accounted for in the Monte Carlo simulations.

Given M_0 , the amounts of ^{235}U and $^{239}\text{Pu} + ^{241}\text{Pu}$ in the MOX fuel can be inferred from the behavior of the 2-neutron correlation function Y_{2F} for time intervals on the order of milliseconds when the fuel assembly is surrounded by an external CH_2 moderator. Fig. 4 shows the 2-neutron correlations we obtained from our Monte Carlo calculations when the MOX fuel is surrounded by a 5-cm thick CH_2 layer. Assuming $M_0 = 1.16$, the detection efficiency and total multiplication M obtained from the Monte Carlo calculation of Y_{2F} for the MOX fuel with a 5cm CH_2 layer are $\epsilon = 3.5\%$ and $M = 1.54$. It is expected the bare multiplications are the same in the moderated and unmoderated cases. Again, the source strength inferred from the observed count rate is consistent with the spontaneous fission rate expected from the 0.6 kg of ^{240}Pu in 125 kg of MOX fuel.

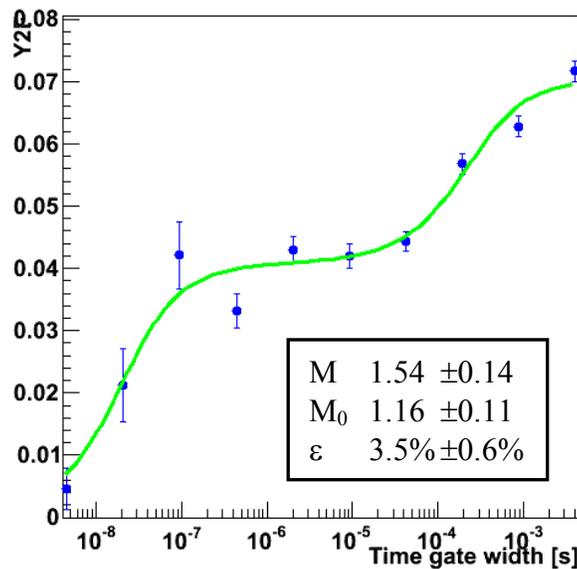


Figure 4. Monte Carlo Y_{2F} for 125 kg MOX fuel surrounded by a 5 cm thick CH_2 layer

The amount of $^{239}\text{Pu} + ^{241}\text{Pu}$ in the MOX nuclear fuel can be inferred by comparing the asymptotic value for Y_{2F} for moderation times (milliseconds) with the plateau value for Y_{2F} for times on the order of a microsecond. As is evident from Eq. (2), the ratio of these plateau values is $(M/M_0)^2$. The excess of M over M_0 is due to the contribution of fissions in the ^{239}Pu , ^{241}Pu , and ^{235}U induced by thermalized neutrons. The value $M = 1.54$ we obtained is consistent with the amounts of these isotopes assumed in the simulation.

From a safeguards perspective, one way of masking the diversion of Pu would be to replace the Pu with a small amount of ^{252}Cf . If the 4 kg of Pu in our 125 kg of MOX fuel is replaced by 25 μg of ^{252}Cf , the spontaneous neutron rates will be approximately the same. Fig. 6 shows the results of Monte Carlo calculations for the 2-neutron Feynman correlation functions without any CH_2 . When compared to Fig. 3, it can be seen that when ^{252}Cf replaces Pu, the Y_{2F} curve becomes flatter for millisecond times and the second shoulder has almost disappeared. Since a total of 3.4 kg ^{239}Pu and 0.2 kg ^{241}Pu were removed from the fuel, the only fissile material left that can thermally induce fissions from the neutrons bouncing off the xylene detector cells is the 0.7 kg ^{235}U . While fast neutrons still induce many fissions in ^{238}U , the fraction of thermal induced fissions has decreased significantly.

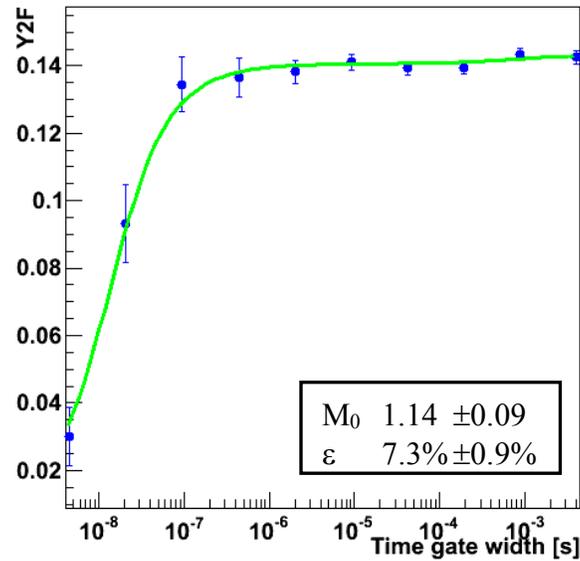


Figure 6. Monte Carlo Y_{2F} for 125 kg MOX fuel with no CH_2 layer and ^{252}Cf replacing the Pu.

The result of the same calculation as Fig. 6 but with a 5 cm layer of CH_2 surrounding the fuel is shown in Fig. 7. The multiplication inferred from the ratio of the long-time plateau to the short time plateau is 1.27 versus 1.54 in the case when Pu is present (Fig. 5). The removal of all the Pu is reflected in the decrease of the multiplication.

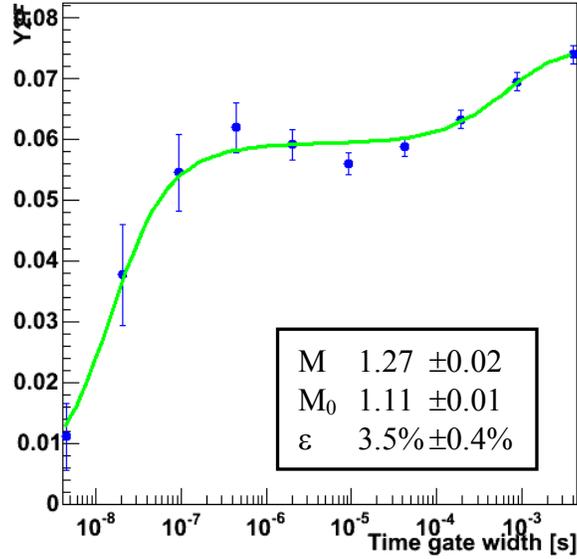


Figure 7. Monte Carlo Y_{2F} for 125kg MOX fuel surrounded by 5 cm CH_2 and ^{252}Cf replacing the Pu.

TABLE I. Actinide masses used in simulations (kg)

Isotopes	MOX fuel	Spent fuel
^{235}U	0.7	20
^{238}U	105	125
^{239}Pu	3.4	1.4
^{240}Pu	0.6	0.5
^{241}Pu	0.2	0.2
^{241}Am	None	0.04
^{242}Cm	None	0.002
^{244}Cm	None	0.004

ASSAYING THE Pu AND ^{235}U IN SPENT REACTOR FUEL

The case of spent fuel differs from MOX fuel in that the predominant source of spontaneous fission neutrons is ^{242}Cm and ^{244}Cm rather than ^{240}Pu . In addition, a thick high Z shield must be placed between the liquid scintillator array and the fuel in order to shield the liquid scintillator array from the enormous γ -ray background from the fission products in the spent fuel. Our hope for using neutron correlation techniques to analyze the ^{235}U and Pu content in spent fuel lies in the idea that a high Z shield that is thick enough to reduce the γ -ray flux to acceptable levels, might still be thin enough to measure fission neutron correlations.

In Fig. 8, we show the results of Monte Carlo calculations of Y_{2F} and Y_{3F} for a section of a simulated assembly of fuel rods with an assumed composition typical for spent reactor fuel with 33000 MWd/ton burn-up and 150 days of cooling, not surrounded by any CH_2 . We assumed that the liquid scintillators could only view a 1 m section of the fuel rod assembly, and that they were shielded with 10 cm of Pb. 10 cm of lead corresponds to about 1 neutron mean free path for fission neutrons. The (α, n) processes,

which contribute 10% to the total neutron source strength, were not accounted for in the Monte Carlo simulations.

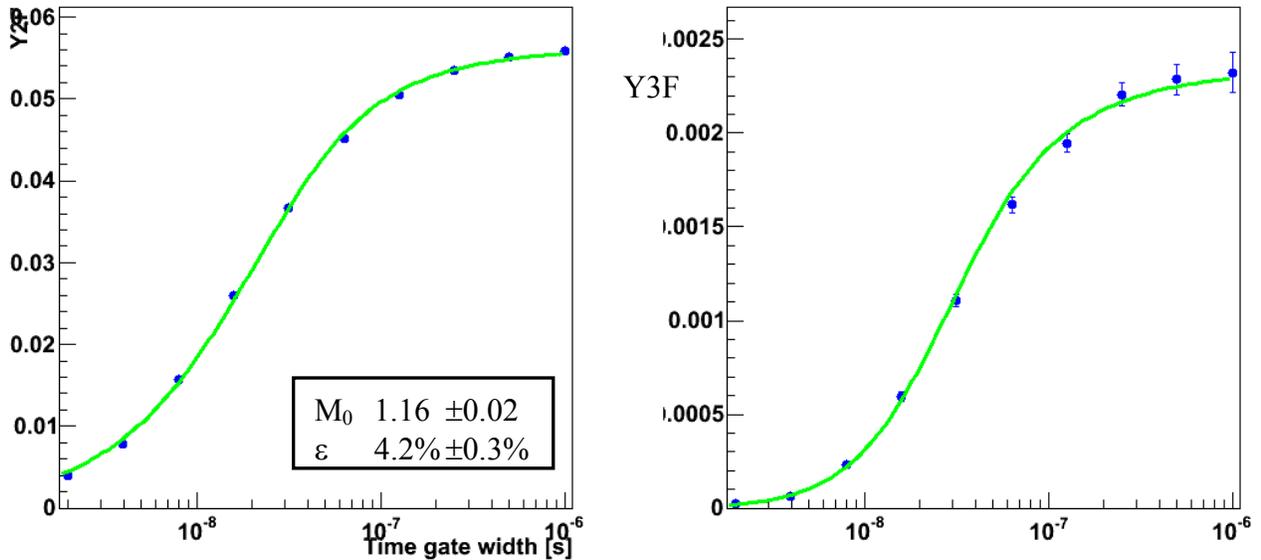


Figure 8. Y_{2F} and Y_{3F} for spent fuel shielded with 10 cm Pb and no CH_2 .

As was the case with MOX fuel, we can use Y_{2F} and Y_{3F} to obtain the spontaneous neutron rate and the bare multiplication M_0 of the spent fuel assembly. The detection efficiency ϵ was determined to be approximately 4.2% and the bare multiplication $M_0=1.16$. The observed count rate in this case was $3.5 \cdot 10^6 \text{ sec}^{-1}$, which is consistent with the assumed amounts of ^{242}Cm and ^{244}Cm . In Fig. 9, we show the results of Monte Carlo calculations of Y_{2F} when the spent fuel is shielded with 5 cm of CH_2 and 10 cm of Pb. The total multiplication M is 1.57. As was the case with MOX fuel, the excess of M over M_0 is due to the fissions induced in $^{239}\text{Pu} + ^{241}\text{Pu} + ^{235}\text{U}$ by thermal neutrons.

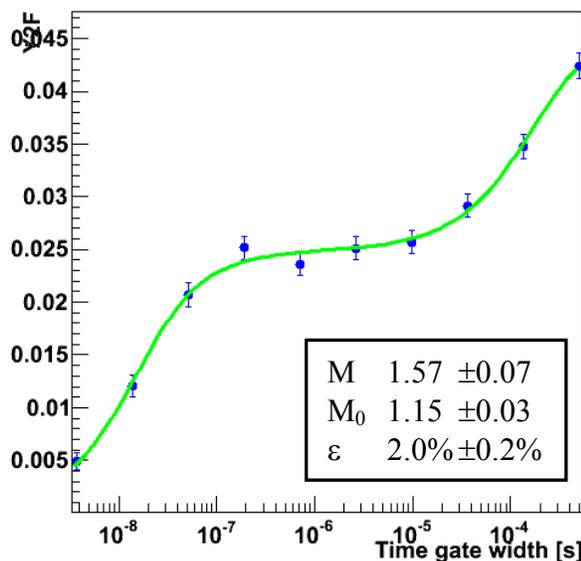


Figure 9. Y_{2F} for spent fuel shielded with 5 cm CH_2 and 10 cm Pb

In order to get some idea whether it is possible to use our methods to determine the amounts of Pu and ^{235}U remaining in spent fuel rods after they have been removed from service, we have repeated the spent fuel Monte Carlo calculation with the Pu and ^{235}U removed. The results for Y_{2F} are shown in Fig. 10.

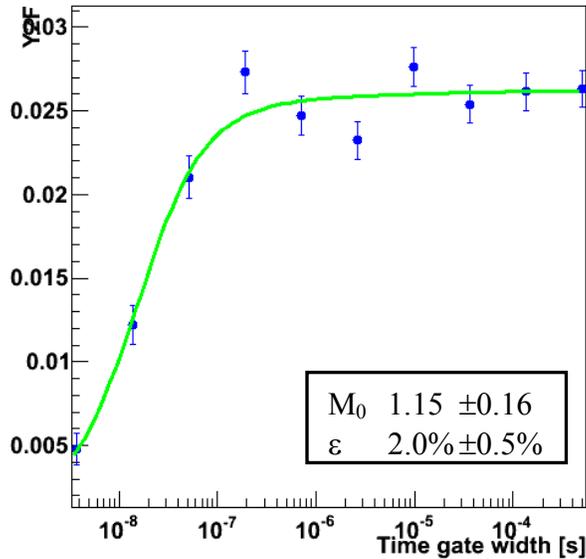


Figure 10. Y_{2F} for same fuel configuration, but with the Pu and ^{235}U removed

The multiplication in this case is 1.15 versus 1.57 with no diversion of Pu and ^{235}U . The difference between the average count rate with and without Pu and ^{235}U is a factor 1.5. The ratio of multiplications tells us that $\approx 30\%$ of the increase in count rate is due to induced fissions in the $^{239}\text{Pu} + ^{241}\text{Pu} + ^{235}\text{U}$. In principle, by using Monte Carlo calculations to map the multiplication M to the combined mass of these isotopes, one could determine this mass by measuring Y_{2F} .

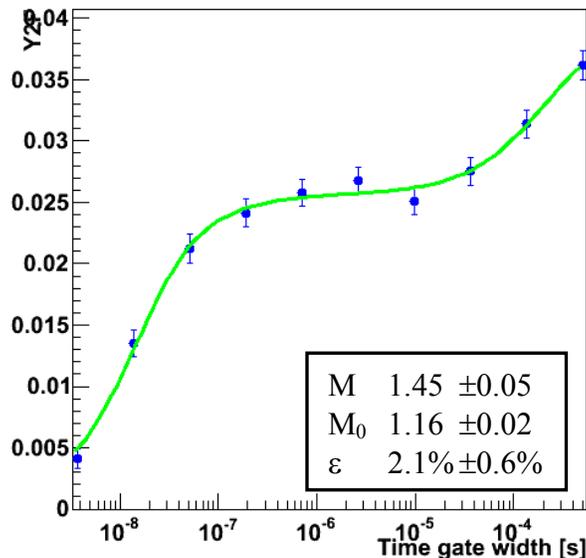


Figure 11. Y_{2F} for spent fuel shielded with 5 cm CH_2 and 10 cm Pb, but containing no Pu.

In Fig. 11, we show a calculation of Y_{2F} for the shielded spent fuel with only the Pu removed. The second shoulder in Y_{2F} , is relatively lower than the one in Fig. 9, which indicates that the total multiplication M is reduced. We estimate the total multiplication M in this case to be 1.45 versus 1.57 with no diversion of Pu. Although the results do appear to be sensitive to the amount of Pu, it is not clear at this point whether we can determine the individual masses of the ^{239}Pu , ^{241}Pu , or ^{235}U isotopes. This will require further study.

CONCLUSIONS

Our Monte Carlo simulation results suggest that fast neutron counting techniques ought to be very useful for assaying the amounts of fissile materials in spent and reprocessed nuclear fuel. In particular the short time behavior of Y_{2F} and Y_{3F} can be used to assay the amounts of ^{240}Pu in the reprocessed fuel and Cm in spent fuel. If the reprocessed fuel is surrounded with a few inches of moderator, then the long time behavior of Y_{2F} can be used to quite accurately determine the absolute amount of $^{239}\text{Pu} + ^{241}\text{Pu}$ in the reprocessed fuel. In the case of spent fuel the neutron counting statistics are somewhat marginal due to the need to use a high Z shield thick enough to block the enormous γ -ray flux. However, with a Pb shield about 1 neutron mean free thick, the statistics are good enough to at least estimate the total amount of $^{239}\text{Pu} + ^{241}\text{Pu} + ^{235}\text{U}$ in the spent fuel. Whether we can separately identify the amounts of Pu isotopes will require further study.

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