



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Distinguishing Plutonium Metal from Plutonium Oxide Using Fast Neutrons, Preliminary Experimental Results

J. M. Verbeke, G. F. Chapline

November 7, 2012

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.



LLNL-TR-599212

Distinguishing Plutonium Metal from Plutonium Oxide Using Fast Neutrons, Preliminary Experimental Results

Jérôme M. Verbeke, G. F. Chapline
Lawrence Livermore National Laboratory
Global Security

6 July 2012

Contact author: Jérôme M Verbeke, (925) 422 8337

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

Auspices

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Distinguishing Plutonium Metal from Plutonium Oxide Using Fast Neutrons, Preliminary Experimental Results

Jérôme M. Verbeke, G. F. Chapline
Lawrence Livermore National Laboratory
Global Security

6 July 2012

1 Introduction

This report is the experimental counterpart of a paper [3] in which algorithms were developed (a) to distinguish chemical forms of plutonium, and (b) to extract the multiplication of a plutonium measurement using liquid scintillators for fast neutron measurements. While we had tested our algorithms using Monte Carlo simulations in Ref. [1], we now propose to apply the same algorithms to experimental measurements.

2 Description of the objects measured for the reference neutron spectra

The algorithms developed previously [1] relied upon the knowledge of two important neutron energy spectra: (a) the energy spectrum of fission neutrons, and (b) the energy spectrum of $O(\alpha,n)$ neutrons. If these two reference neutron energy spectra are sufficiently distinct, an arbitrary measured neutron energy spectrum should be re-constructable by combining the two reference spectra weighed appropriately.

To produce pure neutron spectra for both fission neutrons and $O(\alpha,n)$ neutrons, we need sources that will primarily produce these neutrons of interest.

2.1 Plutonium metal

For trace amounts of metallic plutonium, the system does not multiply, and the neutrons emitted are dominated by the spontaneous fission of ^{240}Pu . For large and dense quantities of metallic plutonium, plutonium multiplies and the fission neutron rate is dominated by induced fissions in ^{239}Pu . For anything in-between, the fission neutron energy spectrum will be a mix of ^{240}Pu spontaneous fissions and ^{239}Pu induced fissions. Fortunately, nature is good to us, and the fission neutron energy spectra from both ^{240}Pu and ^{239}Pu are very similar, as shown in Fig. 1. Therefore, a Pu metal object of any size will give off neutrons with approximately

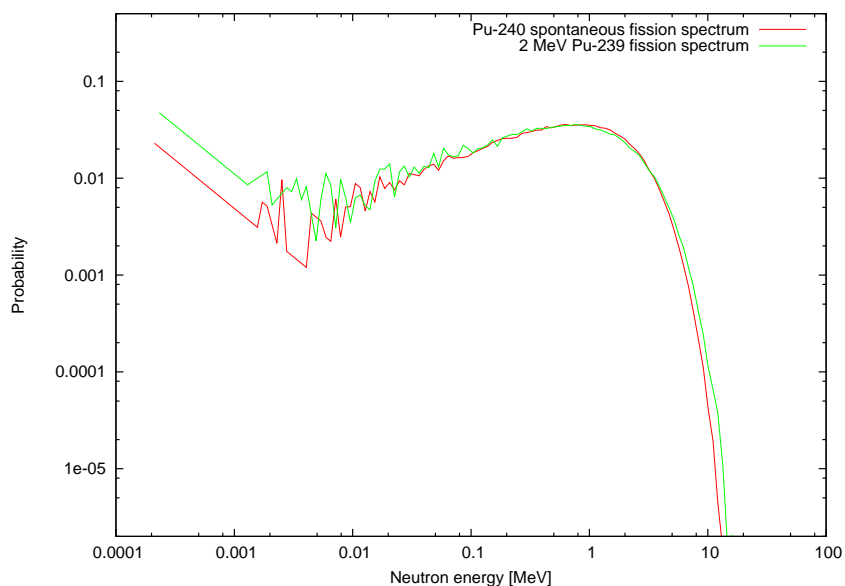


Figure 1: Energy distributions of neutrons from ^{240}Pu spontaneous fission (red) and ^{239}Pu fission induced by 2 MeV neutrons (green). Data from Ref. [2]

the same energy distribution.

The object considered to measure a pure fission spectrum of plutonium with our array of liquid scintillators is a bare plutonium ball of density $\rho = 15.92 \text{ g/cm}^3$. Table 1 lists the isotopic composition of the plutonium ball. The plutonium metal ball was located in the middle of the array of liquid scintillators depicted in Fig. 2. The spectrum of energy deposited in the liquid scintillator by the fission neutrons is shown in red in Fig. 3

Table 1: Weight fractions of isotopes composing the plutonium metal ball.

Isotope	weight fraction [wt %]	weight [g]	spontaneous fission neutron yield [n/s]
²³⁸ Pu	0.01107	0.259	670.81
²³⁹ Pu	93.87881	2195.556	87.463
²⁴⁰ Pu	5.95746	139.328	142,115
²⁴¹ Pu	0.12699	2.968	0.1484
²⁴² Pu	0.02600	0.608	1045.8
²⁴¹ Am	0.36948	8.643	10.199
²³⁷ Np	0.01250	0.293	3.3402e-5
Total	100	2347.66	143,929

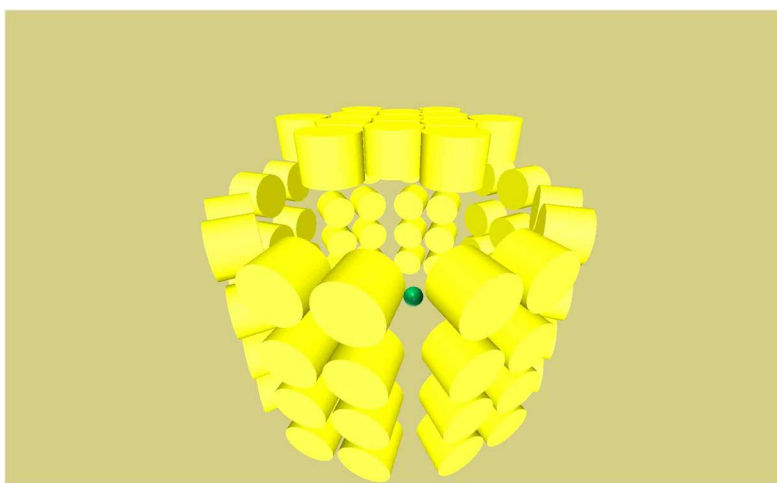


Figure 2: Object in the middle of the 77 liquid scintillator array.

2.2 Plutonium dioxide

For the other chemical form of plutonium, namely PuO_2 , nature is not as easy as for plutonium metal. We saw in Ref. [1, 3] that the ratio of the (α, n) neutron rate to the fission neutron rate, also referred to as the α -ratio is of the order of 0.8. This low α -ratio has two reasons: (a) in the denominator ^{240}Pu is a very strong spontaneous fission neutron source, (b) in the numerator oxygen is a weak (α, n) neutron source. As a consequence, if we were to measure the neutrons coming out of PuO_2 , the majority of them would be plutonium fission neutrons. If we were able to turn off fission reactions for both ^{239}Pu and ^{240}Pu , we could cleanly measure the $\text{O}(\alpha, n)$ neutron energy spectrum — produced by the plutonium decay α -particles. However, while this would be possible in the simulation world, fission reactions can not be turned off in nature.

Since we cannot measure the (α, n) neutron energy spectrum coming from PuO_2 directly, how could we possibly estimate it?

In plutonium, ^{240}Pu yields 1020 n/s/g. On the other side, in HEU, the main source of spontaneous fissions is ^{238}U , which yields 0.0136 n/s/g. While we have a α -ratio of the order of 0.8 for PuO_2 , this difference in spontaneous fission rates translates into an α -ratio of the order of 30 or more for UO_2 (for an uranium composition close to HEU). This means that the vast majority of the neutrons emitted by uranium dioxide are $\text{O}(\alpha, n)$ neutrons. Measuring the spectrum of neutrons coming off of UO_2 would be roughly equivalent to measuring the $\text{O}(\alpha, n)$ neutrons directly, without much pollution from fission neutrons. If we

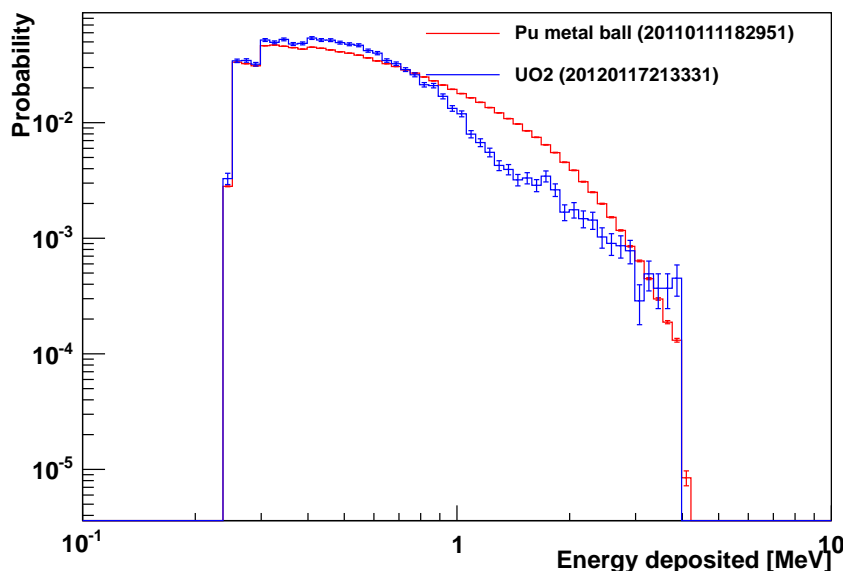


Figure 3: Fast neutron energy spectra for bare plutonium metal ball (red) versus UO_2 objects (blue). The experimental data were taken for 596 s (Pu metal) and 1800 s (UO_2).

can show that the energy spectra of the $\text{O}(\alpha, n)$ neutrons from PuO_2 and UO_2 are similar, we could substitute PuO_2 with UO_2 to estimate the energy spectrum of PuO_2 . Similarly, if we could show that the energies of the α -particles produced by the decay of the most radioactive uranium and plutonium isotopes are similar, there is no reason to believe that these α -particles would produce very different neutron spectra via the $\text{O}(\alpha, n)$ reaction.

Table 2 shows the reactions producing the largest $\text{O}(\alpha, n)$ yields for PuO_2 ¹. These yields were computed using the code SOURCE-4C [4]². By far, the reactions yielding the most (α, n) neutrons are the α -particles from ^{239}Pu on ^{18}O . The 3 α -particles have energies of 5.106, 5.144 and 5.157 MeV and yield 10.1, 13.6 and 66.6 n/sec/cm³. Table 3 shows the same yields for UO_2 ³. For UO_2 , the reactions yielding the most (α, n) neutrons are the α -particles from ^{234}U on ^{18}O . The 2 α -particles yielding the most neutrons have energies of 4.722 and 4.775 MeV and yield 0.0652 and 0.169 n/sec/cm³.

Since the energies of the α -particles dominating the neutron production for PuO_2 and UO_2 are quite different, it is not possible to conclude at this point whether the energy spectra of the $\text{O}(\alpha, n)$ neutrons will be identical. Using the information listed in tables 2 and 3, we can compute the spectra of the neutrons produced using the same code SOURCE-4C and check how different they are. Fig. 4 shows the energy distribution of $\text{O}(\alpha, n)$ neutrons (mainly from ^{18}O) for both PuO_2 (red) and UO_2 (green). The (α, n) neutron spectra from PuO_2 and UO_2 are very close, close enough that we can confidently substitute PuO_2 with UO_2 to estimate the energy spectrum of PuO_2 .

The objects used for measurement of the $\text{O}(\alpha, n)$ spectrum were 3 UO_2 objects of weights 1485.9 (MS-Hexcan-0510), 1463.5 (MS-Hexcan-0601), and 1516.7 grams (MS-Hexcan-0603). They were measured

¹The density of PuO_2 is 3 g/cm³, the plutonium isotopic composition is 0.014 w% ^{238}Pu , 93.5 w% ^{239}Pu , 5.97 w% ^{240}Pu , 0.490 w% ^{241}Pu , 0.0255 w% ^{242}Pu .

²The SOURCE-4C input file for the PuO_2 object is shown in appendix A.

³The density of UO_2 is 10.97 g/cm³, the uranium isotopic composition is given in Table 4. The SOURCE-4C input file is shown in appendix B.

Table 2: α -particle energies and yields, and $O(\alpha,n)$ yields in PuO_2 from major plutonium isotopes, computed using the code SOURCE-4C [4].

Isotope	Target	α energy [MeV]	$\alpha/\text{sec}/\text{cm}^3$	$p(e)$ [n/ α]	n/sec/cm ³
²³⁸ Pu	¹⁸ O	4.431	2.5806E+00	8.9716E-09	2.3152E-08
		4.471	2.8152E+00	9.3722E-09	2.6385E-08
		4.525	3.0498E-01	9.6592E-09	2.9458E-09
		4.566	5.8650E-01	1.0172E-08	5.9661E-09
		4.590	2.8152E+01	1.0487E-08	2.9523E-07
		4.661	9.8531E+00	1.1442E-08	1.1274E-07
		4.663	2.1114E-01	1.1463E-08	2.4204E-09
		4.704	1.1730E+02	1.1964E-08	1.4033E-06
		4.724	5.1612E+01	1.2136E-08	6.2638E-07
		5.015	1.5953E+01	1.4742E-08	2.3518E-07
		5.206	7.0380E+03	1.6610E-08	1.1690E-04
		5.358	2.4633E+05	1.8542E-08	4.5675E-03
		5.456	6.7987E+07	1.9838E-08	1.3487E+00
		5.499	1.6635E+08	2.0410E-08	3.3952E+00
Total:		-	-	-	4.7486E+00
²³⁹ Pu	¹⁷ O	5.106	6.5303E+08	1.3306E-09	8.6892E-01
		5.144	8.5746E+08	1.3737E-09	1.1779E+00
		5.157	4.1624E+09	1.3931E-09	5.7986E+00
Total:		-	-	-	7.8454E+00
²³⁹ Pu	¹⁸ O	5.106	6.5303E+08	1.5400E-08	1.0057E+01
		5.144	8.5746E+08	1.5868E-08	1.3606E+01
		5.157	4.1624E+09	1.6008E-08	6.6632E+01
Total:		-	-	-	9.0295E+01
²⁴⁰ Pu	¹⁷ O	4.264	8.3730E+00	7.5617E-10	6.3314E-09
		4.492	2.6581E+02	8.7053E-10	2.3140E-07
		4.655	6.2465E+02	9.7335E-10	6.0801E-07
		4.864	1.4354E+04	1.1184E-09	1.6053E-05
		5.021	1.1324E+06	1.2625E-09	1.4296E-03
		5.124	3.6017E+08	1.3493E-09	4.8598E-01
		5.168	9.6755E+08	1.4116E-09	1.3658E+00
Total:		-	-	-	1.8532E+00
²⁴⁰ Pu	¹⁸ O	4.264	8.3730E+00	6.9164E-09	5.7911E-08
		4.492	2.6581E+02	9.4904E-09	2.5226E-06
		4.655	6.2465E+02	1.1359E-08	7.0954E-06
		4.864	1.4354E+04	1.3165E-08	1.8897E-04
		5.021	1.1324E+06	1.4773E-08	1.6729E-02
		5.124	3.6017E+08	1.5628E-08	5.6286E+00
		5.168	9.6755E+08	1.6148E-08	1.5624E+01
Total:		-	-	-	2.1269E+01
Total:		-	-	-	1.2698E+02

together. The overall composition of these objects is given in table 4. Combining the neutron yield from $O(\alpha,n)$ reactions given in table 3 with the spontaneous fission neutron yield given in table 4, we get an overall neutron yield of $4166.606/10.97 * 0.26083 + 3.05063 = 99.068 + 3.05063 = 102.12$ n/s for the 3 objects. This value is not infallible, it should only be taken as an estimate, for the α -ratio varies with the

Table 3: α -particle energies and yields, and $O(\alpha,n)$ yields in UO_2 from major uranium isotopes, computed using the code SOURCE-4C [4].

Isotope	Target	α energy [MeV]	α /sec/cm ³	$p(e)$ [n/ α]	n/sec/cm ³
²³⁴ U	¹⁷ O	4.109	1.3515E+00	6.3350E-10	8.5617E-10
		4.151	5.0198E+00	6.6269E-10	3.3266E-09
		4.276	7.7228E+00	7.4645E-10	5.7647E-09
		4.603	3.8614E+04	9.1453E-10	3.5314E-05
		4.722	5.4870E+06	1.0029E-09	5.5029E-03
		4.775	1.3781E+07	1.0419E-09	1.4359E-02
Total		-	-	-	1.9897E-02
²³⁴ U	¹⁸ O	4.109	1.3515E+00	5.5983E-09	7.5660E-09
		4.151	5.0198E+00	5.8791E-09	2.9512E-08
		4.276	7.7228E+00	6.9229E-09	5.3464E-08
		4.603	3.8614E+04	1.0449E-08	4.0349E-04
		4.722	5.4870E+06	1.1875E-08	6.5156E-02
		4.775	1.3781E+07	1.2281E-08	1.6924E-01
Total		-	-	-	2.3480E-01
Total		-	-	-	2.6083E-01

chemical form of the uranium oxide.

Table 4: Sums of the weights of isotopes composing the 3 UO_2 objects (MS-Hexcan-0601U, MS-Hexcan-0510U, MS-Hexcan-0603U).

Isotope	weight [g]	atomic density [atoms/cm ³]	spontaneous fission neutron yield [n/s]
¹⁶ O	498.17	4.94×10^{22}	-
¹⁷ O	0.195	1.82×10^{19}	-
¹⁸ O	1.004	8.84×10^{19}	-
²³⁴ U	31.67	2.15×10^{20}	0.15898
²³⁵ U	3425.5	2.31×10^{22}	$3.5650 \times 10^{-2\dagger}$
²³⁸ U	210.00	1.40×10^{21}	2.856
Total	4166.606	7.42×10^{22}	3.05063

The uranium oxide objects were located in the middle of the array of liquid scintillators depicted in Fig. 2. The spectrum of energy deposited in the liquid scintillator by the neutrons is shown in blue in Fig. 3. It can directly be compared with the spectrum in red for the Pu metal ball.

The UO_2 curve seems to have an inflection point between 1 and 1.5 MeV with a long tail past 1.5 MeV. The tail is likely due to one of 3 effects:

1. The few fission neutrons emitted by both the spontaneous fissions of ²³⁸U and the induced fissions in ²³⁵U. Even through the α -ratio is high for UO_2 , there is a few percents probability that neutrons emitted by the UO_2 object will be fission neutrons. This small probability could be the cause of the long tail for large deposited energies.

[†]This value reflects the spontaneous fission yield of 10.41×10^{-6} n/s/g used by SOURCE-4C for ²³⁵U, instead of 299×10^{-6} n/s/g, which would result in a spontaneous neutron yield of 1.024 n/s.

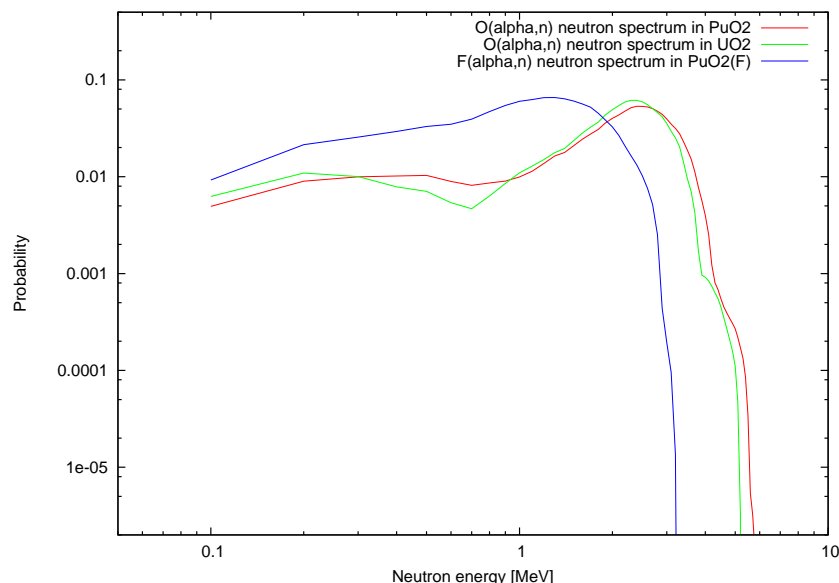
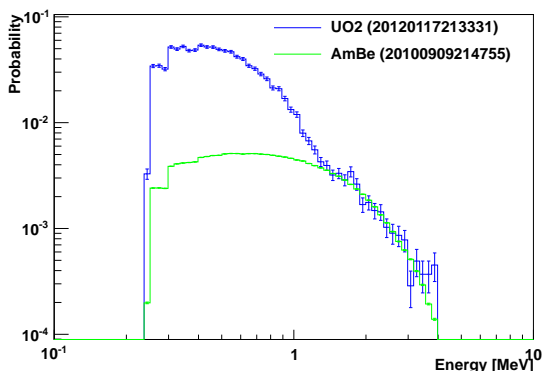


Figure 4: Energy distributions of neutrons from the $O(\alpha,n)$ reactions in PuO_2 (red) and UO_2 (green). Neutron spectrum from the $F(\alpha,n)$ reactions in $PuO_2(F)$ (blue) is given for comparison.

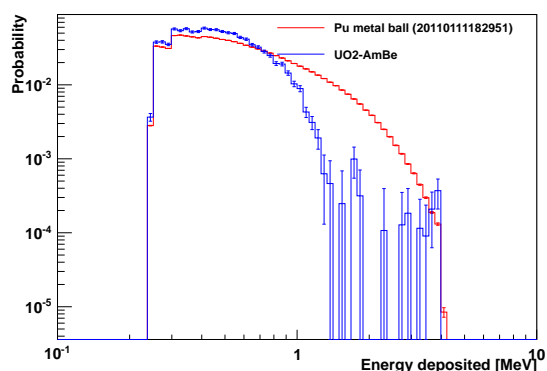
2. The presence of an external source of high-energy neutrons not shielded appropriately, such as an Americium/Beryllium source. One should point out that an Americium/Beryllium source was used 10 minutes after the 2012/01/17 21:33:31 experiment for an experiment where it acted as an interrogating source for UO_2 .
3. The natural cosmic-ray background.

Regarding the first effect, while it is entirely possible for fission neutrons to generate a long tail in the neutron energy spectrum, the tail of the blue distribution in Fig. 3 does not have the same shape as the tail of the Pu metal spectrum in red, which it should resemble if the tail was due to fission neutrons from Pu.

Regarding the second effect, we will check how likely the long blue tail in Fig. 3 is due to the presence of an Americium/Beryllium source. The neutron energy spectrum from a measured bare Americium/Beryllium source is shown in green in Fig. 5(a). It was scaled down to match in height the tail of the UO_2 spectrum. As we can observe, the two curves are within each others error bars from 1.35 MeV all the way up to 3.5 MeV. The scaled down Americium/Beryllium spectrum corresponds to an array detecting Americium/Beryllium neutrons 13.6% of the time. Since the count rate for the measured objects (2012/01/17 21:33:31) was only 13.52 n/s, this means that the detection system could have been measuring 1.84 n/s from the bare Americium/Beryllium source. Given that the Americium/Beryllium source that was in the experimental area had a neutron yield of 6.6×10^4 n/s (and a measured count rate of 4102 n/s when in the liquid scintillator array), this unshielded Americium/Beryllium source would only have to be 14.2 m away from the liquid scintillators to produce a measured count rate of $4102 \left(\frac{0.3}{14.2}\right)^2 = 1.84$ n/s. The room in which the experiment was carried out is about that size, so it is easily believable that the Americium/Beryllium source was in the vicinity of the relatively weak UO_2 object measured during the measurement, and thus the origin of the long tail.



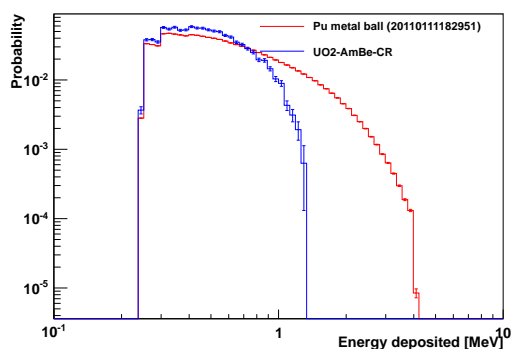
(a) Fast neutron energy spectra for UO_2 objects (blue) and Americium/Beryllium source (green).



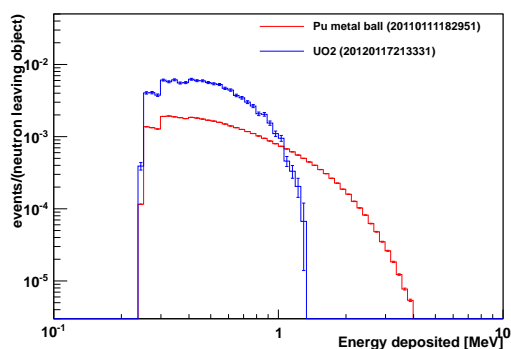
(b) Fast neutron energy spectra for bare Pu metal ball (red) and UO_2 objects where the external Americium/Beryllium source contribution was suppressed (blue).

Figure 5: Fast neutron energy spectra. The experimental data were taken for 1800 s (UO_2) and 579.4 s (Americium/Beryllium source). The measured count rates for the UO_2 objects (2012/01/17 21:33:31) and Americium/Beryllium source (2010/09/09 21:47:55) were 13.52 n/s and 4102 n/s, respectively.

If we subtract from the UO_2 spectrum the spectrum from an Americium/Beryllium source of intensity equal to 13.6% of the total intensity, we obtain the corrected UO_2 spectrum shown in Fig. 5(b). The odd peaks above 1.4 MeV do not quite belong to the rest of the spectrum, and represent less than 3% of the total neutron intensity, i.e. 0.36 n/s. We will assert that they are cosmic-ray induced fast neutrons. Once these odd peaks are removed, one obtains the spectrum shown in Fig. 6(a). One should argue that it is not obvious to



(a) Normalized neutron energy spectra



(b) Number of events per neutron leaving the object.

Figure 6: Bare Pu metal ball (red) and UO_2 objects where the external Americium/Beryllium source and the cosmic-ray induced fast neutron contributions were suppressed (blue). The experimental data were taken for 596 s (Pu metal), 1800 s (UO_2) and 579.4 s (Americium/Beryllium source).

determine whether the long blue tail in Fig. 3 is due to the presence of the Americium/Beryllium source or the cosmic-ray induced neutrons. We have assumed here that most of it was due to the Americium/Beryllium source, because of the shape resemblance. In reality, we should not really pay too much attention to details in the spectrum that produce less than 2 n/s, because such a low rate is definitely at the background level or

even noise level. This is unfortunately what happens when our source of neutrons is so weak to start with. Ideally, a stronger UO_2 source would be better to characterize the $\text{O}(\alpha,n)$ neutron energy spectrum for UO_2 .

The two spectra in Fig. 6(a) resemble their Monte-Carlo calculated counterparts in Fig. 15 of Ref. [1]. A comparison between the experimentally measured and the simulated spectra is shown in Fig. 7. Overall,

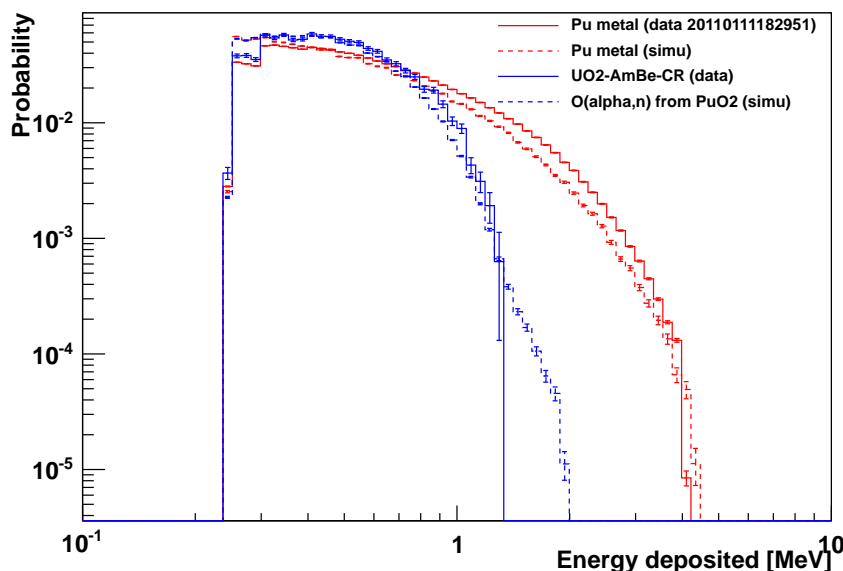


Figure 7: Comparison between experimentally measured (continuous lines) and simulated (dashed lines) fast neutron energy spectra for Pu metal ball (red) and $\text{O}(\alpha,n)$ from UO_2 ⁴/ PuO_2 ⁵ (blue).

the shapes of the experimental and simulated spectra are similar. The simulated spectra are lower than their measured counterparts for high energies, and higher for energies below 400 keV. There are two possible explanations for these differences:

- It is known that the pulse shape discrimination algorithm used experimentally misidentifies more and more neutrons as photons when the energy deposited decreases, resulting in an artificial reduction in the neutron detection efficiency. This dependence of the neutron detection efficiency of the data acquisition system on deposited energy could be the reason why the measured data does not keep up with the simulated data for low energies.
- The quench function used in the simulations to convert the energy deposited by the proton recoils into light-equivalent energy comes from a fairly old paper. While it looks somewhat decent, it should be re-measured with our current data acquisition system and liquid scintillator cells to do a proper comparison of the experimental data with the measured data. This lack of a good quench function is actually the main reason why we did not originally propose to simply reconstruct the energy spectrum from the bare PuO_2 measurement by combining the deposited energy spectra of two separate simulations: one for the Pu metal and a second one for the $\text{O}(\alpha,n)$ neutrons in PuO_2 . The first author is in the process of filling that knowledge gap by re-measuring the quench function. A report should soon be available.

⁴The experimental spectrum is the corrected one shown in Fig. 6(a).

⁵For the simulation, a spectrum of $\text{O}(\alpha,n)$ neutrons from PuO_2 calculated by the code SOURCE-4C [4] was transported to the liquid scintillator cells to produce the spectrum of energies deposited shown.

Above 1.3 MeV, the measured and simulated $O(\alpha,n)$ spectra are substantially different. The simulation predicts a long tail all the way to 2 MeV, while data is lacking that tail. This could well be due to the somewhat arbitrary background (Americium/Beryllium and cosmic-ray) suppression performed above for the data.

3 Reconstruction of an unknown plutonium object

To test out the reconstruction algorithms, we measured a PuO_2 sample with the same liquid scintillator array. The random time gate count distribution for the $1 \mu\text{s}$ time gate along with the count distribution moments are shown in Fig. 8. The values of R_1 , R_{2F} and R_{3F} can be extracted from the graphs.

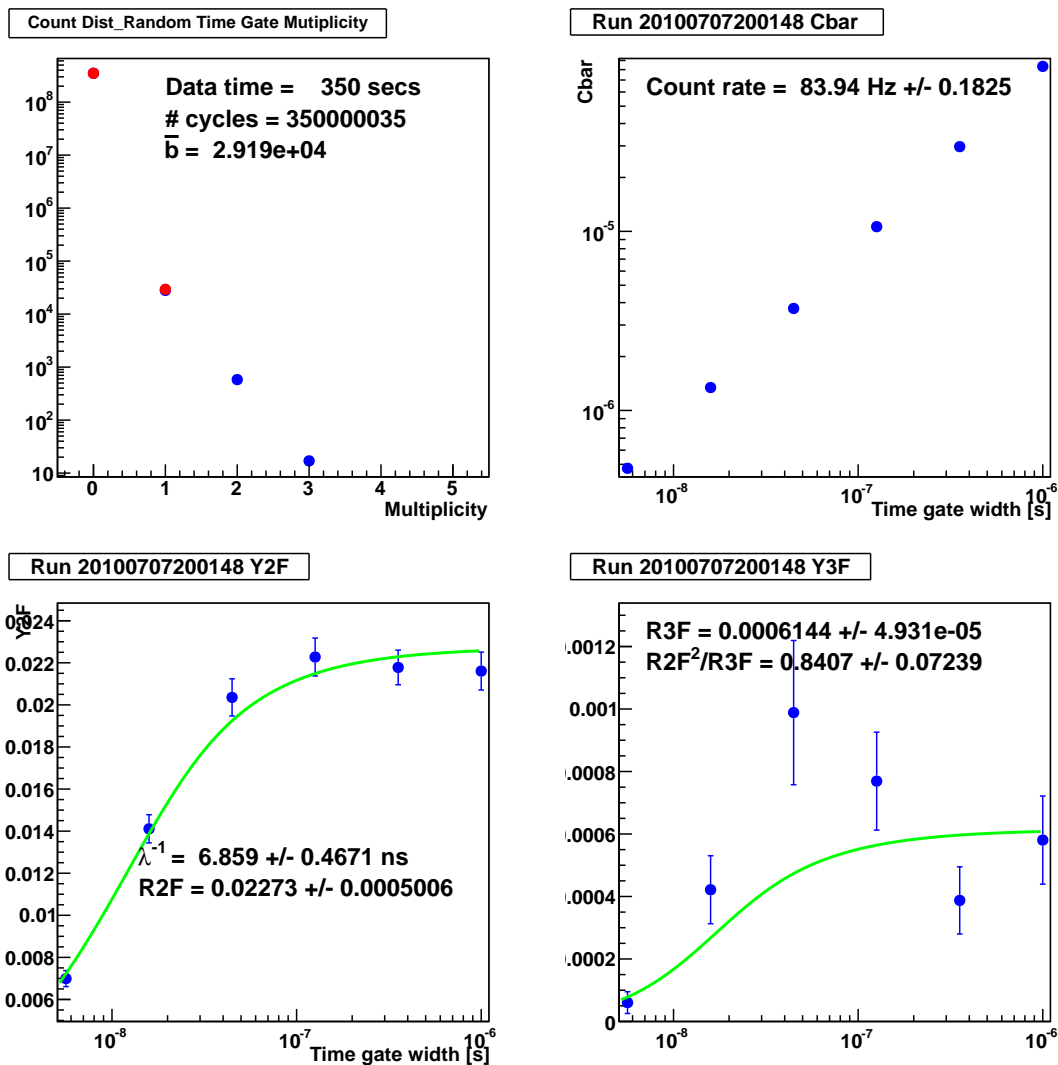


Figure 8: Count distribution $b_n(T)$ for the $1 \mu\text{s}$ time gate, $\bar{C}(T)$, $Y_{2F}(T)$ and $Y_{3F}(T)$ for the PuO_2 sample and T between 5 ns and $1 \mu\text{s}$.

The spectrum of energies deposited by fast neutrons is shown in green in Fig. 9 along with the Pu metal sample (red) and the modified UO_2 (blue) spectra.

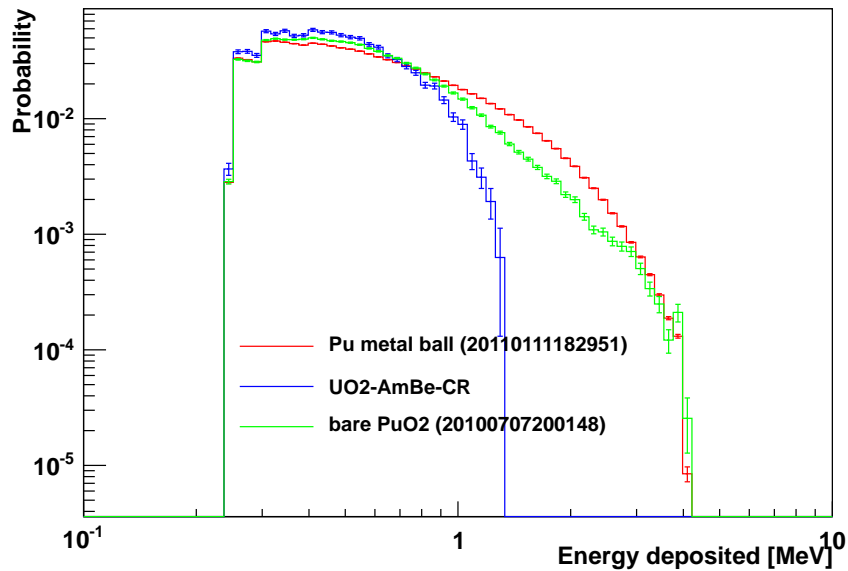
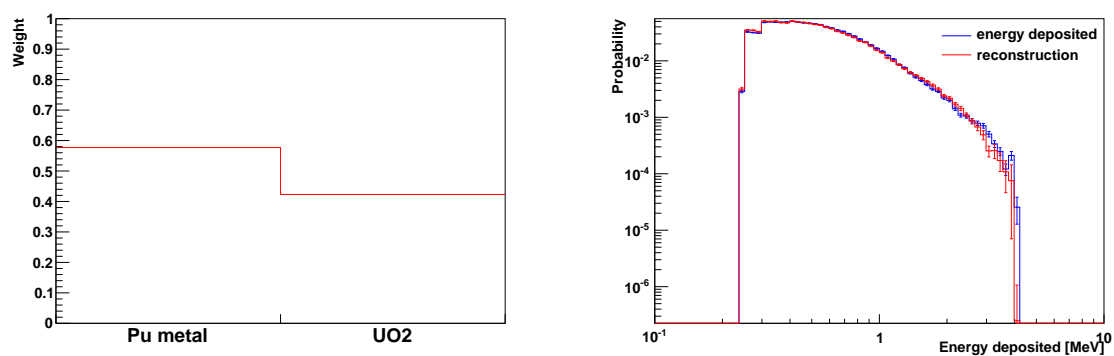


Figure 9: Fast neutron energy spectra for PuO₂ sample (green), along with Pu metal ball (red) and modified UO₂ (blue) spectra. The experimental data were taken for 596 s (Pu metal) and 2521 s (bare PuO₂).

If we assume that the Monte-Carlo determined efficiency ratio $r_\epsilon = \frac{\epsilon_\alpha}{\epsilon_f}$ (see Sec. 6.1 of [1]) is accurate, one can add the red and blue spectra shown in Fig. 9 with appropriate weights to reconstruct the green curve. The set of weights that are optimal for the reconstruction of the bare PuO₂ spectrum is shown in Fig. 10(a). Adding the Pu metal spectrum pre-multiplied by 0.58 to the mostly (α, n) spectrum pre-multiplied by 0.42, the reconstruction of the spectrum produces the red curve in Fig. 10(b), to be compared with the measured PuO₂ spectrum shown in blue.

Setting $\rho = \frac{0.42}{0.58} = 0.74$, the solution to Eqs. 38 and 42 (Ref. [1]) with $M \geq 1$ is $\alpha=0.55$ and $M=1.13$. We can determine the value of ϵ_f to be 3.6% from Eq. 43 (Ref. [1]), while Eq. 20 (Ref. [1]) implies a spontaneous fission source rate of 1160 n/sec. This solution is quite different from the exact value of 519 n/s [5]. Also, our experience tells us that the computed efficiency is lower than expected.



(a) Factors by which the two spectra shown in Fig. 6(a) must be multiplied to reconstruct the spectrum of energies deposited by the fast neutrons emitted by the PuO_2 sample: 0.58 for the Pu metal spectrum and 0.42 for the UO_2 sample spectrum.

(b) Spectrum of energies deposited by fast neutrons in liquid scintillator cells for the PuO_2 sample (blue), along with its reconstruction (red) from the two spectra shown in Fig. 6(a) and the optimal weights.

Figure 10: (a) Factors by which the two spectra shown in Fig. 6(a) must be multiplied to reconstruct the spectrum of energies deposited by the fast neutrons emitted by the PuO_2 sample: 0.58 for the Pu metal spectrum and 0.42 for the UO_2 sample spectrum. (b) Spectrum of energies deposited by fast neutrons in liquid scintillator cells for the PuO_2 sample (blue), along with its reconstruction (red) from the two spectra shown in Fig. 6(a) and the optimal weights.

4 Conclusion

In this report, we tried to apply the algorithm formulated in Ref. [3] to characterize a PuO₂ ball in terms of multiplication, α -ratio and ²⁴⁰Pu mass. The method of using the spectra of energies deposited in the liquid scintillators to determine whether the measured object is Pu metal or PuO₂ proved effective. Because energy spectra can be quickly measured with good statistics, it only takes a few seconds of data to unambiguously determine the chemical form of plutonium. In this respect, this method proved successful.

On the other side, the characterization of the PuO₂ did not fare that well. The method predicted a spontaneous fission rate of 1160 n/sec, while the true rate was 519 n/s. Also, based on the author's experience, it predicted a lower efficiency than expected. We believe these discrepancies arise from the many approximations that were made in the course of this work:

- The spectrum of O(α ,n) neutrons produced by PuO₂ was assumed to be identical to the one produced by UO₂. While this is true in the first order, it might affect the reconstruction of the PuO₂.
- The UO₂ data suggest that the UO₂ measurement was likely made in the presence of an Americium/Beryllium source in the vicinity, which polluted the UO₂ data. We had to clean up the UO₂ energy spectrum in a haphazardous manner. It would be much better to take some clean UO₂ measurements.
- The efficiencies for detecting fission neutrons and O(α ,n) neutrons were determined from simulations, instead of experimental data. Because of the Americium/Beryllium pollution of the data, it was unfortunately not possible to measure the detection efficiencies from the data.

If one could remedy the last 2 points above by doing clean measurements of UO₂, we believe the reconstruction algorithms would work and produce the expected results.

A SOURCE-4C input file for PuO₂

(alpha,n) neutrons from PuO2

```
1 2 1
2 0
  008 .6667
  094 .3333
100 10.0 0.0
6
  0942380 9.3722e17
  0942390 62.331e20
  0942400 3.9633e20
  0942410 3.2394e19
  0942420 1.6789e18
  0952410 4.6991e17
2 4000
  0080170 0.00026
  0080180 0.00134
```

B SOURCE-4C input file for UO₂

(alpha,n) neutrons from UO2 (HX-0603+HX-0510+HX-0603)

```
1 2 1
2 0
  008 .6667
  092 .3333
100 10.0 0.0
3
  0922340 2.15e20
  0922350 2.31e22
  0922380 1.40e21
2 4000
  0080170 0.00026
  0080180 0.00134
```

References

- [1] J.M. Verbeke, G.F. Chapline, “Distinguishing Plutonium Metal From Plutonium Oxide,” LLNL-TR-518451, Lawrence Livermore National Laboratory (2011).
- [2] J.M. Verbeke, C. Hagmann, D. Wright, “Simulation of Neutron and Gamma Ray Emission from Fission and Photofission,” LLNL-AR-228518, Lawrence Livermore National Laboratory (2010).
- [3] J.M. Verbeke, G.F. Chapline, L.F. Nakae, R.E. Wurtz, S.A. Sheets, “Distinguishing Pu Metal From Pu Oxide Using Fast Neutron Counting,” *Proc. INMM 2012*, LLNL-PROC-559636, Lawrence Livermore National Laboratory (2012).
- [4] W.B. Wilson, R.T. Perry, E.F. Shores, W.S. Charlton, T.A. Parish, G.P. Estes, T.H. Brown, E.D. Arthur, M. Bozoian, T.R. England, D.G. Madland, and J.E. Stewart, “SOURCES 4C: A Code for Calculating (alpha,n), Spontaneous Fission, and Delayed Neutron Sources and Spectra,” LA-UR-02-1839, Los Alamos National Laboratory (2002).
- [5] Private communication with Darrell Pugh, October 9, 2012, Lawrence Livermore National Laboratory.