Distinguishing Pu Metal from Pu Oxide and Determining α -ratio using Fast Neutron Counting

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Abstract

We describe a new method for determining the ratio of the rate of (α , n) source neutrons to the rate of spontaneous fission neutrons, the so called α -ratio. This method is made possible by fast neutron counting with liquid scintillator detectors, which can determine the shape of the fast neutron spectrum. The method utilizes the spectral difference between fission spectrum neutrons from Pu metal and the spectrum of (α , n) neutrons from PuO₂. Our method is a generalization of the Cifarelli-Hage method for determining k_{eff} for fissile assemblies, and also simultaneously determines k_{eff} along with the α -ratio.

Keywords: liquid scintillators, fast neutron counting, neutron multiplicity, neutron correlation, k_{eff} , α -ratio, fissile materials

1. Introduction

Methods based on time-correlated neutron signals have long been used to characterize fissile materials. Typically one uses ³He tubes to record the arrival times of neutrons from the fissile source, and then by segmenting the arrival times using time windows of varying width, one can use the statistics of the number of neutron counts in the time window as a function of the width of the time window to characterize the neutron source. Unfortunately the cross-section for neutron capture in ³He is only large enough for the purposes of collecting these counting statistics after the fission neutrons have been thermalized in a moderating material. Therefore the time windows must be at least tens of microseconds long to pick up counts from the same spontaneous fission or chain. In the case of a strong neutron source such as Pu this means that many fission chains will be generated within the time windows. Because the neutron time correlations of interest are generated by individual fission chains, the time correlation information that can be extracted using ³He tubes is diluted, requiring high statistics to disentangle the contribution from separate chains.

Liquid scintillators on the other hand, can directly detect unmoderated fission neutrons, because the reaction used for detection is inelastic scattering of neutrons primarily on hydrogen, producing a recoil proton from which scintillation light is produced promptly. Consequently, counting can now be on the nanosecond time scale. One no

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27 longer needs to open time windows 100's of microseconds long to pick up the cor-
28 relation signal as with ^3He , but only of order 100 nanoseconds. These shorter time
29 windows will enormously reduce the number of overlapping chains within a time win-
30 dow, and we will be in a regime where time windows encompass neutrons from a single
31 fission chain.

32 In contrast to bare ^3He tubes, which can detect only thermal neutrons, liquid scin-
33 tillators can detect only neutrons above a threshold of about 1 MeV. The threshold is
34 because below an MeV the recoil protons do not produce a sufficiently unique scin-
35 tillation light pulse to confidently distinguish them from the light pulse produced by
36 gamma-ray Compton interactions. The total light collected by the fast proton recoil in
37 the scintillator is statistically proportional to the incident neutron energy. This enables
38 a statistical energy spectrum to be determined. This capability allows the current new
39 analysis.

Figure 1: Liquid scintillator array used for measuring the double and triple neutron correlations.

40 This report describes our efforts to use the time correlations of fast neutrons to-
41 gether with their energy spectra as measured directly in a liquid scintillator array to
42 distinguish Pu metal from Pu oxide and also determine the α -ratio of the Pu oxide¹.
43 Some earlier work [1, 2] had conceptually shown by way of Monte Carlo simulation
44 that this method could work. In this paper, we demonstrate that the method actually
45 works experimentally. The liquid scintillator array used to obtain the results that we
46 will discuss below is illustrated in Fig. 1. If the object at the center of the array con-
47 tains a multiplying material, each spontaneous fission will typically generate a chain of
48 detected neutrons in the liquid scintillator array. An important difference between Pu
49 metal and Pu oxide is that the α particles produced by the α -decay chain of Pu carry
50 enough energy to cause ^{18}O to emit a neutron via an (α, n) reaction. Although the av-

¹the α -ratio is the ratio of the (α, n) neutron source strength to the spontaneous fission source strength in units of neutrons/second.

51 erage energy of this (α ,n) reaction is 1.9 MeV, close to the average for fast fission, the
52 energy distributions are different. We will show in the following that these differences
53 can be exploited to determine the α -ratio. These neutrons are emitted randomly, and
54 in the case of Pu oxide the emission rate of these neutrons is comparable to the rate
55 of neutron emission due to spontaneous fission. It is important to note that while the
56 proposed method has been tested successfully for unmoderated systems, its possible
57 application to moderated systems remains yet to be determined.

58 The key idea is that the count rate, for fast neutron counting, can be partitioned
59 into a contribution with (α ,n) spectrum and a contribution with fission spectrum. The
60 separate contributions can be determined from liquid scintillator data by a principal
61 component analysis. The ratio of these contributions gives a relation between α -ratio
62 and multiplication. This enables the Cifarelli-Hage [3, 4] correlated moment analysis
63 for determining multiplication given α -ratio to be solved for both quantities.

64 2. Description of Pu objects measured for the reference neutron spectra

65 The algorithms developed in this paper rely upon the knowledge of two impor-
66 tant neutron energy spectra: (a) the energy spectrum of fission neutrons, and (b) the
67 energy spectrum of O(α ,n) neutrons. If these two reference neutron energy spectra
68 are sufficiently distinct, an arbitrary measured neutron energy spectrum should be re-
69 constructable by combining the two reference spectra weighed appropriately.

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

72 2.1. Metallic plutonium

73 For objects containing only gram quantities of plutonium, the system does not mul-
74 tiply, and the neutron emission will be dominated by the spontaneous fission of ^{240}Pu .
75 For kilogram quantities of metallic plutonium, the multiplication can be significant, in
76 which case the fission neutron rate is dominated by induced fissions in ^{239}Pu . In gen-
77 eral, the fission neutron energy spectrum will be a mix of ^{240}Pu spontaneous fissions
78 and ^{239}Pu induced fissions. Fortunately, the fission neutron energy spectra for both
79 ^{240}Pu and ^{239}Pu are very similar, as shown in Fig. 2. Therefore, the fission spectrum of
80 objects containing plutonium will be insensitive to multiplication.

81 In Fig. 3(b), we show the spectrum of energy deposited in our liquid scintillator by
82 the fission neutrons for a bare 2.35 kg plutonium ball of density 15.92 g/cm³ consisting
83 of 93.88% ^{239}Pu , 5.96% ^{240}Pu , 0.13% ^{241}Pu , 0.37% ^{241}Am plus trace amounts of other
84 isotopes.

85 2.2. Plutonium dioxide

86 For the oxide form of plutonium, namely PuO_2 , the fast neutron spectrum will also
87 contain a contribution from (α ,n) neutrons. We have found from computer simulations
88 that the ratio of the (α ,n) neutron rate to the spontaneous fission neutron rate, also
89 referred to as the α -ratio is of the order of 0.8. This α -ratio is a consequence of the fact
90 that ^{240}Pu is a very strong spontaneous fission neutron source, and therefore in general
91 the neutrons coming out of PuO_2 will contain comparable numbers of fission and α ,n

Figure 2: Energy distributions of neutrons from (a) ^{240}Pu spontaneous fission (red), (b) ^{239}Pu 2 MeV induced fission (green), O(α ,n) reactions in (c) PuO_2 (blue) and (d) UO_2 (magenta). Fission spectra [5], (α ,n) spectra computed using SOURCE-4C [6].

neutrons. If we were able to turn off fission reactions for both ^{239}Pu and ^{240}Pu , we could measure the O(α ,n) neutron energy spectrum produced by the plutonium decay α -particles. While this would be possible in simulation, fission reactions cannot be turned off in nature. Of course, one might try using a theoretically derived (α ,n) neutron spectrum to separate an observed fast neutron spectrum into a fission piece and an (α ,n) piece. Unfortunately, converting a theoretical neutron spectrum into a liquid scintillator spectrum would require knowing how to convert an intrinsic neutron energy spectrum into a spectrum of liquid scintillator pulses with the pulse shape that one somewhat arbitrarily identifies as a neutron. This is very difficult to do in practice.

While we have a α -ratio of the order of 0.8 for PuO_2 , the α -ratio is 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 O(α ,n) neutrons. Measuring the spectrum of neutrons coming off of UO_2 would be roughly equivalent to measuring the O(α ,n) neutrons directly, without much pollution from fission neutrons. If it happened that the energy spectra of the O(α ,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 . Fig. 2 shows the energy distribution of O(α ,n) neutrons (mainly from ^{18}O) for both PuO_2 (blue) and UO_2 (magenta). The (α ,n) neutron spectra from PuO_2 and UO_2 are very close; therefore when analyzing the fast neutron spectrum from objects containing PuO_2 one can use the UO_2 (α ,n) spectrum as a surrogate for the PuO_2 (α ,n) spectrum. The objects used for measurement of the O(α ,n) spectrum were 3 UO_2 objects of weights 1485.9, 1463.5, and 1516.7 g. The uranium in these objects consisted of 93.4% ^{235}U , 5.7% ^{238}U , 0.86% ^{234}U . The oxygen consisted in 99.8% ^{16}O , 0.04% ^{17}O and 0.2% ^{18}O . The total ^{18}O mass was 1 g, which is the source of the (α ,n)

neutrons.

The 3 uranium oxide objects were located in the middle of our array of liquid scintillators depicted in Fig. 1. The spectrum of energy deposited in the liquid scintillator by the UO_2 neutrons is shown in blue in Fig. 3(a). Because the UO_2 objects are weak neutron sources, we need to subtract the background that was present during the measurement to get to the UO_2 neutron spectrum. A 15-hour background spectrum was taken within 2 days of the experiment. The neutron energy spectrum from that background measurement is shown in green in Fig. 3(a). The rate of that background is 1.6 n/s and is likely due to cosmic-rays. Since the count rate for the measured objects is 13.52 n/s, the detection system system is measuring background neutrons 12% of the time. Thus, the blue curve in Fig. 3(a) contains both the UO_2 neutrons and 12% of background neutrons. Subtracting the background spectrum in green from the UO_2

Figure 3: Fast neutron energy spectra. The UO_2 curve represents 1800 s of data, the background 53,442 s, and the Pu metal ball 596 s. The measured count rates for the UO_2 objects, background and Pu metal were 13.52 n/s, 1.6 n/s, and 9123 n/s, respectively.

spectrum in blue, we obtain the background-suppressed UO_2 neutron spectrum shown in blue in Fig. 3(b).

Regarding the fission neutrons emitted by both the spontaneous fissions of ^{238}U and the induced fissions in ^{235}U , since only 2 to 3% percents of the neutrons are due to fission in UO_2 , the red curve in Fig. 3(b) would have to be shifted down by almost 2 orders of magnitude before being subtracted from the blue curve to produce a pure (α, n) neutron spectrum from uranium decay α -particles on oxygen. We can thus see that the normalized U fission curve would very small compared to the observed UO_2 curve, and therefore the effect of subtracting the U fission spectrum would be negligible. Given the resemblance between the (α, n) neutron energy spectra from UO_2 and PuO_2 (see Fig. 2), we will from this point on use the (α, n) neutron energy spectrum from UO_2 as a substitute for the (α, n) neutron energy spectrum for PuO_2 .

3. Moment equations with different nuclear data and efficiencies for (α, n) neutrons and fission neutrons

The times of arrival of the neutrons in each of the liquid scintillator cells were recorded. Randomly splitting the sequence of time tags into N segments of length T — where T is of the order of nanoseconds to hundreds of microseconds — one can count how many neutrons arrive in the first segment, how many in the second segment, in the third one, etc. and build distributions $b_n(T)$ of the number n of neutrons arriving in the segments of length T . For the sake of illustration, one such count distribution is shown in Fig. 5. By repeating this procedure for segments of different lengths T , multiple count distributions $b_n(T)$ can be obtained.

These count distributions $b_n(T)$ can be used to determine the strength F_s of the spontaneous fission sources in the object, the efficiency ε of the liquid scintillator array, and multiplication M of fissile material [7]. We will show these count distributions can also be used to determine the rate of neutrons from the (α, n) reactions. This will be shown by way of the first three moment equations for the count distributions.

The count rate can be partitioned [9] into a contribution with (α, n) spectrum and a contribution with fission spectrum,

$$R_1 = S_\alpha q_\alpha \varepsilon_\alpha + F_s \bar{v}_s \varepsilon q M + S_\alpha p_\alpha \bar{v}_\alpha \varepsilon q M. \quad (1)$$

The three contributions to Eq. 1 are illustrated in Figs. 4. The first term is due to (α, n) neutrons that escape without inducing fission, Fig. 4(c). Because of the spectrum difference of efficiency for the liquid scintillator detectors, ε_α can be different from ε . The second term is the usual count rate from spontaneous fission induced chains, Fig. 4(a). The last term is due to (α, n) neutron induced fission. As noted by Böhnel [8] and Hage-Cifarelli [4], the probability for an (α, n) neutron to induce fission is in general different from a fission spectrum neutron. (This is most obvious for ${}^9\text{Be}(\alpha, n){}^{12}\text{C}$, where the high energy neutron induces fission with a higher probability p_α than fission spectrum, and with a higher $\bar{v}_\alpha$.) Also $q_\alpha = 1 - p_\alpha$, the probability for neutron escape can be different for (α, n) spectrum neutrons than fission spectrum. It is further assumed that the induced fission by the (α, n) neutron emits neutrons with a fission spectrum. The (α, n) neutron induced fission acts like an effective spontaneous fission for the rest of the chain, Fig. 4(b).

The ratio of neutron source rates is the α -ratio, here denoted A ,

$$A = \frac{S_\alpha}{\bar{v}_s F_s}.$$

For time-gated fast neutron counting, the time dependent second and third moments of the random time gate distribution $b_n(T)$ are a generalization of the formulas given by Prasad-Snyderman [7]. Normalizing by the count rate,

$$Y_{2F}(T) = \frac{(\varepsilon q M)^2}{(\varepsilon q M)(1 + p_\alpha \bar{v}_\alpha A) + \varepsilon_\alpha q_\alpha A} [D_{2s} + \bar{v}_\alpha p_\alpha A D_{2\alpha} + D_2(M-1)(1 + \bar{v}_\alpha p_\alpha A)] \left(1 - \frac{1 - e^{-\alpha T}}{\alpha T}\right) \quad (2)$$

$$(3)$$

(a) Fission chains generated by spontaneous fission neutrons.

(b) Fission chains generated by \$(\alpha,n)\$ neutrons.

(c) Direct \$(\alpha,n)\$ neutron detection.

Figure 4: Three classes of neutrons contributing to the detector.

$$Y_{3F}(T) = \frac{(\epsilon q M)^3}{(\epsilon q M)(1 + p_\alpha \bar{v}_\alpha A) + \epsilon_\alpha q_\alpha A} [D_{3s} + p_\alpha \bar{v}_\alpha A D_{3\alpha} + (M-1)(1 + p_\alpha \bar{v}_\alpha A) D_3] \left(1 - \frac{3 - 4e^{-\alpha T} + e^{-2\alpha T}}{2\alpha T} \right) + \frac{(\epsilon q M)^3}{(\epsilon q M)(1 + p_\alpha \bar{v}_\alpha A) + \epsilon_\alpha q_\alpha A} [2D_2(M-1)(D_{2s} + p_\alpha \bar{v}_\alpha A D_{2\alpha}) + 2(M-1)^2 D_2^2(1 + p_\alpha \bar{v}_\alpha A)] \left(1 - \frac{2 - (2 + \alpha T)e^{-\alpha T}}{\alpha T} \right) \quad (4)$$

For the correlated triples, the two terms with different time dependences correspond to the following topological classes : in the first term, all counted neutrons have a single common ancestor, while for the second time dependence term two of the counted neutrons have a common ancestor fission, and the third counted neutron has an ancestor with the neutron that induced that fission.

In contract with ^3He counting, the ratio of the two terms in Eq. 1 becomes measur-

able for fast counting,

$$\begin{aligned}
 \rho &= \frac{S_\alpha q_\alpha \varepsilon_\alpha}{(\varepsilon q M) [F_s \bar{v}_s + S_\alpha p_\alpha \bar{v}_\alpha]} \\
 &= \frac{A q_\alpha \varepsilon_\alpha}{(\varepsilon q M) [1 + A p_\alpha \bar{v}_\alpha]} \\
 &\simeq \frac{A r_\varepsilon}{1 + (M-1)(1+A)}. \tag{5}
 \end{aligned}$$

173 The approximations for PuO₂ are that (a) $p_\alpha \simeq p$, so $q_\alpha \simeq q$ and (b) $\bar{v}_\alpha \simeq \bar{v}$. The
 174 ratio $r_\varepsilon = \frac{\varepsilon_\alpha}{\varepsilon}$ is an intrinsic property of the liquid scintillator spectral response, and can
 175 be determined. For our liquid scintillators we find $r_\varepsilon \simeq 1.6$. For PuO₂, the additional
 176 measured quantity defined in Eq. 5 will turn out to give an additional relation between
 177 M and A, and enable one to determine both A and M by the Cifarelli-Hage moment
 178 method.

With the relation provided in Eq. 5, the Cifarelli-Hage equation becomes

$$\frac{R_{2F}^2}{R_{3F}} = \frac{[D_{2s} + (1+A)(M-1)D_2]^2}{D_{3s}(1+A) + 2(1+A)(M-1)D_{2s}D_2 + 2[(M-1)(1+A)]^2 D_2^2 + (1+A)(M-1)D_3(1+A)} \tag{6}$$

(7)

For A=0, this formula is the familiar Hage-Cifarelli equation for M. For A≠0, all the M dependence is in the combination $(1+A)(M-1)$, and this can be expressed in terms of the measured ρ , and the unknown A using Eq. 5,

$$(1+A)(M-1) = \frac{r_\varepsilon A}{\rho} - 1. \tag{8}$$

Substituting Eq. 8 into Eq. 7, we find that A satisfies the following quadratic equation:

$$2D_2 \left(\frac{r_\varepsilon}{\rho} A - 1 \right) \left[D_{2s} + D_2 \left(\frac{r_\varepsilon}{\rho} A - 1 \right) \right] + (A+1) \left(D_{3s} + \left(\frac{r_\varepsilon}{\rho} A - 1 \right) D_3 \right) = \frac{\frac{\rho}{r_\varepsilon} + 1}{\rho + 1} \left[D_{2s} + \left(\frac{r_\varepsilon}{\rho} A - 1 \right) D_2 \right]^2 \frac{R_{3F}}{R_{2F}^2} \tag{9}$$

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180 After solving Eq. 9 for A, Eq. 8 then determines M. In the next sections, we show
 181 that this equation, together with the neutron spectral information provided by the liquid
 182 scintillators, can be used to measure both the α -ratio and the multiplication of pluto-
 183 nium oxide samples of interest.

184 4. Liquid scintillator time correlation results

185 For the 2.35 kg Pu ball previously described, the count distribution for T=1 μ s, and
 186 the moments $Y_{2F}(T)$ and $Y_{3F}(T)$ are shown in Fig. 5. The total time is shared among
 187 the 6 time gates in such a way as to keep the errors on the $Y_{2F}(T)$ and $Y_{3F}(T)$ values
 188 for the different time gates approximately equal. Fast neutron counts are not re-used

189 among different time gates. These data correspond to a measurement of 113 seconds,
 190 21.2 seconds of which contributed to the $1 \mu\text{s}$ time gate, and the remaining time was
 191 shared among the other time gates. The values of R_1 , R_{2F} and R_{3F} can be extracted
 192 from the graphs by fitting the data to the time dependence, $\bar{C} = R_1 T$, and for $Y_{2F}(T)$,
 Eq. 3, and for $Y_{3F}(T)$, Eq. 4.

Figure 5: Count distribution and moments for Pu metal ball. T between 5 ns and $1 \mu\text{s}$. The set of parameters used for the moment reconstruction $(M, \varepsilon, \alpha) = (2.1, 4.6\%, 0.006)$ was determined using the measured moments and spectral information.

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Fig. 6 shows the results for a PuO_2 sample containing 8.72 g of plutonium. The measurement thereof was much longer as the weight of PuO_2 was just a few grams.

Figure 6: Count distribution and moments for PuO_2 sample. T between 5 ns and $1 \mu s$. The set of parameters used for the moment reconstruction $(M, \varepsilon, \alpha) = (1.06, 5.1\%, 0.86)$ was determined using the measured moments and spectral information.

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5. Liquid scintillators spectral information

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In this section we describe experimental results which illustrate how the information contained in the spectrum of energies deposited by the fast neutrons in the liquid scintillator cells can be used to differentiate metallic plutonium from plutonium dioxide. For the measured PuO_2 sample, the spectrum of energies deposited by the fast neutrons is shown in green in Fig 7, along with the Pu metal neutron spectrum in red and the pure (α, n) neutron spectrum from α -particles on oxygen in blue (the latter two directly copied from Fig. 3(b)). In Fig. 8 we show that the red and blue spectra can be added with suitable weights to reconstruct the green curve. In particular, by adding the ^{240}Pu spectrum pre-multiplied by 0.45 to the (α, n) spectrum pre-multiplied by 0.55, one obtains the reconstruction spectrum of energies deposited shown in red in

Figure 7: Fast neutron energy spectra for PuO₂ sample (green), along with Pu metal ball (red) and modified UO₂ (blue) spectra. The experimental curve for PuO₂ represents 7200 s of data.

207 Fig. 8(b). On the other hand, when one measures the Pu metal ball, the weights that
 208 are optimal for the reconstruction of the spectrum of deposited energies are 0.995 of
 209 the ²⁴⁰Pu spectrum and 0.005 of the (α ,n) spectrum. So we find that just measuring the
 210 spectrum of energies deposited in a liquid scintillator by fast neutrons is sufficient to
 211 distinguish Pu metal and Pu oxide.

(a) Factors by which the blue and red spectra shown in Fig. 7 must be multiplied to reconstruct the spectrum of energies deposited by the fast neutrons emitted by the PuO₂ sample: 0.45 for the Pu metal spectrum and 0.55 for the UO₂ sample spectrum.

(b) Spectrum of energies deposited by fast neutrons in liquid scintillator cells for the PuO₂ sample (blue), along with its reconstruction (red) from the blue and red spectra shown in Fig. 7 and the optimal weights shown on the left.

Figure 8: Reconstruction of the PuO₂ objects using the sum of two weighed energy spectra.

212 Setting $\rho=1.24$, the solution to Eqs. 8-9 with $M \geq 1$ is $A = 0.86 \pm 0.08$, $M = 1.06 \pm$
 213 0.09 . The exact value of 0.8 for A is within 1 standard deviation of our solution. Using
 214 $R_{2F} = R_2/R_1$ calculated from Eqs. 1 and 3 where $R_2 = \lim_{\alpha T \rightarrow \infty} Y_2(T)$, we determined
 215 the value of ϵ to be 5.1%, while Eq. 1 implies a spontaneous fission source rate of $662 \pm$

216 57 neutrons/sec. Although not exactly the same strength as the true value of 519 n/s,
 217 the implied spontaneous fission rate is within 20% of the correct answer. If we measure
 218 10 times longer, the solution with $M \geq 1$ becomes $A = 0.84 \pm 0.02$, $M = 1.05 \pm 0.02$,
 219 $\varepsilon = 5.3\%$, and the spontaneous fission source rate becomes 638 ± 11 neutrons/sec. For
 220 the metallic plutonium ball, our algorithm gives $\rho = 0.005$. The solution to Eqs. 8-9 with
 221 $M \geq 1$ is $A = 0.006 \pm 0.0001$, $M = 2.1 \pm 0.04$. The value of ε is 4.6% and the source
 222 strength is $149,015 \pm 2,700$ neutrons/sec, which is off the true value by less than 1.5%,
 223 or less than 1 standard deviation.

224 6. Conclusion

225 In this paper, we have shown first of all that measuring the energy spectrum of
 226 the fast neutrons using a liquid scintillator allows one to immediately distinguish the
 227 metallic and oxide forms of plutonium. In addition, combining this spectral informa-
 228 tion with the Feynman 2-neutron and 3-neutron correlations allows one to extract the
 229 α -ratio without explicitly knowing the multiplication. Given the α -ratio one can then
 230 extract the multiplication as well as the ^{239}Pu and ^{240}Pu masses directly from the mo-
 231 ment equations. In principle the same techniques could be used to distinguish metallic
 232 Pu from other compounds of Pu, such as PuF_2 , where (α, n) neutron emission is also
 233 significant.

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