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Pulse-height distributions of neutron and gamma rays from plutonium-oxide samples

S.A. Pozzi [a,*], S.D. Clarke [a], M. Flaska [a], P. Peerani [b]

[a] Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor, MI 48109, USA
[b] European Commission EC–JRC–IPSC, Ispra, Italy

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ABSTRACT

We present new results on neutron and gamma-ray pulse-height distributions (PHDs) measured with liquid scintillators from five plutonium-oxide samples of varying mass and burnup and a ^{252}Cf isotopic source. We show that the analysis of the pulse-height distributions can be used to easily distinguish the fissile material (plutonium oxide) from the ^{252}Cf source. Moreover, the slope of the measured pulse-height distributions can be analyzed to distinguish the burnup of the samples, independent of their masses. Finally, the measurement scenarios are modeled using the MCNP-PoliMi code and good agreement is observed between the measured and the simulated pulse-height distributions.

1. Introduction

Recently, considerable effort has been placed in the development of tools and techniques for the measurement of neutrons and gamma rays emitted by isotopic neutron sources or bulk fissile material such as plutonium and uranium. Within these studies, several researchers have focused on the use of organic scintillators of various types for the measurement and characterization of neutron and gamma-ray sources within the areas of homeland security [1,2] or basic nuclear physics [3]. In some cases, the neutron energy spectra were analyzed to obtain information on the radioactive sources or nuclear material composition [4,5]. Finally, some studies have addressed the problem of neutron energy spectrum unfolding on the basis of pulse-height distributions (PHDs) measured with organic scintillators [6,7].

In a past study, we showed that neutron PHDs can be used for neutron source identification by comparing measured neutron PHDs from isotopic neutron sources having significantly different initial neutron energy spectra, i.e., ^{252}Cf, Am–Be, and Am–Li sources [4]. In the current study, we show that source identification is possible also when comparing bulk plutonium-oxide samples and a ^{252}Cf source. Specifically, we present new measured PHDs acquired from ^{252}Cf and plutonium-oxide samples of mass varying from 50 to 500 g. The analysis of gamma-ray PHDs is also included in the present study. The results show that direct observation of the neutron or gamma-ray PHDs allows one to easily distinguish ^{252}Cf from plutonium oxide of any mass in the range considered.

2. Description of measurements

The measurements were performed in August 2008 at the Performance Laboratory of the Joint Research Center (JRC) in Ispra, Italy, jointly by the University of Michigan and JRC staff. In these measurements, pulse heights, multiplicities, and cross-correlation functions were acquired for ^{252}Cf and plutonium-oxide (PuO_2) samples using a multi-detector system developed at the University of Michigan that uses digital processing techniques and post-processing algorithms. The results from the cross-correlation measurements are described in a previous publication [5], whereas the analysis of neutron and gamma-ray PHDs is the subject of this paper.

2.1. Description of measurement setup and fissile material

The measurement system was developed at the University of Michigan and consists of six cylindrical, EJ-309 liquid scintillation detectors (13.3 cm high and 13 cm in diameter), a digital data acquisition system, and algorithms for pulse shape discrimination

* Corresponding author.
E-mail address: pozzisa@umich.edu (S.A. Pozzi).

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and time correlation. In these measurements, the detectors were placed symmetrically around the sample approximately 30 cm from the center of the sample. Lead shielding sleeves with a total thickness of approximately 2.6 cm were placed around each sample to reduce the gamma-ray background. Fig. 1 shows two photographs of the measurement setup: the EJ-309 scintillators and the centrally located sample holder.

Data acquisition was performed with a CAEN V1720, 12-bit, 250 MHz digitizer: pulses were sampled and stored directly from the detector anodes. Each detector was connected to a separate digitizer channel and each channel was triggered independently. A 100 keVee detection threshold (corresponding to a deposited neutron energy of approximately 600 keV) was applied to each channel. This threshold was chosen to minimize the gamma-ray background from the gamma-ray decay of ^{241}Am and other isotopes in the plutonium-oxide samples.

The voltage at the detectors was set using a ^{137}Cs source: the Compton edge was aligned to a value of approximately 0.55 V for all six detectors. This procedure ensured that the detectors were operating at similar effective gains. The dynamic range of the digitizer was 2 V.

The fissile material available at the JRC Ispra consists of PuO_2 powder of varying burnups, stored in cylindrical steel containers with various heights and diameters. The measurements were performed on five samples of varying masses and burnups: 100, 300, and 500 g low-burnup samples and 50 and 100 g high-burnup samples, and on one ^{252}Cf isotopic neutron source. Table 1 lists the isotopic composition of plutonium in the low- and high-burnup samples [8], obtained from the reprocessing of fuel from a Magnox and a pressurized water reactor, respectively. The ^{252}Cf source had a certified intensity of 2500 n/s.

2.2. Digital pulse shape discrimination

An offline digital pulse shape discrimination (PSD) method was used to distinguish neutrons from gamma rays [4]. This method is based on a standard charge integration technique, which calculates the ratio of the integrals of two different pulse intervals. The first interval covers the tail of a pulse, while the second interval covers the total pulse. The integration parameters for the pulse tail are optimized for the specific detector system. In these measurements, the tail start integration time was set to 20 ns past the pulse peak, and the stop integration time (for both integrals) was set to 272 ns past the peak. These values were obtained from an optimization procedure that examined the effect of varying the start and stop integration times on the particle classification accuracy.

Fig. 2 shows a plot of tail and total integrals for pulses collected from a single EJ-309 scintillator. The digital PSD methodology used in this study is based on a discrimination curve that separates the neutron and the gamma-ray region. The chosen discrimination curve for the two detectors is shown in Fig. 2: pulses above this line are classified as neutrons and below this line as gamma rays. The parameters for this curve were determined by fitting several arbitrarily chosen points in the region between the two distributions with a discrimination curve. This procedure was repeated for each detector to account for slight differences in the light collection efficiency and response of the individual detectors. Note that standard figures of merit parameters cannot be used to quantify the accuracy of the PSD methodology in this case; this is due to the non-linearity of the discrimination curve being used. Instead, visual inspection shows that the PSD achieved with the proposed methodology is very good, even for the small pulses close to the measurement threshold of 100 keVee. For all plutonium-oxide samples, approximately 6.8% of the measured pulses were due to neutrons, whereas for the ^{252}Cf source this value was approximately 13%.

Table 1
Isotopic composition (mass percent) of low- and high-burnup PuO_2 samples.

Isotope	Low burnup (%)	High burnup (%)
^{238}Pu	0.20	1.72
^{239}Pu	70.96	58.10
^{240}Pu	24.58	24.77
^{241}Pu	3.29	9.77
^{242}Pu	0.98	5.65

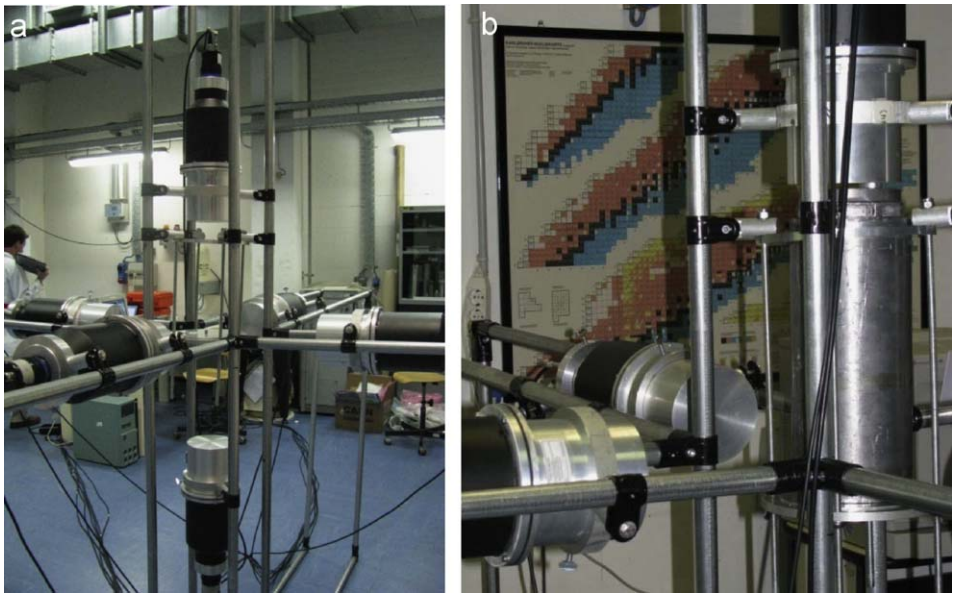

Fig. 1. Photographs of the measurement setup showing: (a) six EJ-309 liquid scintillators and centrally located ^{252}Cf source and (b) plutonium-oxide sample shielded by lead sleeves of thickness approximately 2.6 cm.

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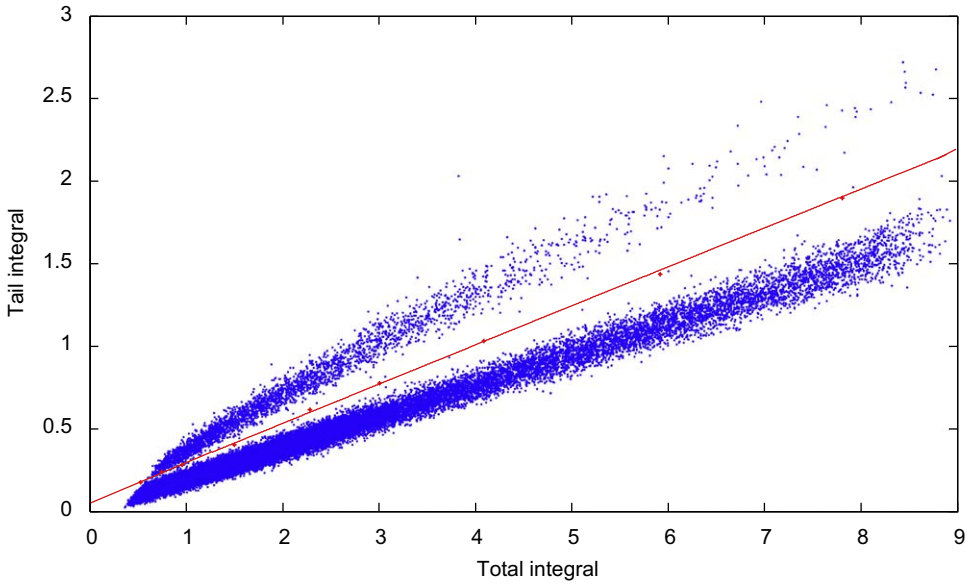

Fig. 2. Tail and total pulse integrals for measured pulses from a 50 g, high-burnup PuO_2 sample. The discrimination curve is shown: pulses above the curve are classified as neutrons and below as gamma rays.

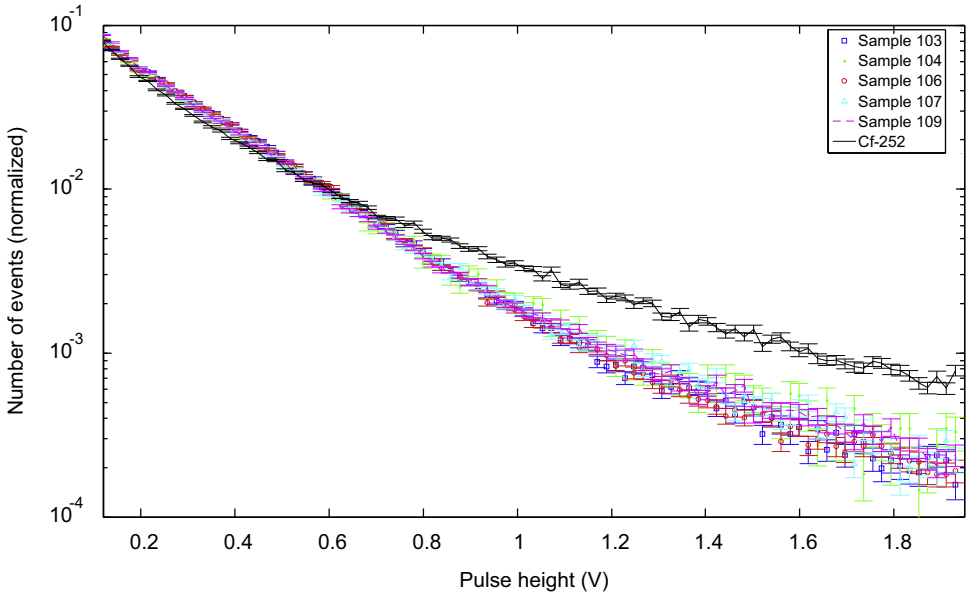

Fig. 3. Measured neutron pulse-height distributions for PuO_2 powder of varying mass and burnup and ^{252}Cf isotopic source.

3. Measurement results

PHDs were obtained by binning the pulse heights of pulses from neutrons and gamma rays as determined by the PSD method described above. One hundred pulse-height bins with a width of 0.02 V were used. The results were normalized to allow for direct comparison of the various samples.

Fig. 3 shows the measured neutron PHDs with their statistical error for the five PuO_2 samples and the ^{252}Cf source. The results show that the ^{252}Cf PHD has a more pronounced tail that clearly separates it from the PHDs of all the plutonium-oxide samples. This effect is clearly due to the harder neutron spectrum of ^{252}Cf with respect to plutonium. The PHDs from the five PuO_2 samples are visually similar in shape.

Fig. 4 shows the measured gamma-ray pulse-height distributions for the five PuO_2 samples, the ^{252}Cf source, and a ^{137}Cs source. The results show that the ^{252}Cf PHD has a distinctly different shape, which clearly separates it from the PHDs of all the plutonium-oxide samples. It should be noted that the PHDs from the plutonium-oxide samples resemble that of the ^{137}Cs source, which consists of the Compton continuum resulting from the 662 keV gamma rays. This effect is probably due to the 662.40 keV ^{241}Am gamma-ray line, which is prominent in the PuO_2 samples.

4. Data analysis

The measured neutron PHDs described in Section 3 were analyzed by linear fit of the logarithm of the normalized distributions; these fits are shown in Fig. 5. Clearly, better fits can be obtained using a higher order polynomial or other curves to fit the data. However, the simple linear analysis gives the possibility of using a single value (the slope of the line) for the

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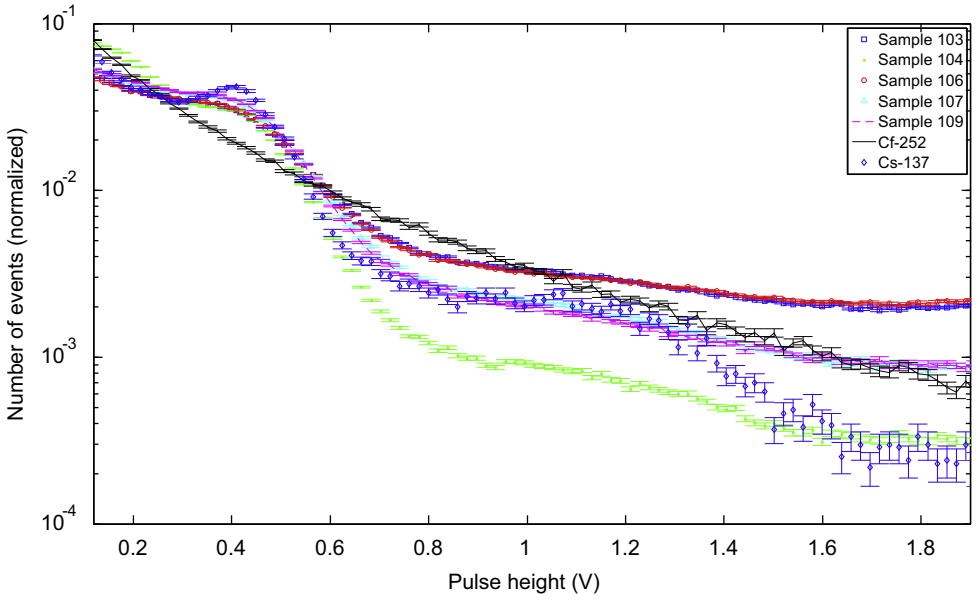

Fig. 4. Measured gamma-ray pulse-height distributions for PuO_2 powder of varying mass and burnup, ^{252}Cf, and ^{137}Cs isotopic sources.

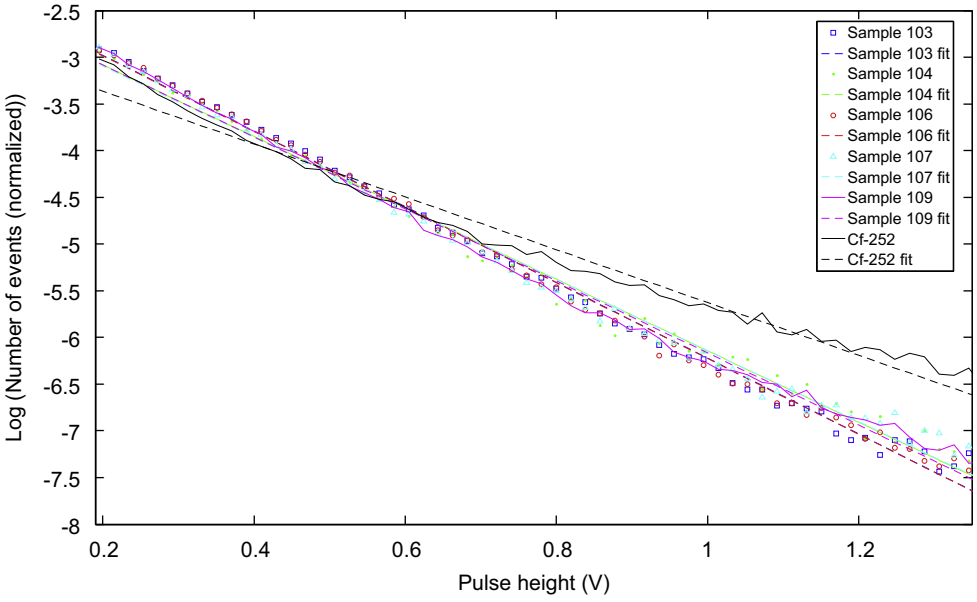

Fig. 5. Measured neutron pulse-height distributions for PuO_2 powder of varying mass and burnup and ^{252}Cf isotopic source: linear fits of the logarithm of the normalized number of events are shown with the dotted lines.

Table 2
Slope of fitted line of measured neutron PHDs.

Burnup	Sample mass (g)	Sample number	Slope of fitted line
Low	100	104	−3.82
	300	107	−3.80
	500	109	−3.85
High	50	103	−4.05
	100	106	−4.05
^{252}Cf	tbd	N.A.	−2.83

determination of the source type and burnup type (high or low burnup). In addition, this slope has the physical significance of being the rate of decay of the exponential distribution of the pulse heights, as shown in Fig. 3. The slopes of the fitted lines are given in Table 2. The slopes are indicative of the PuO_2 composition: the low-burnup samples have slope values between −3.80 and −3.85, the high-burnup samples approximately −4.05, and the ^{252}Cf source approximately −2.83.

5. Monte Carlo analysis

5.1. Description of MCNP-PoliMi

The simulation of neutron PHDs in an organic scintillator must take into account multiple scatterings and detector non-linearities. Therefore, the simulation tools must describe the neutron interactions within the scintillator on an event-by-event basis. A modified version of MCNP4C called MCNP-PoliMi, and its associated post-processor, has been developed to simulate these

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quantities [9]. MCNP-PoliMi version 1.2.5 is capable of running with all standard MCNP source types and includes several specific spontaneous-fission-source distributions (i.e., ^{252}Cf, ^{240}Pu, ^{242}Pu, etc.). In addition, MCNP-PoliMi also contains distributions for (α, n) reactions for plutonium isotopes in oxides. These special sources were used in the simulations described here.

5.2. Description of Monte Carlo models

The MCNP-PoliMi model of the measurement setup included a detailed model of the container with the PuO_2 and surrounding lead sleeves as well as all six EJ-309 scintillators. For simplicity, only the active volume of each scintillator was modeled.

Each measurement was simulated with the MCNP-PoliMi code. The source for each simulation was modeled using the MCNP-PoliMi internal definitions for plutonium spontaneous fission and neutron production from (α, n) reactions. The relative contributions for each neutron source were computed from the isotopic compositions given in Table 1 and the specific activities of each isotope for spontaneous fission and (α, n) in oxides. Table 3 summarizes the fraction of total source neutrons emitted from each reaction for the low- and high-burnup PuO_2 samples. Some contributions were neglected due to their low relative intensity (see Table 3). For simplicity, the few ^{238}Pu spontaneous fissions were normalized into the ^{240}Pu and ^{242}Pu energy distributions.

5.3. Simulation results and comparison

Fig. 6 shows the simulated neutron energy spectra incident on the detector face from the five PuO_2 samples and the ^{252}Cf source. Note that these distributions include self-shielding effects in the PuO_2 powder and the shielding effect of the lead sleeve placed around the samples. As expected from the data reported in Table 3, in the case of the high-burnup samples (samples 103 and 106), the (α, n) contribution is more prevalent than the case of the low-burnup samples (samples 104, 107, and 109). This fact contributes to the peak at neutron energies of approximately 2.3 MeV, which is present in all high-burnup samples. In both cases the (α, n) and spontaneous fission neutrons contribute to the total neutron energy spectrum. While the (α, n) spectrum is harder on average than the spontaneous fission spectrum, the spontaneous fission spectrum is greater at high energies (greater than 4 MeV); this is clearly observable in Fig. 6. Due to the above-mentioned shielding effects, the spectrum of the neutrons incident on the detector is significantly softer than that of the emitted neutrons for the PuO_2 samples. There are also observable gaps in the low-energy (less than 2 MeV) portion of the spectra, which are present only in the PuO_2 samples. This effect is given by neutron elastic scattering interactions in oxygen [10]. Table 4 gives the average neutron energy for each of these cases. The ^{252}Cf source consists of only spontaneous fission neutrons and has no shielding present.

Table 3
Fraction of spontaneous fission (S.F) and Alpha-*N* neutrons emitted from low- and high-burnup PuO_2 samples.

Isotope	Low burnup (%)	High burnup (%)
^{238}Pu(*s.f.*)	1.43	6.53
^{238}Pu(α, n)	7.39	33.80
^{239}Pu(*s.f.*)[a]	<0.01	<0.01
^{239}Pu(α, n)	7.49	3.24
^{240}Pu(*s.f.*)	69.43	37.03
^{240}Pu(α, n)	9.60	5.12
^{241}Pu(*s.f.*)[a]	<0.001	<0.01
^{241}Pu(α, n)[a]	0.01	0.02
^{242}Pu(*s.f.*)	4.64	14.23
^{242}Pu(α, n)[a]	0.01	0.02

[a] These contributions were neglected in the MCNP-PoliMi source specification.

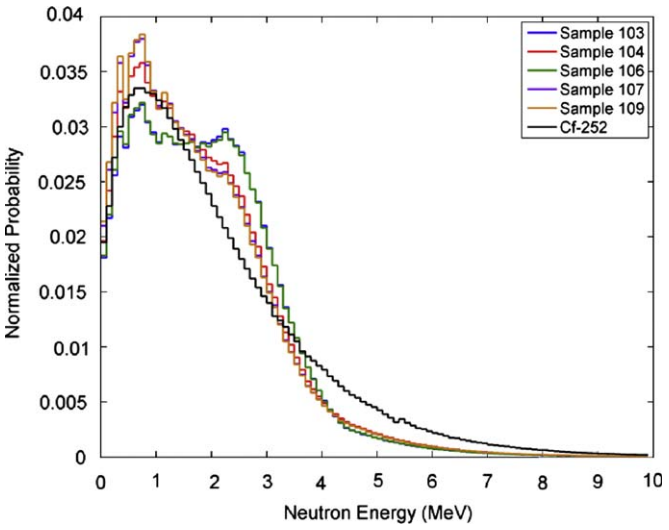

Fig. 6. MCNP-PoliMi simulated neutron energy spectra for PuO_2 powder of varying mass and burnup and ^{252}Cf source.

Table 4
Simulated average neutron energy incident on detector for low- and high-burnup PuO_2 samples and ^{252}Cf source.

Burnup	Sample number	Sample mass (g)	Average neutron energy (MeV)
Low	104	100	1.87
	107	300	1.83
	109	500	1.82
High	103	50	1.95
	106	100	1.95
^{252}Cf	N.A.	tbd	2.13

Fig. 7 shows a comparison between simulated and measured neutron PHDs for three of the plutonium-oxide samples. The comparison was performed by normalizing the neutron PHDs to the total number of detected neutrons in both cases. Good agreement was obtained between the simulated and the measured data.

6. Conclusions

This paper presented new measured and simulated results on neutron and gamma-ray pulse-height distributions measured with liquid scintillators from five plutonium-oxide samples of varying masses and burnups, and ^{252}Cf and ^{137}Cs isotopic sources. These measurements were performed with a fast, multi-detector digitizing system of 250 MHz and 12-bit resolution developed at the University of Michigan. For real-time detection and data collection, the typical measurement time is of the order of a few minutes for PuO_2 samples in the mass range considered here (100–500 g). The digital system requires algorithms for pulse shape discrimination (PSD) and pulse-height analysis. An optimized PSD approach based on charge integration was presented here. One of the advantages of using a digital system is that the user can analyze the data as many times as needed. However, simplified systems could be developed that are even more cost-effective and field-deployable; in fact, this type of measurement could be performed with a single detector, though sacrificing efficiency when compared to the multi-detector measurement system described in this paper.

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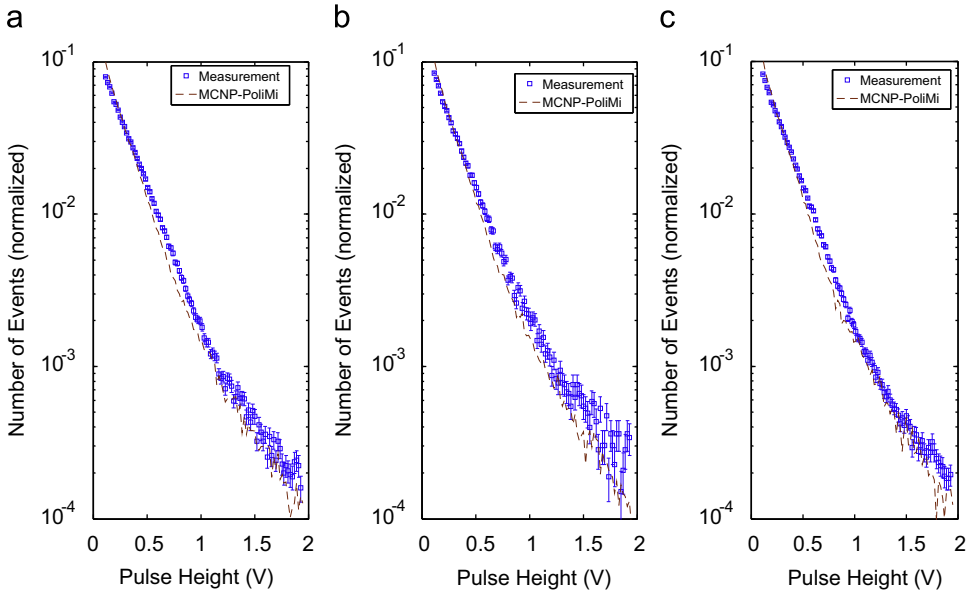

Fig. 7. Comparison between simulated and measured neutron PHDs for three PuO_2 samples: (a) sample 103, (b) sample 104, and (c) sample 106.

In this study, we showed that the analysis of the neutron and gamma-ray pulse-height distributions can be used to easily distinguish the fissile material (PuO_2) from the ^{252}Cf isotopic source. Moreover, the analysis of the slope of the neutron pulse-height distributions can be used to distinguish the burnup (high or low) of the samples, independent of their mass. This capability is of great interest in fields such as nuclear non-proliferation and safeguards.

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