Plutonium Metal vs. Oxide Determination with the Pulse-Shape-Discrimination-Capable Plastic Scintillator EJ-299-33

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Abstract

Neutron measurements can be used to distinguish plutonium in metal or oxide form, a capability that is of great interest in nuclear nonproliferation, treaty verification, and other applications. This paper describes measurements performed on well-characterized samples of plutonium oxide and plutonium metal using the pulse-shape-discrimination-capable plastic scintillator EJ-299-33. Results are compared to those obtained with a same-sized detector cell using the liquid scintillator EJ-309. The same optimized, digital pulse shape discrimination technique is applied to both detectors and the neutron pulse height distributions are compared. Results show that the EJ-299-33 plastics can be successfully used for plutonium measurements, where the gamma ray to neutron detection ratio is much higher than for typical radioactive sources. Results also show that EJ-299-33 detectors can be used to characterize plutonium samples, specifically to discriminate between plutonium metal and oxide.

1. Introduction

The discrimination of plutonium in metal vs. oxide form is of interest in nonproliferation, safeguards, and treaty verification applications. Previous efforts have shown that neutron pulse height distributions measured with liquid organic scintillators can be used to characterize nuclear materials of various forms [1]. However, for some applications the use of liquid scintillators in the field is discouraged, due to concerns about possible leaks. For this reason, plastic scintillators would be preferable to liquid scintillators. Until very recently, plastic scintillators were not able to discriminate between neutrons and gamma rays.

This paper presents new results obtained with EJ-299-33, a new plastic scintillator capable of pulse shape discrimination (PSD) [2-5]. Specifically, we present new results from measurements performed at the Joint Research Centre at Ispra, Italy in May 2013 on plutonium oxide and plutonium metal samples, and a Cf-252 source.

2. Measurement Setup

Four detectors were arranged in a square configuration around each of the samples. The samples were placed inside a lead pig, which had a thickness of 3.3 cm and was used to partially shield the many gamma rays emitted from the plutonium samples. Two 7.6-cm diameter by 7.6-cm thick EJ-299-33 plastic scintillators

were arranged at 10 cm from the center of the sample. Two 7.6-cm diameter and 5.1-cm thick liquid scintillators, one EJ-309 and one EJ-315, were arranged at 15 cm from the center of the sample. The plastic scintillator detector assemblies used ETL 9821B photomultiplier tubes and were provided by Eljen technologies [6]. The EJ-309 and EJ-315 liquid scintillator assemblies used the same photomultiplier tube model as the plastics. Figure 1 shows a photograph of the measurement setup.

Fig. 1. Measurement setup used for this experiment.

2.1 Samples

The first sample consists of a pellet of plutonium oxide of mass 6.62 g, having initial composition (June 1986) Pu-238 1.20 wt%, Pu-239 62.5 wt%, Pu-240 25.4 wt%, Pu-241 5.68 wt%, Pu-242 4.19 wt%, and Am-241 1.44 wt%. The second sample is a plutonium metal alloy (PuNiCu) pellet of mass 20.1 g, with initial composition (January 2001) Pu-238 0.03 wt%, Pu-239 91.3 wt%, Pu 240 8.47 wt%, Pu-241 0.18 wt%, and Am-241 0.83 wt%. Each of the two plutonium samples was measured for one hour. A one-hour measurement with a Cf-252 source was also performed. Table I shows the current sample composition and the neutron emission properties of each sample.

Table I: Composition of each Sample on the Measurement Day, with Neutron Emission Rate.

Sample	Wt % for each Isotope						Neutron Emission Rate
	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	
Plutonium Oxide	0.01%	61.6%	25.0%	1.5%	4.1%	5.5%	4,430 n/s
Plutonium Metal	0.0002%	85.1%	7.9%	0.1%	0.04%	0.9%	1,650 n/s

The plutonium metal sample emits neutrons primarily by spontaneous fission of Pu-240. The plutonium oxide sample emits neutrons primarily by spontaneous fission of Pu-240 and by (alpha, n) reactions resulting from the alpha decay of Pu and Am and subsequent (alpha, n) reaction on oxygen. The Cf-252 source emits neutrons primarily by spontaneous fission, with source strength of approximately 95,900 neutrons per second. The energy spectra of the neutrons emitted by these samples are different. According to the Watt spectrum fits in the ENDF/B-VII data library [7], spontaneous fission emits neutrons of average energy approximately 2.1 MeV for Cf-252 and 1.9 MeV for Pu-240, and the most probable neutron energy is approximately 730 keV for Cf-252 and 800 keV for Pu-240. Neutrons from (alpha, n) have an average energy of approximately 2 MeV and are limited in energy by the energy of the alpha particle at the reaction site and the Q-value of the reaction, which is 590 keV for (alpha, n) reactions on oxygen. Figure 2 shows the neutron energy spectrum simulated for each measured source as emitted from sample. When measuring at a 110-

keVee threshold, 44% of Cf-252 neutrons and 55% of Pu-240 spontaneous fission neutrons were below threshold in the EJ-299-33 plastic.

Fig. 2. MCNPX-PoliMi simulation of emitted neutron energy spectra. Statistical error bars are not shown as they are smaller than the symbols used.

2.2 Data acquisition

A CAEN 4-channel, 250 MHz and 12-bit digitizer was used for data acquisition. The full pulse waveforms were recorded for each of the detectors, with a pulse window of 400 ns. The measurement threshold was set to 115 keVee. Triggering was performed independently on each channel; the four detectors combined for a total count rate of approximately 4300 Hz for Cf-252, 3000 Hz for plutonium oxide, and 1900 Hz for plutonium metal. For this measurement, the systematic dead time never exceeded 0.2%. An optimized, offline, digital pulse-shape discrimination (PSD) algorithm was applied to the measured data [8].

3. Results

Measurement results from the campaign are given in this section, including calibration, pulse shape discrimination, and neutron pulse height distributions.

3.1 Calibration

Figure 3 shows the calibration of the three detectors performed using a Cs-137 source. The EJ-299-33 voltage settings were chosen so that the Compton edge calibration is set to 0.35 V to offset the lower light output of the EJ-299-33 plastic scintillator compared to the EJ-309 liquid scintillator [3, 5]. A difference of approximately 4% is present in the Compton edge calibration of the two EJ-299-33 plastic scintillators.

Fig. 3. Detector calibration performed using a Cs-137 source.

3.2 Pulse Shape Discrimination

Figure 4 shows the PSD results for the Cf-252 source and plutonium metal and oxide samples measured with the two EJ-299-33 and the EJ-309 detector. PSD is performed by integrating two regions of each digitized pulse: the tail and the total. Two regions of pulses can be seen, one corresponding to gamma rays and one corresponding to neutron pulses. For the former, the result of the tail integration is smaller than for the latter, given an equal total integral value.

The pulse shape discrimination line was determined by using a slicing algorithm on the Cf-252 data for each detector such that the discrimination line is dependent on pulse height [9]. A bias parameter is available that reduces the misclassification rate at the cost of neutron efficiency. A single parameter was used to obtain the discrimination line for both EJ-299-33 plastics. Due to slight differences in response, we applied a slightly different discrimination line to each detector. The misclassification rate is greater for pulses with lower total integrals; this effect is observed by overlapping of the neutron and photon regions at low total integral values. The EJ-309 liquid gave clear separation between the two regions, so no bias was applied to the EJ-309 liquid.

Fig. 4. Measured neutron and gamma ray PSD integrals for three samples for EJ-299-33 detectors 1 (a, b, and c) and 2 (d, e, and f), and EJ-309 detector (g, h, i). Each figure shows, in logarithmic scale, a total of 250,000 pulses.

Figure 4g-4i shows PSD results for two plutonium samples and the Cf-252 source measured with the EJ-309 detector. Here, the photon and neutron regions are more clearly separated, ensuring correct classification of a greater number of photon and neutron pulses than in the EJ-299-33 detector.

Table II gives the measured ratio of PSD-attributed photons to neutrons. We do not expect these ratios to be the same for the two detectors because the detection efficiency and the chosen PSD discrimination curves are different. Overall, the EJ-299-33 detector measured more photons and less neutrons emitted by the Cf-252 and plutonium sources than the EJ-309.

Table II: PSD-attributed photon-to-neutron ratio for each detector, for each sample.

Detector	Cf-252	Pu Metal	Pu Oxide
EJ-299-33 det1	2.250(3)	38.1(2)	25.78(6)
EJ-299-33 det2	2.709(3)	56.1(3)	38.4(1)
EJ-309	1.416(2)	35.9(2)	22.23(7)

3.3 Neutron Pulse Height Distributions

Figure 5 shows the measured neutron pulse height distributions (PHDs) for two plutonium samples and the Cf-252 source measured with the EJ-299-33 and EJ-309 detectors. This result is obtained after applying the PSD procedure described in section 2.2. Simulation results obtained with MCNPX-PoliMi are also shown. Figure 5a and 5b show that the two plastic scintillator detectors provided responses that differed significantly, possibly due to differences in the calibration (see Figure 3) or in some other aspect of the experimental setup. Despite these differences, the plutonium oxide neutron PHD is well-separated from the plutonium metal and Cf-252 neutron PHDs. This effect is observed for all the detectors tested.

Fig. 5. Measured neutron pulse height distributions using EJ-299-33 detectors 1 (a) and 2 (b) and EJ-309 detector (c) for the Cf-252 source and plutonium oxide and metal samples. Simulations performed with MCNPX-PoliMi are also shown.

4. Discussion

We present a discussion of the results obtained in this work in terms of the detection of spontaneous fission neutrons in Cf-252 and Pu-240 (relative to measurements of the Cf-252 source and plutonium metal sample, respectively) and of the ability of each of the detectors to discriminate plutonium metal from plutonium oxide.

4.1 Spontaneous fission of Cf-252 and Pu-240

For both EJ-309 and EJ-299-33 detectors used in this measurement, the neutron PHDs obtained from plutonium metal and the Cf-252 source are the same within statistical uncertainty. This result does not agree completely with the simulation, where a slightly different Watt spectrum is used for Cf-252 and Pu-240, in agreement with our knowledge of the Pu-240 neutron spectrum being slightly softer than the Cf-252 neutron spectrum. Our current measurement result agrees with a previous measurement performed with a setup of

liquid organic scintillator cells of larger size [10]. The difference with the simulation result requires further investigation in terms of the accuracy of two parameters used in the simulations: (i) the neutron energy spectrum and (ii) the detector resolution.

4.2 Plutonium metal vs. Oxide

We show that the difference in the emitted neutron spectrum in plutonium metal and oxide (see Figure 2) can be used for sample characterization. Specifically, the neutron spectrum emitted by the plutonium oxide sample has a peak between 2 and 4 MeV, which is not present in the plutonium metal spectrum. This difference results in a measured neutron PHD for the plutonium oxide that has a steeper slope than that of the plutonium metal. We analyzed the PHD from the threshold of 0.115 to 1 MeV, and fit to an exponential:

$$f(L) = C_1 e^{-aL}, 0.115 \leq L \leq 1 \text{ MeV}$$

Table III shows the fitted exponential parameters for each detector and source.

Table III: Fit parameter a for normalized pulse height spectra for each source.

Detector	Cf-252	Pu Metal	Pu Oxide
EJ-309	3.4	3.6	4.2
EJ-299-33 det2	3.2	3.5	4.4
EJ-299-33 det1	3.7	4.4	5.4

The results of the fit show that, for all detectors tested, the value of the a parameter for the plutonium oxide is higher than the a parameter for the plutonium metal. The average difference is 22 percent. We also found that the value of the a parameter cannot be used “across” detectors to make the metal vs. oxide distinction: it cannot be used as an absolute measure. This result indicates that prior detector calibration with known sources is likely needed for the method to be valid.

The results show that the EJ-299-33 scintillator can be used to distinguish a plutonium metal sample from a plutonium oxide sample. In fact, whereas the efficiency of the EJ-299-33 detector is lower than that of the EJ-309 detector, the difference in the a parameter determined by a fit of the measured neutron PHDs is of the same order of magnitude.

5. Conclusions

We presented new results on the measurements of plutonium oxide and metal samples with the EJ-299-33 plastic scintillator. The pulse shape discrimination capabilities and neutron pulse height distributions were compared to the EJ-309 liquid scintillator with the same scintillator dimensions and readout system. It was shown that the EJ-299-33 plastic can be used to perform plutonium sample measurements, where the detected gamma ray to neutron ratios are much higher than those of Cf-252 measurements (over 30 to less than 3, respectively).

Neutron pulse height distributions obtained after pulse shape discrimination were presented for the plutonium samples and for the Cf-252 source. Results showed that, for both detector types, the Cf-252 and the plutonium metal neutron pulse height distribution was the same, within measurement error. The neutron PHDs were used to successfully discriminate the plutonium metal from the plutonium oxide samples: an exponential fit of the neutron PHD from 0.115 to 1 MeV was 22% higher, on average, for the oxide sample than it was for the metal sample.

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