PASSIVE (NMIS) MEASUREMENTS ON PLUTONIUM OXIDE SAMPLES

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ABSTRACT

This paper describes the measurements and Monte Carlo simulations that were performed in March and April, 2005 on plutonium oxide standards stored at the EURATOM Joint Research Center in Ispra, Italy. The simulations were performed with the MCNP-PoliMi code, which correctly simulates the statistics of the neutron and photon field in a multiplying medium. The plutonium samples were oxide powder having mass in the 50 to 2500 g range, with three compositions corresponding to three different burnups. The measurements were performed with 8 large (25 by 25 by 8 cm) liquid scintillators, arranged in several different configurations. The measurements rely on the Pu-240 spontaneous fission, which emits multiple neutrons and gamma rays. These particles are detected with the scintillators and correlations among detectors are studied, on the time scale of a few tens of ns. The correlations include gamma-gamma, gamma-neutron, and neutron-neutron coincidences.

The paper presents results from the measurements, together with Monte Carlo simulations and analysis. Contamination of the signature by (alpha, n) reactions and detector cross-talk are analyzed and discussed. Finally, a method to determine Pu-240 mass is described. The masses were determined to ±1.3% using the Pu samples as calibrations. This method can be used in conjunction with gamma ray spectrometry to obtain the Pu-239 mass.

1. INTRODUCTION

The present paper describes the results of passive NMIS measurements performed jointly by ORNL and JRC staff on plutonium oxide samples that are stored at the Performance Laboratory of the JRC Ispra. The measurements took place in March and April, 2005 using the Nuclear Materials Identification System (NMIS) [1] with eight liquid scintillators having dimensions 25 by 25 by 8 cm each. The scintillators are sensitive to fast neutrons and gamma rays. The plutonium oxide samples have two compositions, denominated high and low burnup, and the mass varies from 8 g to 2500 g. The measurements were performed on the samples both inside and outside the AT-400 container. This paper describes the measurements performed on the containerized samples.

2. MEASUREMENT SETUP

The plutonium oxide powder is stored in cylindrical steel containers having variable height and diameter. In all the measurements, the sample heights were determined on the basis of an average plutonium oxide powder density of 2.6 g/cm^3. The AT-400 container (diameter 49.5 cm and height 49.5 cm) was placed on a 1 cm thick aluminum plate, at a height of 70 cm from the floor. The

sample containers were set at a height such that the center of the sample was at the same height as the center of the detector assembly. A lead shield with cylindrical shape and 1 cm thickness was placed around the sample containers.

A number of detector assemblies were used. In the first, four detectors were placed symmetrically around the AT-400 container. In the second, detectors were paired vertically, and the four pairs were placed symmetrically around the AT-400 container. Finally, two sets of four detectors were placed in a two by two array and the two sets were placed facing each other around the container. In all cases, the detectors were placed in contact with the container. A photograph of the measurement configuration with the two by two arrays of detectors is shown in Fig. 1.

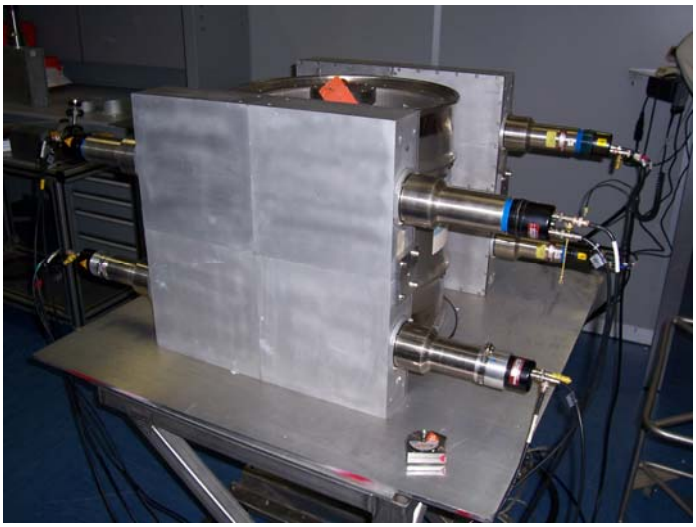

Figure 1. Photograph of measurement configuration showing AT-400 container and eight liquid scintillators, in two arrays.

For all the measurements, the signals from the detectors were input to constant fraction discriminators that eliminated pulses having amplitude less than 40 mV. This procedure eliminates pulses from gamma rays having energy less than 160 keV. The voltages on the detectors were set so that the Compton edge of a Cs-137 source had an amplitude of 120 mV, approximately.

3. RESULTS

The time correlations between signals from the detectors were acquired using NMIS. A typical signature is shown in Fig. 2. The signature is comprised of four different contributions that arise from combinations of neutron and gamma ray detections from the spontaneous fission events that occur inside the sample. The dominating contributions are from gamma, gamma and gamma, neutron pairs. Other reactions contribute to the correlation function, but their contribution is much smaller than the contribution from spontaneous fission. These reactions include induced fission, (alpha, n) neutrons and related gamma rays, and other beta decays of the isotopes present in the samples. The contribution of induced fission can be estimated using Monte Carlo and this is shown in Section 4.

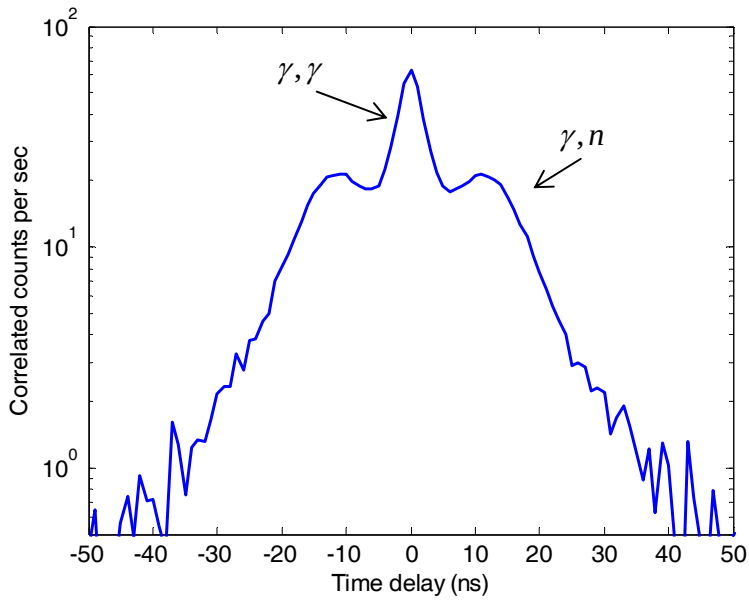

Figure 2. Cross correlation function of signals from two detectors facing each other, 2500 g Pu sample in AT 400 container.

Figure 3 shows the right half of the cross correlation functions of the signals from the two detectors facing each other in the measurement configuration given in Fig. 1.

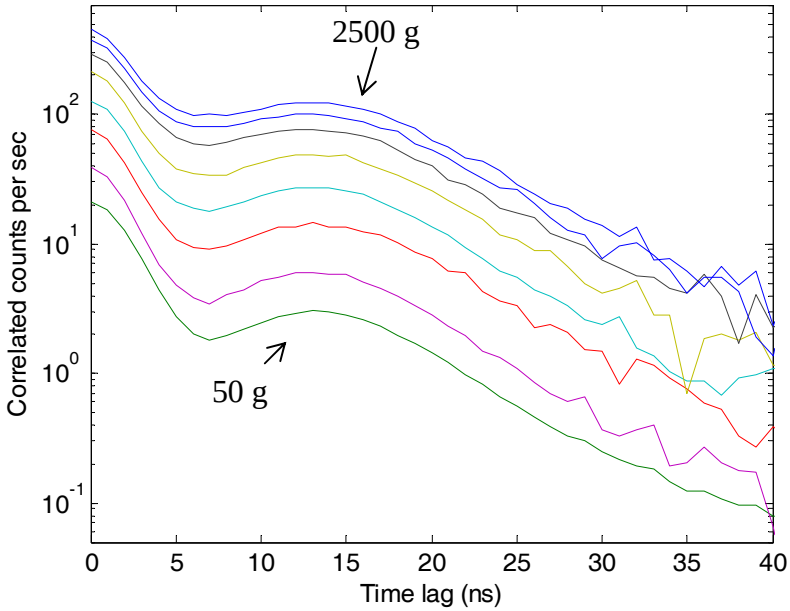

Figure 3. Cross-correlation functions for samples of total mass 50 to 2500 g. Positive and negative sides of the cross-correlations are averaged.

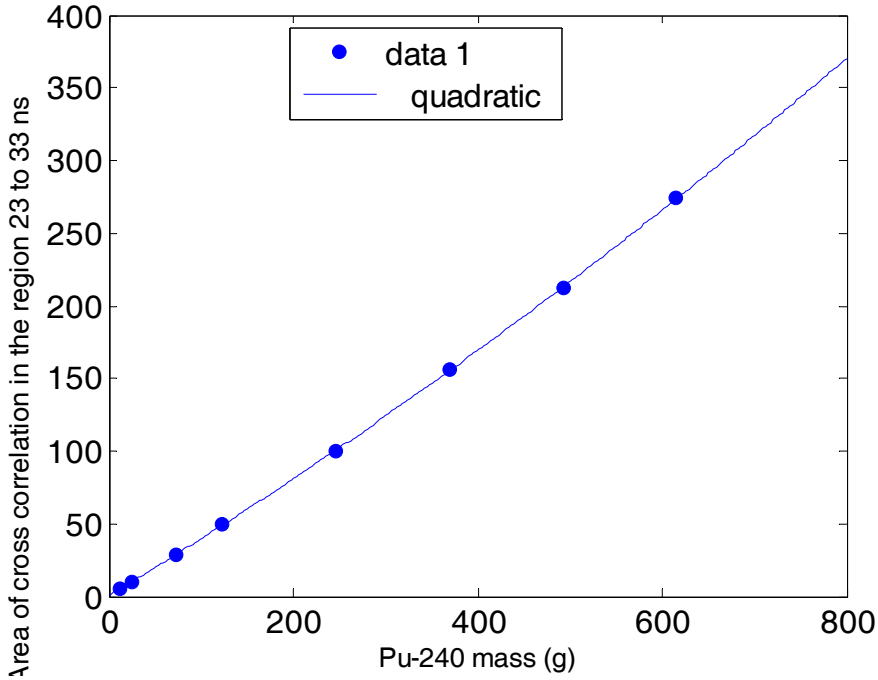

Figure 4. Area of the cross-correlation functions shown in Fig. 3 from 23 to 33 ns as a function of Pu-240 mass (g).

Figure 4 shows the integral of the cross-correlation function shown in Fig. 2 from time lags 23 to 33 ns as a function of the Pu-240 mass. The integration region was determined by minimizing the mean square error in the mass prediction for each possible integration region in the correlation time window. The mass prediction was assumed to have a quadratic form[1] of the type:

$$y = ax^2 + bx + c, \tag{1.1}$$

where x is the integral of the cross-correlation function of Fig. 3 and y is the Pu-240 mass in g. The coefficients for the fit are given in Table I.

Table I. Coefficients for Eq. (1.1)

a	*b*	*c*
1e-4	0.38	0.53

Figure 5 shows the error in the mass prediction using Eq. 1.1 with the coefficients of Table I. The error is less than 1.3% in the entire mass range of the samples, from 50 to 2500 g. Figure 6 shows the mean square error in the mass prediction for each possible integration region in the correlation time window.

[1] Other forms were explored but the quadratic gave the best results.

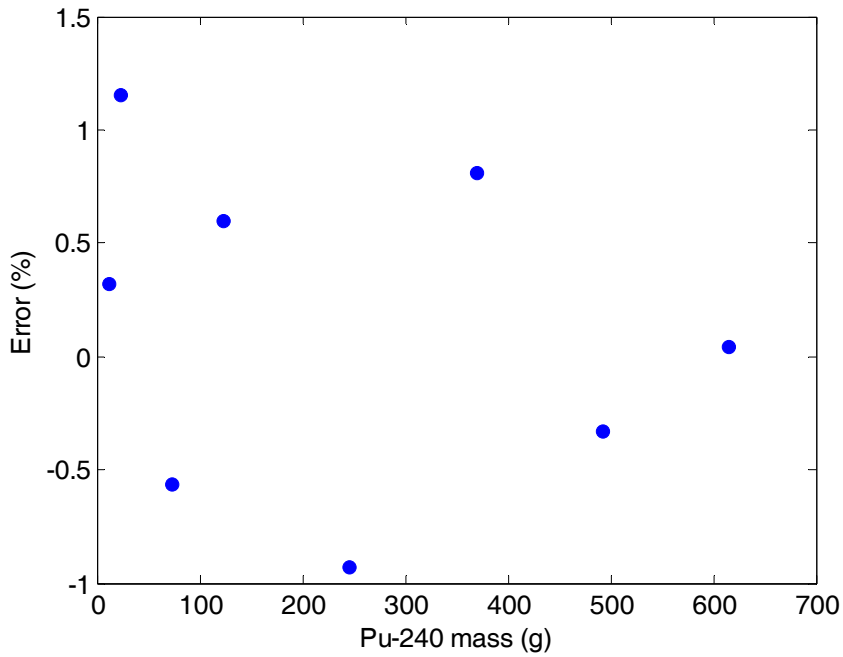

Figure 5. Percentage error in the prediction of Pu-240 mass using Eq. (1.1).

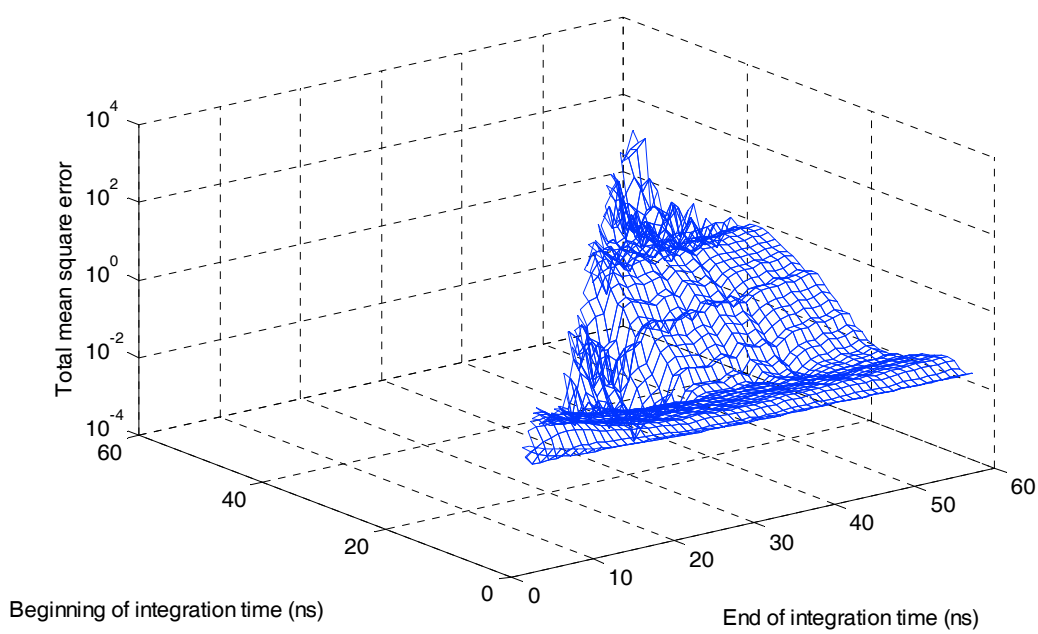

Figure 6. Mean square error in the prediction of Pu-240 mass using Eq. (1.1) for all possible integration intervals.

4. MONTE CARLO SIMULATIONS

The design and analysis of the measurements described in this paper rely on simulations performed with Monte Carlo methods. The measured signatures can be simulated using MCNP-PoliMi [2], a modification of the Monte Carlo code MCNP-4C. Figure 7. shows the measurement and corresponding simulation for a Cf-252 source placed at 100 cm from one liquid scintillator.

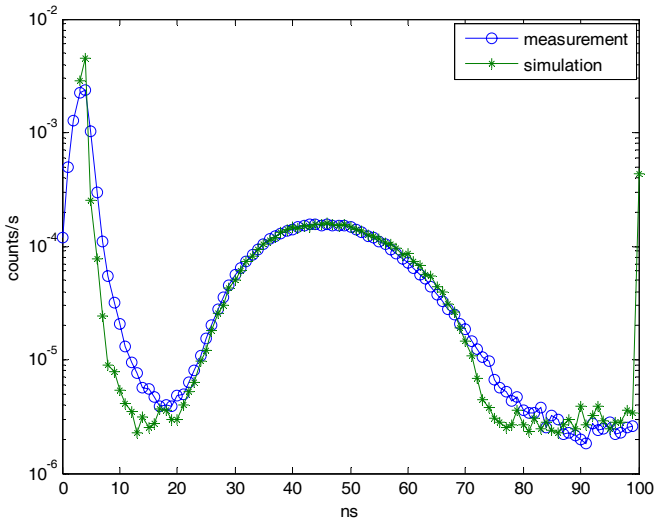

Figure 7. Measurement and simulation of a time of flight measurement with Cf-252 and a liquid scintillator.

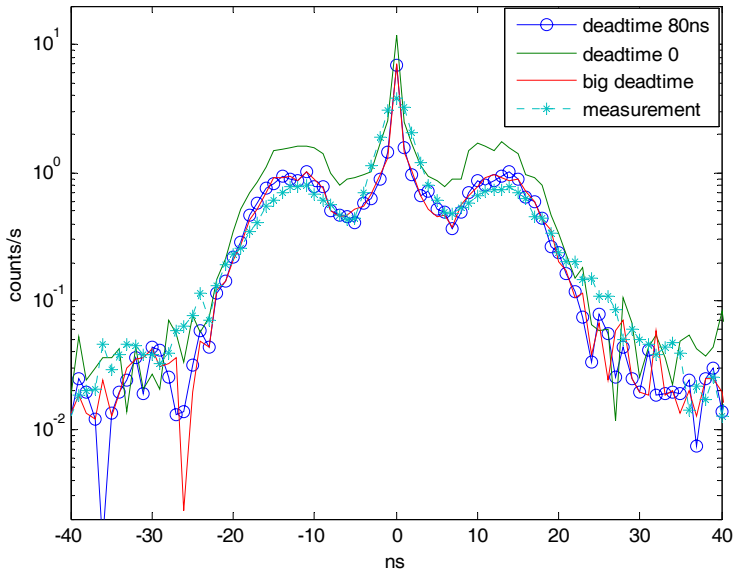

Figure 8 Measurement and simulation of a time of flight measurement with Cf-252 and a liquid scintillator.

5. CALIBRATION USING CF-252 SOURCE

When a series of samples of different masses is not available for the generation of a calibration curve such as that shown in Fig. 4, an alternative method is to use a Cf-252 source for calibration purposes. The prediction of the number of spontaneous fission events in the samples can be then obtained by scaling the measured feature *x* by an appropriate factor:

$$\chi = \frac{x_{Cf}}{x_{Pu}} \tag{1.2}$$

χ is related to the neutron and photon multiplicities emitted by Cf-252 and Pu-240. These are given in Table II. Factors such as multiplication and attenuation complicate the dependence of χ on these physical parameters.

Table II Average number of neutrons and photons from Cf-252 and Pu-240.

	Cf-252	**Pu-240**
Neutron	3.76	2.14
Photon	8.12	6.53

Because the number of spontaneous fissions of the Cf-252 source is known (f_{Cf}), the feature x_{Cf} can be normalized to obtain

$$x'_{Cf} = \frac{x_{Cf}}{f_{Cf}} \tag{1.3}$$

Then, the predicted number of spontaneous fissions in the sample (f_{Pu}) is given by

$$f_{Pu} = \frac{x_{Pu}}{x'_{Cf}} \chi \tag{1.4}$$

Use of Eq. (1.4), with χ measured using the data from the 1500 g sample, yields the estimate of the number of spontaneous fissions in the sample that is shown in Fig. 10. The relative error in the prediction is less than 6.6% for the sample masses from 50 to 2500 g. If the 2500 g sample is excluded, the relative error is less than 5% on the remaining samples.

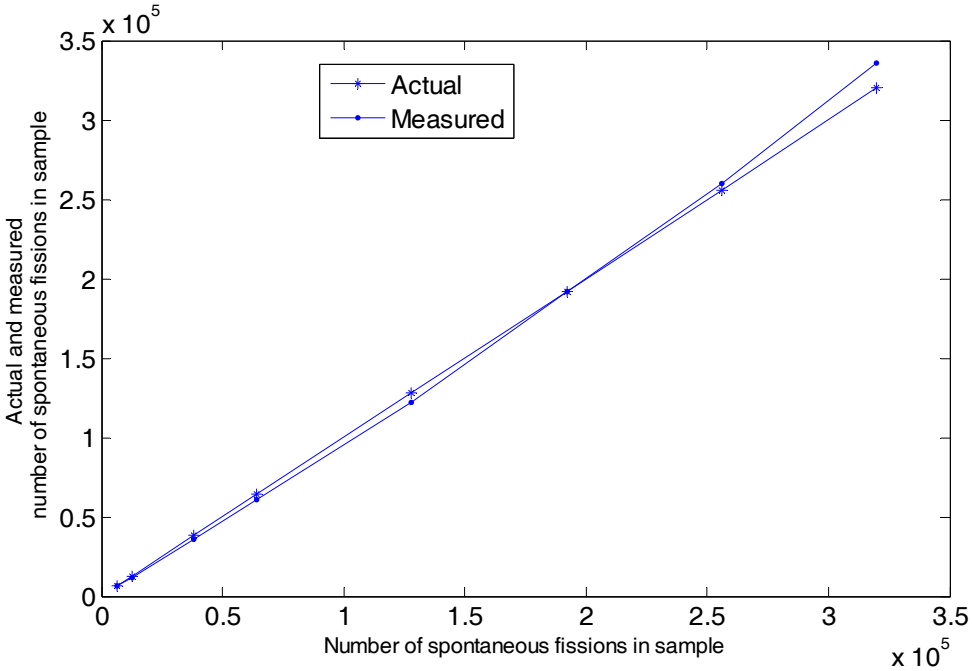

Figure 9 Actual and predicted number of spontaneous fissions in the samples using Eq. (1.4).

6. CONCLUSIONS

This paper presented the results and a preliminary analysis of the passive coincidence measurements performed on plutonium oxide samples stored at the Joint Research Center in Ispra, Italy. The analysis showed that a calibration curve can be produced to obtain estimates of the mass of plutonium in the samples. The error in the mass prediction was less than 1.3% in the mass range from 50 to 2500 g. The error in the prediction using a Cf-252 source and one of the samples as the calibration is less than 5% in the mass range 50 to 2000 g.

REFERENCES

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2. S. A. Pozzi, E. Padovani, and M. Marseguerra, "MCNP-PoliMi: A Monte Carlo Code for Correlation Measurements," *Nuclear Instruments and Methods in Physics Research A*, 513/3 pp. 550-558, 2003.