Journal of Nuclear Materials 385 (2009) 131–133

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Nondestructive determination of plutonium by gamma spectrometry and neutron well coincidence counting

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A B S T R A C T

Different NDA methods based on gamma ray and neutron measurements developed for the determination of Pu in solid samples is reported. In the gamma spectrometric measurements for solid samples, a method which takes advantage of the multiple γ-rays emitted by Pu and relies on the empirical relation between apparent mass of the sample and γ-ray energy was used. The method is applicable for the determination of small quantities of plutonium samples of non standard geometry by gamma ray spectrometry. The gross and coincidence neutron count rates for two different sets of standard Pu oxide powder samples were found to fall on different calibration lines. Isotopic composition of the two sets were determined using gamma ray spectrometry to obtain effective ^{240}Pu content in the samples. A common calibration curve could be obtained when coincidence count rates were plotted vs. effective ^{240}Pu content.

1. Introduction

Plutonium is a principal nuclear material whose isotopes produce characteristics gamma or neutron radiation spontaneously. Measurements of these radiations form the basis of nondestructive assay (NDA) of Pu in samples of different forms and shapes [1]. In the present work, we report different NDA methods based on gamma ray or neutron measurements developed for the determination of Pu in different types of samples. Assay based on the measurement of pulse height spectra of gamma rays from Pu samples have been employed for both solid and liquid samples. Assay based on measurement of neutron was done for solid samples. Since such measurements require isotopic composition of plutonium, a program was developed to determine the isotopic composition of plutonium.

2. Experimental

2.1. Estimation of Pu in solid samples by gamma ray spectrometry

A series of pure plutonium oxide powder standard samples were wrapped in paper and doubly sealed in polyethylene packets. The standards were prepared on weight basis. No independent analytical method was used to verify the amount of plutonium in the sample. The samples were rectangular in shapes (3 cm × 4 cm). The powders were distributed within the packet and there was no knowledge about the distributions. Hence, density and thickness of the samples were not known. The samples were rotated manually by 180° in steps of 45° and counting was done at each geometry to average out any asymmetry in the actual distribution of the oxide powders in the samples. The sample to detector distance was about 30 cm so that point source efficiency could be used for the determination of Pu amount. The standards were counted in a 20% coaxial HPGe detector coupled to 4 k channel analyzer. The resolution of the detector was 2 keV at 1332 keV. Cd filters (1 mm) were used to reduce the count rate of 59.54 keV ^{241}Am gamma rays. The gamma ray spectra were analyzed using the PHAST software [2]. The efficiency of the detector was obtained using standard point source of ^{152}Eu and ^{133}Ba.

2.2. Estimation of Pu by neutron counting

Neutron counting was done with two sets (A & B) of standard Pu oxide powder. Set A used in this counting was same as used for the gamma spectrometric determination of Pu. Set B was prepared separately by the same method as described above. The geometry of both the sets was same as given in the previous section. Samples were counted using a shift-register based Neutron Well Coincidence Counter having 24 ^{3}He counters of 2.54 diameter and 50 cm length arranged in an annular geometry. The counters were embedded in HDPE (moderator) of thickness 10.5 cm. The efficiency of the counter was 15%. Both gross as well as coincidence counting was done. A representative sample from each set was also counted on the 2cc HPGe detector having a thin Be window for gamma ray spectrum. This was required to obtain the isotopic composition [3] which is needed to obtain Pu amount from the coincidence counting data.

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doi:10.1016/j.jnucmat.2008.09.038

Table 1
Gamma ray energies and intensities of the nuclides used in the analysis.

Nuclide	Energy (keV)	Gamma ray intensity (%)
^{239}Pu	129.29	6.31E−03
	203.55	5.69E−04
	345.01	5.56E−04
	375.05	1.55E−03
	413.71	1.47E−03
^{241}Pu	208.00	5.34E−04
^{241}Am	125.30	4.08E−03

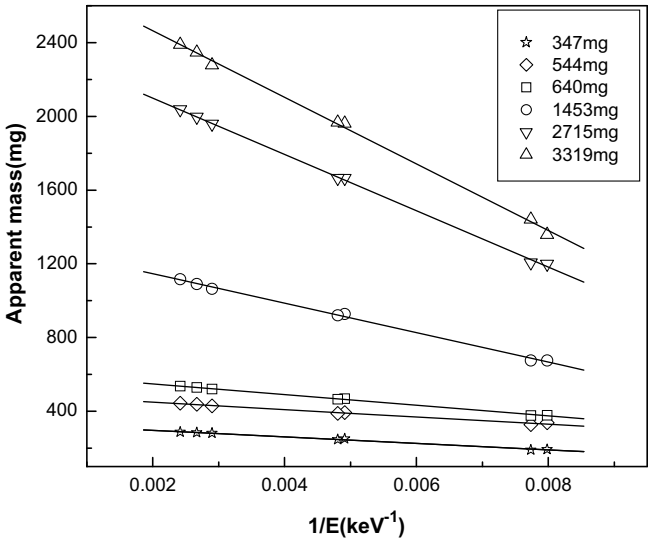

Fig. 1. The apparent mass of Pu as a function of 1/E for different samples, where E is the γ-ray energy in keV.

Table 2
The expected and the obtained Pu amount in the samples.

Expected amount (mg)	Amount obtained (mg)	% Deviation
640.06	611.48 ± 3.25	4.5
543.77	496.27 ± 5.19	8.7
346.65	335.44 ± 3.15	3.2
1453.03	1307.63 ± 9.32	10.0
2714.64	2409.17 ± 10.39	11.3
3319.36	2806.57 ± 18.48	15.4

3. Results and discussion

3.1. Estimation of Pu in solid samples by gamma ray spectrometry

The procedure of Venkataraman and Croft [4,5] was used to obtain the apparent mass of the Pu samples (m(E)) at the given energy E using the Eq.:

$$m(E) = C_{net}/(\varepsilon_\gamma I_\gamma W_A S_A) \quad (1)$$

where C_{net} is the net count rate of the γ-ray peak, I_γ is the intensity of the γ-ray, w_A is the weight fraction of the plutonium isotope

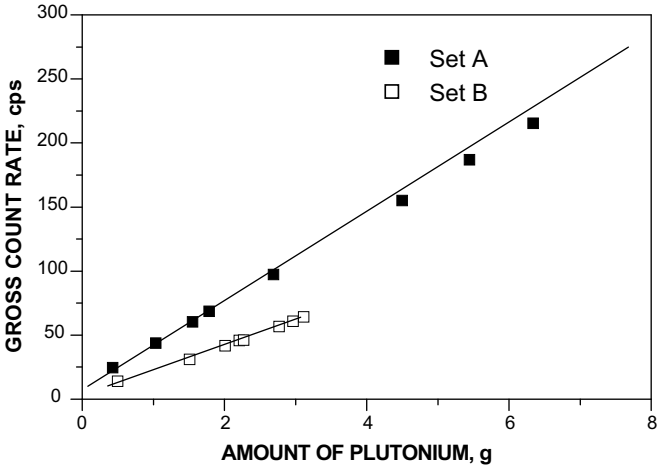

Fig. 2. Gross count rates for the two sets as a function of Pu amount.

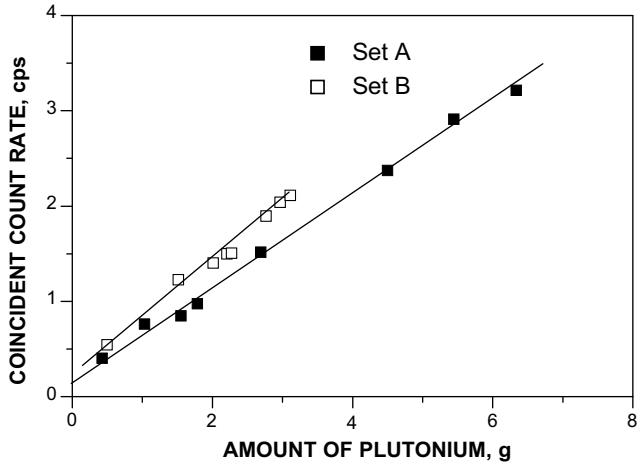

Fig. 3. Coincidence count rates for the two sets as a function of Pu amount.

in the sample emitting the gamma ray, ε_γ is the peak efficiency of the detector obtained using point ^{152}Eu and ^{133}Ba source and S_A is the specific activity of the isotope concerned. Table 1 gives the gamma ray energies of the nuclides and their corresponding gamma ray intensities used in this study.The apparent masses were fitted to the functional form:

$$m(E) = m_0(1 - B/E) \quad (2)$$

where m_0 is the true mass of the sample obtained as intercept, in the limit (1/E)→0 for which attenuation correction becomes unity. Fig. 1 shows the plot of m(E) vs. 1/E. The error bars in the Fig. 1 are of the size of the symbols and are due to counting statistics. It is seen that a good straight line fit is obtained for all the samples. Also, the slope of the curves systematically decreases with decreasing amount of the sample, ultimately becoming parallel to x-axis for less-attenuating samples. 'B' in Eq. (2) is treated as free parameter in the fitting. The apparent masses at different energies obtained

Table 3
Result of isotopic composition of two sets of Pu samples.

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
Set A	$(9.6 \pm 0.5) \times 10^{-3}$	94.16 ± 0.13	5.73 ± 0.13	$(66.5 \pm 0.4) \times 10^{-3}$	0.034	$(3.17 \pm 0.11) \times 10^{-1}$
Set B	$(1.2 \pm 0.2) \times 10^{-2}$	93.66 ± 0.18	6.18 ± 0.18	0.12 ± 0.001	0.034	$(0.46 \pm 0.09) \times 10^{-1}$

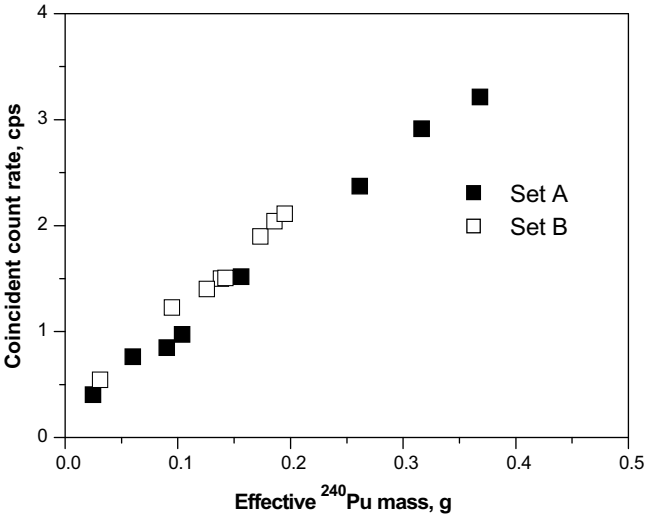

Fig. 4. Coincident count rates as a function of effective Pu amount.

in the present work for plutonium samples of different amount were fitted to Eq. (2) to obtain m_o for different samples. The values of m_o obtained for different samples and the expected values are given in Table 2. The errors on the measured Pu amount reflects the error on the intercept. It is seen that the m_0 values are mostly within 10% of the expected values. However, the deviations cannot be accounted for by the statistical uncertainty on the data, indicating some bias. Slight adjustment of the exponent of E in Eq. (2) could remove this uncertainty which was not attempted. This method is very useful for the determination of small quantities of plutonium samples where appropriate standard cannot be used due to inhomogenity of the real Pu-bearing samples.

3.2. Plutonium estimation by neutron counting

Fig. 2 shows the gross neutron count rates vs Pu amount of the two sets of standards counted. The two sets were known to have almost identical isotopic composition. They fall on different lines showing the gross neutron counting data is not sufficient to obtain the plutonium content in unknown samples. The corresponding coincident count rates vs Pu amount for the same set of samples is shown in Fig. 3. Here also the count rates from different sets do not fall on the same curve. It was observed that the set giving higher coincidence count rates give lower gross count rate for the same Pu amount. A representative sample of each set was subjected to isotopic analysis by gamma ray spectrometry. Table 3 gives the isotopic composition of Pu for the two sets of samples. ^{242}Pu content was obtained from isotopic correlations. It was seen set B has higher ^{240}Pu content, thus giving higher coincidence count rate as shown in Fig. 3. The isotopic composition was used to obtain effective ^{240}Pu mass. Fig. 4 shows the plot of coincidence count rate vs effective ^{240}Pu mass which falls on single curve. This shows the importance of knowledge of isotopic composition of Pu for obtaining Pu mass from neutron coincidence counting data. The higher gross count rate observed for set A can be attributed to the higher (alpha, n) reaction due to elevated ^{241}Am content in this set as observed from the isotopic composition data.

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