Concentration Of The ^{241}Pu In Air Samples From Chernobyl At Belgrade Site Estimated By a ^{241}Am In Growth Method

VUKANAC Ivana, NOVKOVIĆ Dušan, DJURAŠEVIC Mirjana, OBRADOVIĆ Zorica, KANDIĆ Aleksandar*

Institute for Nuclear Sciences Vinča, POB 522, Belgrade, Serbia and Montenegro

Abstract

The surface air samples collected in the first half of May 1986 at Vinča-Belgrade site were prepared and measured at the end of the 1991 and beginning of the 1992. year. Activity concentrations of the ^{137}Cs immediately after the Chernobyl accident were determined by means of gamma spectrometry, while the air activity concentration of ^{238}Pu and ^{239,240}Pu were determined by alpha spectrometry, after the plutonium radiochemical separation. The ^{236}Pu was used as a tracer. The same samples were remeasured after 13 years, during the 2004. The surface air activity concentrations of ^{241}Pu were estimated by a ^{241}Am in-growth method. The built up activities of ^{236}Pu progenies were determined from the recorded spectra and also calculated using the Bateman's equations. The ^{241}Am activity in the remeasured samples, obtained by complex spectral analysis was confirmed by gamma spectrometry. The ^{241}Pu activity concentration in measured air samples ranged from 240 μBq/m^3 to 7800 μBq/m^3. The average activity concentration ratio ^{241}Pu/^{239,240}Pu originated from Chernobyl accident was approximately 100.

1. Introduction

Immediately after the Chernobyl accident the largest contribution to the radioactivity of plutonium was due to ^{241}Pu ($t_{1/2}$ = 14.4 y). This is a pure beta-emitting isotope with a low maximum energy of 21 keV. The most convenient method for determination of ^{241}Pu concentrations is liquid scintillation counting. However, the daughter product of ^{241}Pu is the alpha and gamma emitting ^{241}Am ($t_{1/2}$ = 432.7 y), convenient for alpha and gamma spectroscopic measurements.

Over the past several years, there have been reported some results of measured activity concentrations of ^{241}Pu from environmental samples collected after the Chernobyl accident (Ikäheimonen, 1999; Mietelski, 1999; Holm, 1988). The data show that the isotopic ratio ^{241}Pu/^{239,240}Pu is much higher in the fallout from the Chernobyl nuclear power plant accident than the same ratio of plutonium isotopes originating from nuclear weapon tests.

In this paper we estimate the ^{241}Pu activity in old air samples collected during the first half of May 1986 at Vinča - Belgrade site by the ^{241}Am in-growth method (Ikäheimonen, 1999; Holm, 1988).

The activity of the built up ^{241}Am in the source was estimated using the measured activity of ^{228}Th, determined from the remeasured spectra and its daughter products in secular equilibrium (^{224}Ra, ^{220}Rn, ^{216}Po and ^{212}Po). Since the ^{236}Pu was used as a tracer, due to its relatively short half-life, spectra obtained almost 13 years after electrochemical separation, were complex in the energy region of ^{241}Am.

The contributions of ^{236}Pu progenies to the spectra were also calculated by application of the Bateman's equations (Evans, 1955). In both procedures the ^{238}Pu contribution to the peak area of ^{241}Am was estimated from the $^{239,\ 240}Pu$ content in the sources. The $^{238}Pu/^{239,\ 240}Pu$ activities ratios were calculated from the spectra previously recorded (Manić-Kudra et al., 1995).

Plutonium samples were also measured by means of gamma spectrometry. Activities of ^{241}Am were in good agreement with alpha specrometric measurements.

2. Experiment

The surface air concentrations of ^{238}Pu and $^{239,240}Pu$ at the Vinča-Belgrade site, obtained during the period May 01 to May 15, 1986, were measured in aerosol samples of total volume 576 m^3 - 683 m^3 collected by an air sampler over a period of 24h, and reported in a previously published paper (Manić-Kudra et al., 1995). The radiochemical procedure included the thermal mineralization of aerosol filters, fusion with the ammonium hydrogen sulfate for dissolution of insoluble plutonium oxide, plutonium coprecipitation with iron(III) hydroxide and separation by anion exchange resin (Dowex 1-X8, chloride form, (100-200) mesh). An activity of 0.1 Bq of ^{236}Pu per sample was added as a tracer during sample preparation after the thermal mineralization. The samples were radiochemically treated from December 1991 to February 1992, and measured immediately after plutonium fraction separation and electro-deposition. The previous experimental setup was described in Paligorić et al. (1989), Manić-Kudra et al. (1995) and Jerome et al. (1995).

The recent measurements were performed by a Canberra 7401 alpha spectrometer, incorporating a silicon surface-barrier detector with energy resolution of 21 keV (300 mm^2) and an Ortec 916 MCA. The energy and efficiency calibration of the spectrometer was performed by using a standard alpha spectrometric $^{239,\ 240}Pu$ source. The detection efficiency was 15.29 %.

Details of the applied ^{241}Am in-growth method will be explained by reference to the alpha spectra of plutonium source, separated from the

air sample collected on May 05, 1986 and measured on January 27, 1992 (Fig. 1) and on October 10, 2004 (Fig. 2). The spectrum primarily shows peaks originating from plutonium isotopes separated from the air sample (plus ^{236}Pu, used as a tracer). Smaller peaks due to ^{237}Np and ^{210}Po are also recorded as expected, due to the applied radiochemical treatment (Paligorić et al., 1989; Manić-Kudra et al., 1995).

The same sample was remeasured 12.9 years after the first measurement; its spectrum is presented in Fig. 2. After almost 4.6 half-lives of the ^{236}Pu source ($t_{1/2}$ = 2.851 y), not only could its alpha peak be detected but also the peaks of its daughter products. The peak of ^{237}Np in the plutonium fraction was also observed. The complexity of the obtained spectra in the energy region of ^{241}Am is due to ^{238}Pu and the progenies of ^{236}Pu used as a tracer, i.e. ^{232}U and ^{228}Th. Thus, the spectral analysis, i.e. estimation of built up ^{241}Am activity, was very complicated and will be explained below.

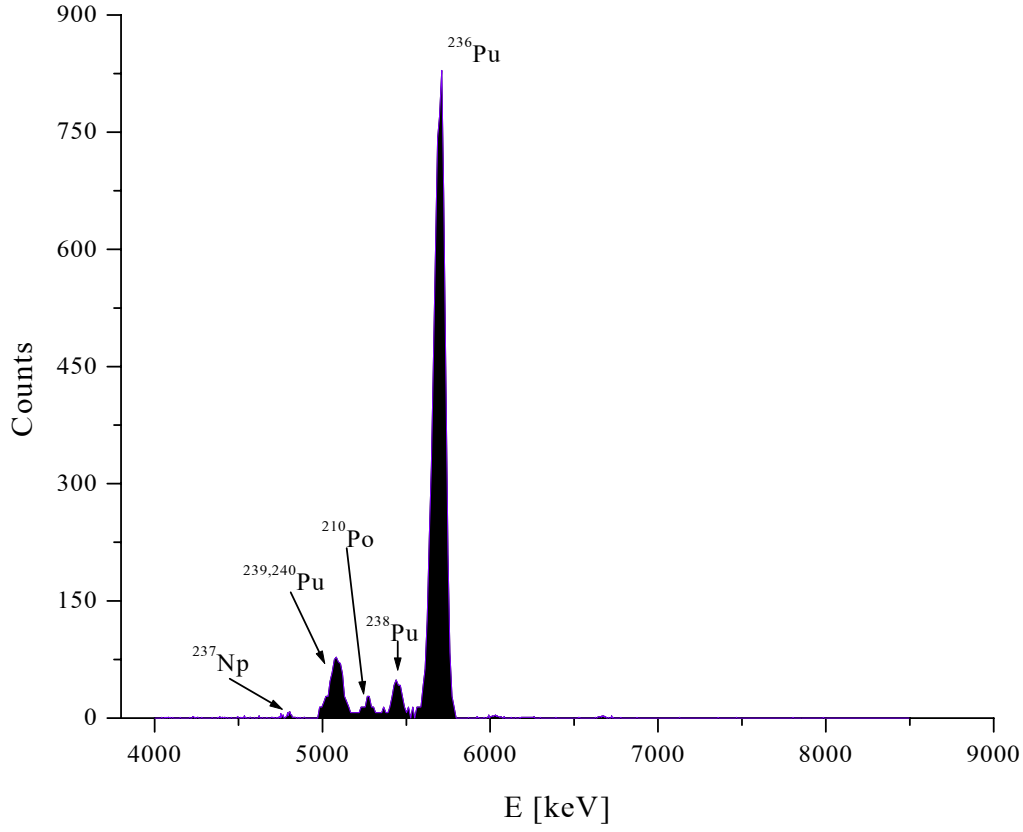

Fig.1. Alpha spectrum of plutonium isotopes separated from the air sample collected on May 05, 1986 and measured (t = 2.5 x 10 ^{5} s) on January 27, 1992.

The gross area of the energy region from 5.4 MeV to 5.55 MeV contains the following alpha peaks: ^{224}Ra (5.449 MeV, 4.9 %), ^{228}Th (5.423 MeV, 72.7 %), ^{238}Pu (5.465 MeV, 28.3 % and 5.499 MeV, 71.6 %) and ^{241}Am (5.443 MeV, 12.8 % and 5.486, 85.2 %). ^{224}Ra and ^{228}Th are the

progenies of ^{236}Pu. In order to estimate the contributions of ^{224}Ra and ^{228}Th in the energy region of ^{238}Pu and ^{241}Am, we evaluated the peak area of ^{224}Ra from the interference peaks areas of ^{224}Ra and ^{236}Pu. To resolve these two peaks we fitted the left end of the ^{236}Pu peak with a Gaussian (Sánchez et al., 1996; Bortels and Collaers, 1987). Since the ^{236}Pu daughter products, ^{228}Th and ^{224}Ra, are in secular equilibrium, their activities are the same. This calculation was confirmed by activities obtained from the well defined peaks of ^{220}Rn, ^{216}Po and ^{212}Po. Their activities were the same within the limits of the statistical error.

The gross area in the energy interval from 5.4 MeV to 5.55 MeV was 731 counts (the spectrum shown in Fig. 2.). The contribution of ^{238}Pu was 181 counts. This value was calculated from the previously measured activities ratio ^{238}Pu/^{239,240}Pu (Manić-Kudra et al., 1995). The activity of ^{228}Th was determined from the ^{224}Ra peak (5.685 MeV). The contribution of ^{228}Th in this energy region was 104 counts. The contribution of ^{224}Ra line at 5.449 MeV was 7 counts. The sum of these values and background were subtracted from the total counts obtained in the observed energy range, i.e.:

$$a\left(^{241}Am\right)=\frac{N_{tot}-N\left(^{228}Th\right)-N\left(^{224}Ra\right)-N\left(^{238}Pu\right)-B}{\varepsilon\cdot\Delta t},$$

(1)

where:

N_{tot} is the gross area in the energy region from 5.4 MeV to 5.55 MeV,

N(^{228}Th), N(^{224}Ra), and N(^{238}Pu) are the contributions of these radionuclides in the observed energy range,

▫ is the total counting efficiency,

Δt is measurement time and

B is background.

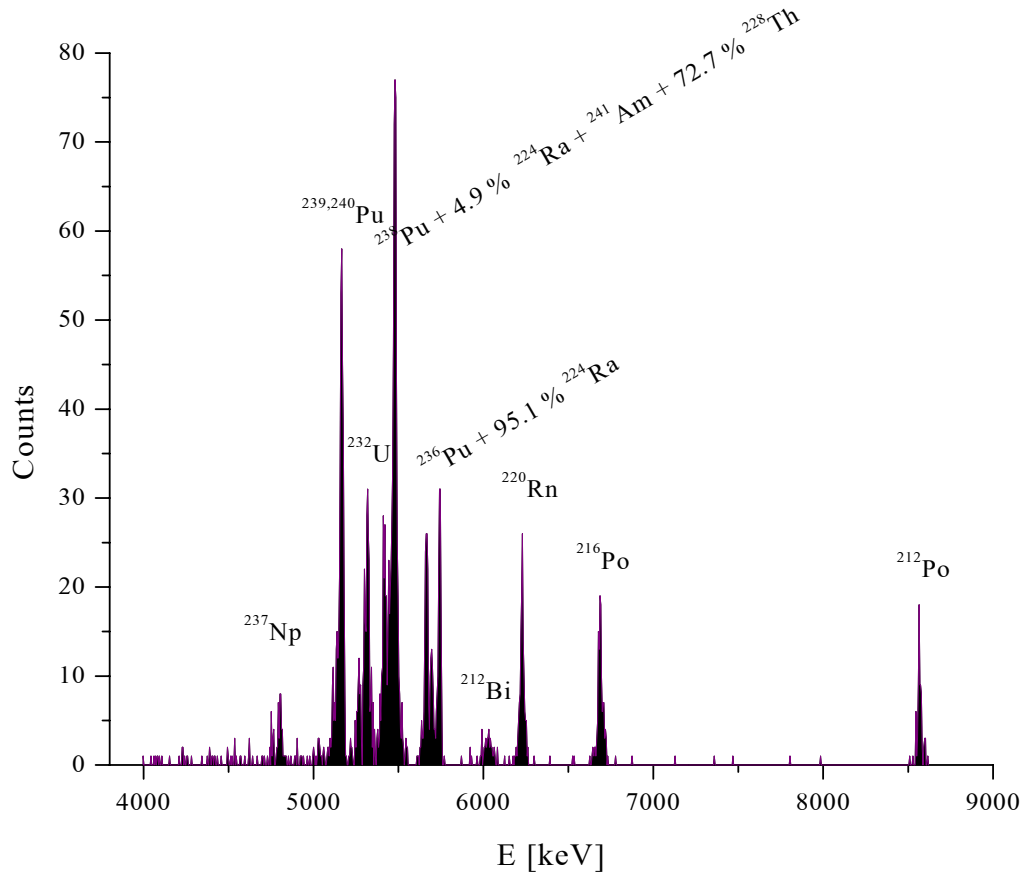

Fig.2. Alpha spectrum of plutonium source from the air sample collected on May 05, 1986 and measured ($t = 5.3 \times 10^5$ s) on October 10, 2004.

The calculated activity of the built up ^{241}Am was 5.4 mBq for this sample. The simple radioactive decay equations lead to the value of 452 mBq of ^{241}Pu, i.e. 1474.36 μBq/m^3 on May 05, 1986. The value of ^{241}Pu/$^{239, 240}$Pu activity ratio was 95 for this sample.

The same procedure was performed for the other measured sources, and the obtained activity ratio values, shown in Table 1, were in the limits quoted in the literature (Jerome et al., 1995).

Table 1. The results obtained by spectral analysis of the remeasured samples; the errors are quoted at a confidence level of 95 %.

collection date	^{241}Am activity [mBq]	^{241}Pu activity* [μBq/m^3]	^{239,240}Pu activity [μBq/m^3]	^{241}Pu/$^{239, 240}$Pu*
May, 01-02 1986	5.2 ± 0.9	1662 ± 57	12.2 ± 1.5	137
May, 03 1986	10.5 ± 1.4	3461 ± 91	38.5 ± 3.1	90
May, 04 1986	2.3 ± 0.7	7775 ± 43	69.3 ± 14.9	112

May, 05 1986	*5.4 ± 0.9*	*1474 ± 56*	*15.6 ± 1.6*	*95*
May, 12 1986	*1.3 ± 0.7*	*360 ± 45*	*5.0 ± 1.0*	*72*

(on collection date)*

3. Application of Bateman's equations

To confirm the validity of the obtained results we have developed a simple algorithm based on the Bateman's equations for calculation of the ^{241}Am in-growth in the sample. For this calculation only the activity of ^{236}Pu tracer and the gross peak area of ^{241}Am and ^{238}Pu in the energy region from 5.4 MeV to 5.55 MeV are necessary. The obtained results with the total error are given in Table 2.

Table 2. The results obtained by application of Bateman's equations; the errors are quoted at a confidence level of 95 %.

collection date	*^{241}Am activity [mBq]*	*^{241}Pu activity* [μBq/m^3]*	*^{241}Pu/^{239,240}Pu**
May, 01-02 1986	*5.5 ± 0.9*	*1771 ± 60*	*145*
May, 03 1986	*11.4 ± 1.7*	*3774 ± 106*	*98*
May, 04 1986	*2.3 ± 1.5*	*7803 ± 97*	*113*
May, 05 1986	*5.3 ± 0.8*	*1473± 54*	*95*
May, 12 1986	*0.9 ± 0.2*	*232 ± 12*	*47*

(on collection date)*

4. Results and discussion

The activity concentrations obtained from the spectra and by application of Bateman equations are approximately the same. The values for ^{241}Pu/^{239,240}Pu activity ratios ranged from 50 to 150. The mean value of this ratio was 101.23. The ^{241}Pu concentration in ground level air ranged from 0.2 mBq/m^3 to 7.8 mBq/m^3.

The results obtained in the two described ways are in good agreement. The spectral analysis is more complicated due to complexity of the plutonium isotopes spectra and ^{236}Pu progenies. The application of Bateman's equations is faster and easier and gives results with satisfying accuracy.

The standard procedure to confirm the quantity of ^{241}Am built up in the sample is its radiochemical separation from the source. Our attempts to extract the americium fraction failed, because it was firmly attached to an iron lattice due to burning up. In order to fix the source to the support, after the electrodeposition the source support (stainless steel disc) was heated to the red heat. In this way possible contamination of the detector was avoided.

Activities of ^{241}Am obtained by means of gamma spectrometry show a good agreement with those obtained by alphaspectrometric analysis.

5. Conclusion

The concentrations of ^{241}Pu in air samples in Belgrade after the Chernobyl accident were estimated by in-growth of ^{241}Am in electroplated plutonium fraction sources. The quantities of ^{241}Am were determined by spectrum analysis or by application of Bateman's equations. The input data for this calculation are gross areas in the energy range from 5.4 MeV to 5.55 MeV and initial activity of ^{236}Pu tracer. Although the ^{236}Pu tracer is not suitable for application of the ^{241}Am in-growth method for ^{241}Pu activity determination because of its progenies interference with ^{241}Am peak, it is still possible to obtain acceptable results. Both methods for determination of ^{241}Am activity give results that are in good agreement with each other. The usual confirmation of the obtained Am activity (Holm, 1988) could not be applied for our sources due to their burn up.

6. References

Bortels, G., Collaers, P., 1987. Analytical Function for Fitting Peaks in Alpha-particle Spectra from Si Detectors. Appl. Radiat. Isot. 38, 831-837.

Evans R.D., 1955. The Atomic Nucleus, McGraw-Hill Book Company, Inc. New York.

Holm, E., 1988. Determination of ^{241}Pu in Environmental Samples by a Radiochemical Procedure. Environment International 14, 363-365.

Ikäheimonen, T.K., 1999. Measurement of ^{241}Pu in Environmental Samples. Journal of Radioanalytical and Nuclear Chemistry. 243, 535-541.

Jerome, S. M., Smith, D., Woods, M.J., Woods, S.A., 1995. Metrology of Plutonium for Environmental Measurements. Appl. Radiat. and Isot. 46, 1145-1150.

Manić-Kudra, S., Paligorić, D., Novković, D., Smiljanić, R., Milošević, Z., Subotić, K., 1995. Pluonium Isotopes in the Surface Air at Vinča-Belgrade site in May 1986. Journal of Radioanalytical and Nuclear Chemistry, Letters 199, 27-34.

Mietelski, J.W., Dorda, J., Was, B., 1999. Pu-241 in Samples of Forest Soil from Poland. Appl. Radiat. Isot. 51, 435-447.

Paligorić, D., Kudra, S., Smiljanić, R., Novković, D., Milošević, Z., Zarić, M., 1989. Determination of Fallout at Location of the Boris Kidrič Institute - Vinča. 30th Anniversary Symposium of Radiation Protection, October, 02-06 1989, Dubrovnik, Yugoslavia.

Sánchez, A.M., Tomé, F.V., Montero, P.R., 1996. Fitting of Alpha Spectra. Application to Low-level Measurements. Appl. Radiat. Isot. 47, 899-903.