Analysis of Plutonium containing Materials by Gamma Spectrometry*

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High resolution gamma ray spectrometry (GS) is becoming a very important technique in the analysis of materials containing plutonium and the accompanying actinides. The isotopes of uranium and transuranium elements emit mainly alpha particles. The probabilities of gamma radiation emission of plutonium isotopes are three or more orders of magnitude lower, but the intensities of this radiation can be measured with a standard deviation better than 1% in the time range from 10 to 100 min depending on the mass of the sample. In this work the gamma radiation in the energy range from 40 to 312 KeV has been used. However, energies below 60 KeV can only be used if ^{241}Am is separated from plutonium before the measurement. The plutonium isotopes with the atomic masses from 238 to 241, ^{241}Am, ^{237}Np, and the ^{235}U/Pu ratio in solution and U/Pu ratio in solid samples can be determined with this method without removing the material from a container if its walls are not too massive to absorb the gamma radiation.

The measuring system was as follows: a high purity germanium detector (HPGE) of the planar type (1500 mm^2 or alternatively 500 mm^2 area and 13 mm thick); an amplifier with shaping time constants 6 μs; a pile-up rejection and dead time correction unit when necessary; energy resolution 565 eV and 530 eV, respectively; and a multichannel analyser with 8192 channels covering an energy range from 0 to 614 KeV with zero and gain stabilization. The spectra were transmitted to a PDP 11/44 computer for further evaluation.

To attenuate the intense ^{241}Am peak of 59 KeV, a cadmium absorber was used. The absorber thickness required depends on the age and the isotopic composition of the sample. Its thickness was from 0.5 to 1.5 mm, and the 59 KeV peak should be attenuated to have a height comparable to the peaks in the 100 KeV region. For plutonium concentration measurement in solutions, very good reproducibility is required; a specially constructed sample holder was used.

The following types of plutonium samples have been measured: (1) plutonium products as dried residue from reprocessing plants in penicillin vials, plutonium content about 4 mg; (2) mixed uranium and plutonium oxides or plutonium oxide as pellets or powder in plastic bottles (2–7 g); (3) diluted solutions of plutonium of about 100 ml volume and 4 mg content in plastic bottles; (4) solutions of plutonium in plastic containers with a flat bottom and highly reproducible shape for plutonium concentration determination, 10 ml volume, 0.5 to 6 mg plutonium per container; (5) solutions of plutonium after ^{241}Am separation, solution volume 1 ml, plutonium content about 100 μg.

The programs used for spectra evaluation were provided by Ray Gunnink of the Lawrence Livermore National Laboratory and have been described in detail elsewhere [1, 2]. The following programs have been used: (a) the MGA2 (*M*ulti-*G*roup-*A*nalysis) for samples of types(1), (2), and (3); it calculates the isotopic abundances of ^{238,239,240,241}Pu (^{242}Pu content can, if known, be declared or it can be calculated from isotopic correlations between ^{239,240,241}Pu against ^{242}Pu [3]), ^{241}Am, ^{237}Np (above 0.8%), ^{235}U/Pu ratio, total U/Pu ratio (only for solid samples); (b) the PUANAL (*Pu Anal*ysis) for samples of type (4); it calculates the plutonium concentration, its isotopic composition and ^{241}Am; (c) the LEPA (*L*ow *E*nergy *P*lutonium *A*nalysis) for the samples of type (5); it calculates the isotopic composition of plutonium using low energy gamma rays (below 59 KeV).

The results can be summarized as follows:

(1) The results of isotopic composition of plutonium calculated with MGA2, based on mass spectrometry (MS) values for ^{242}Pu in 15 samples, agree within 1% with MS values of ^{239,240,241}Pu and within 4% of the alpha spectrometry (AS) of ^{238}Pu (only in three cases the difference is more than 1%). When the ^{242}Pu abundance is calculated from the isotopic correlation, the agreement for ^{238,239,240,241}Pu is within the same range, but ^{242}Pu is only within ±15%.

(2) The ratios of ^{235}U/Pu and U/Pu calculated with MGA2 for 15 samples agree with those determined by titrimetry (and MS in the case of ^{235}U/Pu) within 5%.

(3) The results of ^{241}Am concentration calculated with MGA2 for 15 samples agree within 3 to 10% with those of ^{241}Am after separation and its determination with GS by the 59 KeV measurements against standards (the accuracy of this measurement is ±2%).

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(4) The results of ^{237}Np determination calculated with MGA2 for 15 samples are within 10% of the results from the measurements of the ^{233}Pa 312 KeV gamma line which are measured with ±2% accuracy resulting from counting statistics; uniform distribution of ^{233}Pa in the measured sample is assumed.

(5) The results of plutonium concentration determination in 10 ml volume performed on eight solutions of reference materials SRM947 and SRM948 and calculated with the PUANAL program agree within 1% with those by titrimetry.

(6) The results of plutonium isotopic composition determination calculated with the LEPA program for 15 freshly separated plutonium samples agree within 1% with MS values in most cases for $^{239,240,241}Pu$, and within 4% with AS values for ^{238}Pu (these results are in all cases too high).

References

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3 R. Gunnink, *Nucl. Mat. Manag., 9,* 83 (1980).