Transactions of the Korean Nuclear Society Spring Meeting
Chuncheon, Korea, May 25-26 2006

Calculation of Minimum Detectable Activity for a Measurement of Pu Isotopes in Environmental Samples

Myung Ho Lee, Jong Yun Kim, Kyoung Kyun Park, Kih Soo Joe,
Young Jai Park, Won Ho Kim

Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute

Yuseong-gu, Daejeon 305-353, Korea
mhlee@kaeri.re.kr

1. Introduction

The detection limits are based upon the ratio of two measurements of an efficiency and background for a given instrument. The minimum detectable activity (MDA) is a level (not a limit) of activity concentration which is practically achievable by an overall measurement method, while the detection limits only provide information about the intrinsic performance of the instruments [1-4].

Disintegration of a radioactivity is a random process which can be described accurately by the statistical decay law only when the number of observed events is large, for example, in a high level of radioactivity sample. However, in a very low level of radioactivity in environmental samples, the results of radioactivity measurements have a possibility for a misinterpretation and inappropriate conclusions due to a random statistical fluctuation of the count rate of the background. Therefore, it is necessary to set up a reliable detection limit or MDA of the radionuclides in a very low level of radioactivity for environmental samples to help determine whether the measured radioactivity concentration is a real radioactivity level or background.

The purpose of this study was to investigate the parameters which affect the MDA values of Pu isotopes and to calculate the MDA values by considering the measuring times of the background and the sample for environmental samples.

2. Methods and Results

2.1 Determination of the Pu isotopes in environmental samples

The Pu isotopes in the environmental samples were purified with an anion exchange resin and electroplated onto stainless steel platelets [5], and then, ^{239,240}Pu and ^{238}Pu were measured by an alpha spectrometer. After an α-spectrometric measurement of the Pu isotopes on the stainless steel planchet, the Pu isotopes were withdrawn into an 8 M HNO_3 solution. ^{241}Pu was measured [6] by a liquid scintillation counter (Quantulus 1220), after removing the hindrance elements such as Fe and Ni with an exchange resin.

2. 2 Calculation of the MDA for the Pu isotopes

In the determination of ^{239,240}Pu and ^{238}Pu for the environmental soil samples, the values of the average chemical yield (65 %), background count rate (0.002 cpm) and counting efficiency (23 %) for the alpha spectrometer the values of the MDA for ^{239,240}Pu and ^{238}Pu were expressed by using Eq.(1). Also, after inserting the values of the average chemical yield (65 %), background count rate (1.5 cpm) and counting efficiency (41 %) for the liquid scintillation counter into Eq.(3), the values of the MDA for ^{241}Pu were expressed by using Eq.(2).

$$MDA_{Pu-239,240}(Bq/S) = \frac{2.71 + 0.147(t_S)^{\frac{1}{2}}\left(1+\frac{t_S}{t_B}\right)^{\frac{1}{2}}}{t_S \cdot 8.97 \cdot S} \quad \text{--------(1)}$$

$$MDA_{Pu-241}(Bq/S) = \frac{2.71 + 4.03(t_S)^{\frac{1}{2}}\left(1+\frac{t_S}{t_B}\right)^{\frac{1}{2}}}{t_S \cdot 15.99 \cdot S} \quad \text{----------(2)}$$

2.3 Parameters for the determination of the MDA value for the Pu isotopes

With an increasing sample mass, the MDA was dramatically decreased, as shown in Fig. 1. The MDA of the Pu isotopes was calculated from Eqs. (1) and (2), where the background counting time is equal to that of the sample. The counting time was set to be 1,000 min for the alpha-emitting nuclides, and 500 min for the beta-emitting nuclides, respectively. The MDA for ^{241}Pu was higher than that for ^{239,240}Pu and ^{238}Pu, since the background count rate of the liquid scintillation counter (μ_B = 1.5 cpm) was much higher than that of the alpha spectrometer (μ_B = 0.002 cpm) as well as the counting time of the liquid scintillation counter (t_s = 500 min) which was shorter than that of the alpha spectrometer (t_s = 1,000 min), although the detector efficiency of the liquid scintillation counter (E = 41%) was higher than that of the alpha spectrometer (E = 23%).

The MDA values with the detector efficiency and the chemical yield were found to be within a narrow range. The effect of the detector efficiency and the chemical yield on the MDA values was insignificant when compared to that of the sample amount because the variations of the detector efficiency and the recovery yield were so small that they were reported as reproducible for environmental samples. The MDA

decreased more rapidly with the sample amount than with the counting time. Thus, increasing the sample amount is more effective for reducing the MDA values than increasing the counting time in the MDA calculation, when taking into account that the background count rate may increase by increasing the counting time.

2.4 MDA values of the Pu isotopes by varying the counting time of the sample and background

In a Pu isotope analysis for environmental samples, it is very important that the background and sample counting times are chosen carefully. In a general radioactivity analysis for environmental samples, the background and sample counting times are set as 1,000 min to calculate the MDA of ^{239,240}Pu and ^{238}Pu. But in the case of the samples containing a highly activity concentration of the Pu isotopes, it is not necessary to measure it for such a long time, and therefore a selection of the background and the sample counting times should be optimized to determine more accurate MDA values.

The MDA values of ^{239,240}Pu and ^{238}Pu calculated at a fixed background counting time (t_b = 1,000 min) were decreased by increasing the sample counting time from 50 to 10,000 min. The MDA values of ^{239,240}Pu and ^{238}Pu were decreased by increasing the background counting time at a fixed counting time of the sample. Also, the MDA values of ^{241}Pu were decreased by increasing the background and sample counting times. The initial slope (-6.9×10^{-4}) of the plot by varying the background counting time was smaller than that (-2.7×10^{-3}) by varying the sample counting time for ^{239,240}Pu and ^{238}Pu. The tendency of the MDA of ^{241}Pu by varying the background and sample counting times was similar to that of ^{239,240}Pu and ^{238}Pu. The MDA at a fixed 1,000 min of the background counting time by varying the sample counting time range from 50 to 750 min was higher than that at a fixed 1,000 min of the sample counting time by varying the background counting time in the range of 50-750 min. However, the MDA at a fixed 1,000 min of the background counting time by varying the sample counting time in the range of 2,500-10,000 min was slightly lower than that at a fixed 1,000 min of the sample counting time by varying the background counting time in the range of 2,500-10,000 min. Therefore, if an urgent result for the Pu isotopes is needed in an emergency situation or it is not necessary to measure the Pu isotopes for a long time due to the high activity concentration of the Pu isotopes in the samples, it is proper to determine the MDA at a fixed sample counting time (1000 min) by changing the background counting time (50-750 min). Also, if the counting time requires more than 1,000 min due to a very low activity concentration of the Pu isotopes in a sample, to obtain a lower MDA value, the MDA should be determined at a fixed background counting time (1,000 min) by changing the sample counting time (2,500-10,000 min).

3. Conclusion

In the analysis of radioactive nuclides such as Pu, Am, and Sr in a low-level environmental sample, lots of factors are involved in the MDA determination. Sample amount was the most effective parameter to reduce the MDA values among such parameters as the counting time of a sample and the background, the count rate of the background, the detector efficiency, and the chemical yield of the radionuclides.

Proper selection of the counting time of the sample and the background is very important to determine the MDA of the Pu isotopes. In the case of the counting time of more than 1,000 min, fixing the background counting time by varying the sample counting time is more effective whereas fixing the sample counting time by varying the background counting time is more reasonable in the case of a counting time of less than 1,000 min.

References

[1] B. Altschuler, B. Pasternak, Health Phys. 9 (1963) 293.
[2] L. A. Currie, Anal. Chem. 40 (1968) 586.
[3] B. S. Pasternack, N. H. Harley, Nucl. Instr. and Meth. 91 (1971) 533.
[4] J. H. Head, Nucl. Instr. and Meth. 98 (1972) 419.
[5] M. H. Lee, C. W. Lee, Nucl. Instr. and Meth. A 447 (2000) 593.
[6] M. H. Lee, C. W. Lee, Radiochim. Acta 84 (1999) 177.

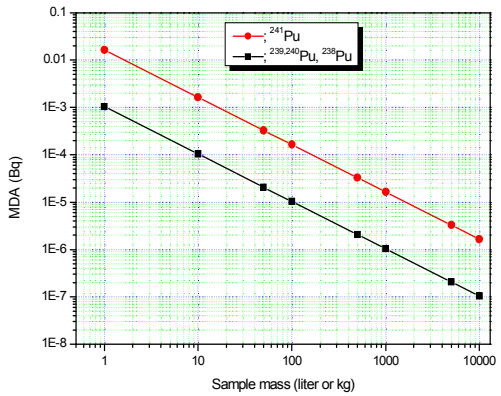

Fig. 1. MDA values of ^{239,240}Pu, ^{238}Pu and ^{241}Pu plotted vs the sample mass.