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Optimization of ^{241}Pu Analysis and Application to Environmental Samples

Myung Ho Lee, Yong Ho Choi, Sang Bok Kim, Kwang Hee Hong,
Chang Woo Lee
Korea Atomic Energy Research Institute

ABSTRACT

An optimized method for determining beta-emitting ^{241}Pu in the presence of alpha-emitting nuclides was developed using a liquid scintillation counting system. PSA-level was setting using pulse-shape discrimination. The ^{241}Pu counting channel was adjusted for maximum value of FM using the ^{241}Pu standard source. The volume of scintillant was determined for the maximum value of counting efficiency. The optimized method of ^{241}Pu has been applied to environmental samples to measure concentration of ^{241}Pu in soils and mosses. Also it has been identified the origin of Pu deposited in Korea from the activity ratio ^{241}Pu / $^{239,240}Pu$.

INTRODUCTION

Recently, modern liquid scintillation spectrometers equipped with pulse height analysis (PHA) and pulse shape analysis (PSA) techniques have been developed[1]. The low-level liquid scintillation counter (Quantulus 1220) has an anticoincidence detector and a massive lead shield to minimize the background radiation. It is also equipped with a pulse shape analyser, which allows discrimination between α and β pulses. The excitation produced in the scintillator depends on the type of radiation. β and γ radiations produce mainly singlet states, which fluoresce in 1-2 ns, whereas α particles produce more triplet

states, which can last 200-300 ns. The shape and duration of the resulting light pulses from the scintillator are different according to nuclides and the pulse shape analyser distinguishes the α and β/γ pulses by comparing the fall time of a light pulse with its amplitude. The use of PSA enables the new liquid scintillation spectrometers to measure the spectra of both α- and β-emitting nuclides simultaneously even at the low-levels present in environmental radioactivity. The purpose of this investigation is the optimization of the procedure for the determination of Pu-241 using liquid scintillation spectrometry, and then the optimized procedure was applied to environmental samples such as moss and soil samples.

EXPERIMENTAL AND METHODS

A low-level liquid scintillation specrometer, Quantulus 1220 (LKB, Wallac), was used for system optimization. A pure alpha and a pure beta sample as standards were used to set the pulse shape analyser. About 1000 dpm of ^{239}Pu was mixed with the water:cocktail having the ratio of 1 ml 0.1 M HCl to 19 ml Ultimagold AB (Packard Instrument Company) that is to be used in the actual analyses. Also a beta sample was prepared in the similar way by about 1000 dpm of ^{36}Cl because ^{36}Cl spectra overlap the alpha energy spectrum of ^{239}Pu. All measurements were carried out with 20ml Teflon vials. Pu was separated from moss samples using a radiochemical slovent extraction and anion exchange technique[2]. The purified Pu fraction was divided into two nearly equal parts: one part was subjected to beta (^{241}Pu) and total alpha counting by a low background liquid scintillation counter, and the other part was used for the measurement of other Pu isotopes (^{239,240}Pu, ^{238}Pu) by means of alpha-ray spectrometry(EG&G ORTEC).

RESULTS AND DISCUSSION

System optimization

To determine ^{241}Pu activities with the highest sentivity, the optimum counting regime

must first be configured. This configuration is affected by a variety of parameters such as α and β counting efficiencies, the pulse-shape analysis discrimination factor of the instrument, background in the region of interest and scintillant volume.

The discrimination of the α and β pulses is based on the well-known difference between the decayed components of their fluorescence. To determined the optimum PSA level, one pure alpha (^{239}Pu) and one pure beta (^{36}Cl) sample were measured in α and β window, respectively, varing the PSA level from 60 to 140. As shown in the Table 1, β percentages in α window for measurement of ^{239}Pu were increased with PSA level while α percentage in β window for ^{36}Cl were decreased. This spillover data were plotted in Fig. 1, which show typical alpha/beta crossover plot for ^{239}Pu and ^{36}Cl standards in Ultimagold AB cocktail. The optimum PSA level is where there is minimum spill of alpha counts into the beta MCA and beta counts into the alpha MCA. Therefore, the PSA level should be set at 100 for optimum counting. Under optimum conditions, misclassification of alpha events into beta MCA and beta into alpha MCA is less than 3%. As the figure of merit (FM), defined as the square of counting efficiency divided by the background in the region of interest (E^2/B), it was considered to be the best indicator of optium counting conditions. The ^{241}Pu counting channel was adjusted for maximum value of FM using ^{241}Pu standard source. L and R channel were adjusted to 23 and 289, respectively, in which the value of FM was maximized.

To establish the optimum volume of scintillant, we determined the optimum scintillant /water (0.1 M HCl) ratio using the total available volume of 20 ml. For this, the FM was calculated for different proportions of scintillant volume and water at the PSA setting of 100. The optimum scintillant volume was 19 ml, which is about the highest FM value.

Using a scintillant volume of 19 ml and PSA setting of 100, the counting efficiency for ^{241}Pu β particles (window 23-289) was 41 % and background count rate was between 1.5 and 2.0 cpm depending measuring time. The α particle efficiency was determined in a similar manner (window 500-800) and found to be 100% and the α background count

rate was 0.06-0.1 cpm. At a counting time 600 min., detection limit was 0.0095 Bq/sample and 0.0030 Bq/sample for ^{241}Pu and alpha activity, respectively.

^{241}Pu measurement of environmental samples

The optimized procedure of ^{241}Pu has been applied to environmental samples such as moss and soil samples in this study. The mean values with standard deviation in soils was 0.55 ± 0.19 (individual data ranged from 0.32 to 0.79) Bq/kg-dry for $^{239,240}Pu$, and 2.32 ± 1.08 (0.91 to 4.12) Bq/kg-dry for ^{241}Pu. In case of the moss samples, the mean values was 2.69 ± 2.60 (individual data ranged from 0.32 to 6.78) Bq/kg-dry for $^{239,240}Pu$, and 10.98 ± 9.51 (1.32 to 24.14) Bq/kg-dry for ^{241}Pu. The highest value for plutonium was found in moss on rock. The average activity concentration of Pu isotopes in moss is greater than its average activity in soil. The measured activity concentrations of Pu isotopes in moss samples are lower than those reported from Sweden[3] and Finland[4]. It may come from deposition influenced by meteorological condition and characteristics of moss such as species and life span of moss colony.

The activity ratio of ^{241}Pu / $^{239,240}Pu$ varies according to the source and can be utilized to identify the different sources of release. The activity ratio ^{241}Pu / $^{239,240}Pu$ in the releases from the Chernobyl accident, nuclear fuel reprocessing facilities, fallout from the 26th Chinese test and in weapon-grade plutonium is in the order of 85, 25, 5.5 and 3, respectively[5]. Data for ^{241}Pu and $^{239,240}Pu$ in environmemtal samples contaminated by global fallout have been published for example by Hakanen et al[1], Holm and Persson[3], Livingston et al[6], Fukai et al[7], Krey et al[8], and Holm et al[9]. The reported activity ratio of ^{241}Pu / $^{239,240}Pu$ was between 4 and 7 for different marine and terrestrial samples. The ^{241}Pu / $^{239,240}Pu$ activity ratios in soils and mosses were found to be in the range of 2.84 to 5.22 with a mean value of 4.2 ± 0.7, a little lower than reported activity ratio of ^{241}Pu / $^{239,240}Pu$ influenced by fallout from nuclear weapon testings. The small difference in the value of activity ratio can be explained by decay of

^{241}Pu ($t_{1/2}$=14.4 years). Reported activity ratio above mentioned was measured at less or more 1980. Correcting their results to 1995 and the estimated value agrees well with our results.

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Table 1. Percentage betas in ^{239}Pu and alphas in ^{36}Cl standard samples observed in the alpha and beta window as a function of PSA level setting in a Quantulus 1220

PSA Level	^{239}Pu (α)			^{36}Cl (β)		
	cpm in α window	cpm in β window	β percentage*	cpm in α window	cpm in β window	α percentage**
60	1274.4	4.6	0.36	742.1	267.3	73.52
70	1286.7	12.9	0.99	598.2	573.1	51.07
80	1254.4	14.3	1.13	396.7	1156.3	25.52
90	1264.8	25.0	1.94	134.5	1322.9	9.23
100	1281.7	36.8	2.79	43.2	1684.8	2.50
110	1265.8	47.4	3.61	10.4	3703.9	0.28
120	1228.3	100.9	7.59	3.1	1406.1	0.22
130	1201.7	241.3	16.72	0.7	499.3	0.14
140	1105.7	429.6	27.98	0.3	333.0	0.09

*:(cpm in β window/total cpm in α and β windows)x100, **: (cpm in α window/total cpm in α and β windows)x100

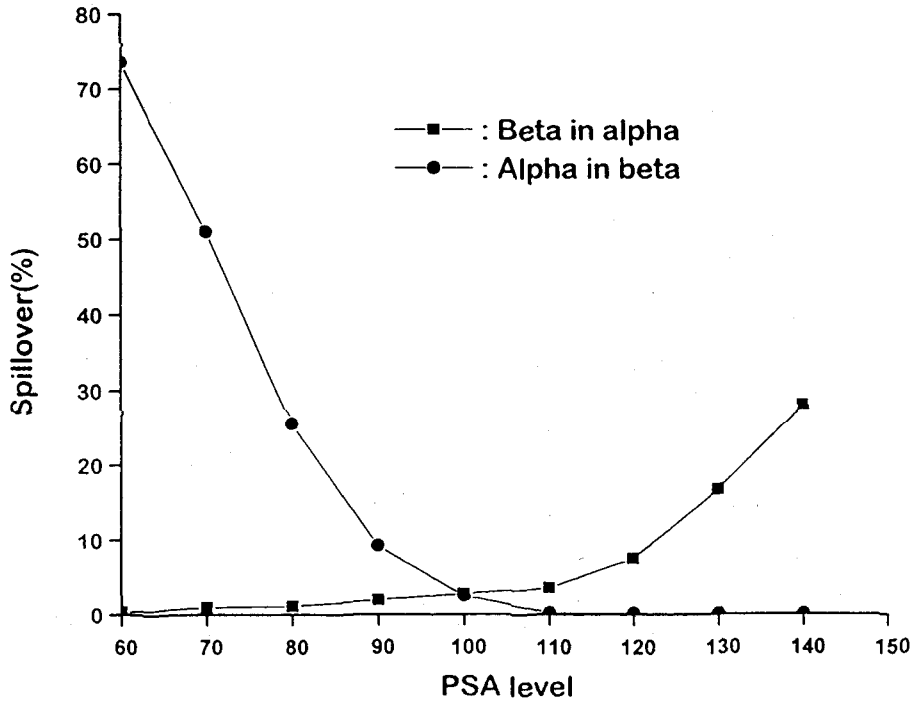

Fig. 1. Typical alpha/beta crossover plot for ^{239}Pu and ^{36}Cl standards in Ultimagold AB cocktail.