MEASUREMENT OF PLUTONIUM ISOTOPIC ACTIVITY RATIO IN LOW LEVEL PLUTONIUM SAMPLES

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INTRODUCTION

In the usual environmental plutonium monitoring around atomic energy plants, such as nuclear fuel reprocessing factories and Atmic power stations, plutonium is chemically extracted from environmental samples in question, purified radiochemically, and electrodeposited on a metallic plate, than α-ray spectrum of the electrodeposition sample is collected and α-activities of $^{239+240}Pu$ and ^{238}Pu of the environmental sample is determined. Alpha-ray energies of ^{239}Pu(5.16 MeV) and ^{238}Pu(5.17 MeV) are so close to each other that α-activities of the two nuclides can not be distinguished.

It is well known that nuclear test explosions and the burn-up of SNAP-9A have caused global plutonium contamination. Information about the plutonium isotopic ratio, $^{239}Pu/^{240}Pu$, gives possibility to presume whether plutonium found in environmental samples has originated from atomic energy plants or from global fallout. Measurements of the plutonium isotopic ratio in environmental samples using nuclear emulsion method[1], α-X ray method[2] and fission track method[3], have been attempted, but these methods were found to be insufficient in the sensitivity. Beside these methods, the Lγ/α-ray activity ratio has recently been used to measure the $^{240}Pu/^{239}Pu$ ratio in environmental samples[4].

PRINCIPLE OF THE METHOD

Plutonium extracted from an environmental sample in quesion and a standard plutonium, the isotopic ratio of which is well known, are both electrodeposited on a metallic plate, respectively, and α-ray spectra of these electrodeposition samples are collected to measure α-counting rates of $^{239+240}Pu$, $\alpha(^{239+240}Pu)_{sample}$ and $\alpha(^{239+240}Pu)_{stand.}$.

Then, these electrodeposition samples are altogether irradiated by thermal neutrons, and γ-ray counting rates of ^{99m}Tc, $\gamma(^{99m}Tc)_{sample}$ and $\gamma(^{99m}Tc)_{stand.}$, are measured.

The $^{239}Pu/^{239+240}Pu$ activity ratio in the environmental sample, $R(^{239}Pu/^{239+240}Pu)_{sample}$, is determined by the following equation.

$$R(^{239}Pu/^{239+240}Pu)_{sample} = \frac{\gamma(^{99m}Tc)_{sample}}{\alpha(^{239+240}Pu)_{sample}} \times \frac{\alpha(^{239+240}Pu)_{stand.}}{\gamma(^{99m}Tc)_{stand.}} \times R(^{239}Pu/^{239+240}Pu)_{stand.} \quad \cdots\cdots\cdots(1)$$

where $R(^{239}Pu/^{239+240}Pu)_{stand.}$ is $^{239}Pu/^{239+240}Pu$ activity ratio of the standard plutonium, and both $\gamma(^{99m}Tc)_{sample}$ and $\gamma(^{99m}Tc)_{stand.}$ are the counting rates converted to the values at the end of the neutron irradiation.

EXPERIMENTAL

Chemical procedures for plutonium extraction from environmental samples, radiochemical purification of the extract and electrodeposition on a metallic plate were carried out mainly according to the mannual issued from the Science and Technology Agency, Japan.[5)]

(I) NBS standard plutonium

Plutonium isotopic abundance of the NBS standard plutonium, which was used as the standard reference in the present study ($R(^{239}Pu/^{239+240}Pu)_{stand.} = 0.7384$).

(II) RCC standard plutonium

Another standard plutonium made by RCC was also electrodeposited on a nickel plate in the same manner as stated above. The contents of ^{238}Pu and ^{240}Pu of this standard plutonium are of negligibly low level.

(III) Environmental samples

Two environmental samples described below were investigated in the present study.

(a) Pit sample(Mud taken from a rain water pit in our laboratory. 1.6 kg. Less than 2 mm in diameter. (b) Sediment sample(Sea sediment, which was collected in the middle of the Nyu Bay, Fukui Prefecture. 240 g. Less than 0.5 mm in diameter.

It was found that, when an electrodeposition nickel plate was irradiated by neutrons, activities induced in the nickel plate made much interference to ^{99m}Tc γ-counting. In order to avoid this difficulty, deposited plutonium film on the nickel plate was wiped off with a piece of cellulose fiber filter paper immersed in hot nitric acid and the filter paper piece was neutron irradiated. Neutron irradiation was made in the Kyoto University Research Reactor(5MW, thermal neutron flux: X10^{13}n.cm^2.sec-1, 1 hour irradiation).

RESULT AND DISCUSSION

In Table 1 are shown measured values of $\alpha(^{239+240}Pu)$ and $\gamma(^{99m}Tc)$.

Sample	α ($^{239+240}Pu$) (cpm)	γ (^{99m}Tc) (cpm)	
		Neutron irradiation I	Neutron irradiation II
P - 1	0.47(2.56)	3.61(48)	
P - 2	1.39(1.09)	4.35(17)	
P - 3	2.13(0.54)	5.10(26)	
S - 1	0.56(1.77)		5.22(27.9)
S - 2	1.22(0.66)		6.19(17.0)
S - 3	2.34(0.75)		8.72(25.5)
RCC-1	17.30(0.69)	53.6(6.5)	
RCC-2	30.46(0.56)	107.0(8.3)	
RCC-3	44.91(0.95)	150.0(5.1)	
RCC-4	88.97(0.64)	281.0(2.6)	
NBS-1	16.69(0.72)	44.9(9.9)	44.8(11.9)
NBS-2	34.94(0.59)	85.5(12.4)	102.5(5.2)
NBS-3	70.46(0.66)	173.0(3.6)	214.5(4.7)
NBS-4	89.23(0.72)	220.0(4.8)	263.8(5.0)

( ): Relative standard deviation in %

Table 1 $^{239+240}Pu$ and ^{99m}Tc counting rate of the samples.

Wipe-off samples of P-1~3, RCC-1~4 and NBS-1~4 were neutron irradiated altogether (Neutron irradiation I in Table 1). About six months later, wipe-off samples of S-1~3 and NBS-1~4 were neutron irradiated altogether (Neutron irradiation I in Table 1). About six months later, wipe-off samples of S-1~3 and NBS -1~4 were neutron irradiated altogether, when activities in the NBS samples at Neutron irradiation I had decayed to be negligibly small (Neutron irradiation II).

Relation between $\alpha(^{239+240}Pu)$ and $\gamma(^{99m}Tc)$, which were found for the NBS samples and the RCC samples are shown in Fig. 1, and those which were found for the pit samples and the sediment samples, are shown in Fig. 2.

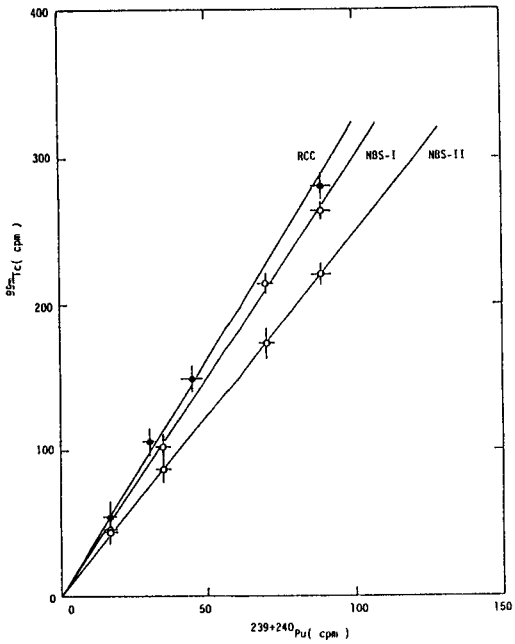

Fig. 1 Standard plutonium.

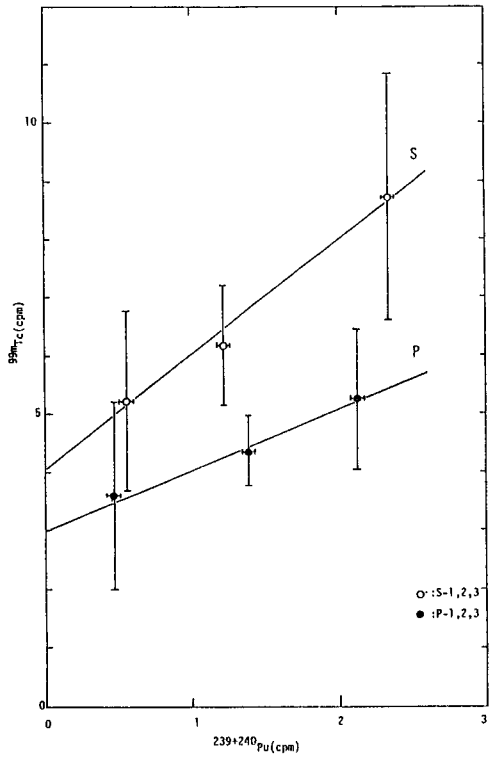

Fig. 2 Nyu Bay sediment and pit samples.

Here, it was assumed that the $\gamma(^{99m}Tc)$ values corresponding to zero $\alpha(^{239+240}Pu)$, namely, 4 and 3 cpm, for the sediment samples and the pit samples, respectively, are ascribed to uranium contamination.

In Table 2 are shown $^{239}Pu/^{239+240}Pu$ ratio values, which were calculated on the basis of the results shown in Table 1 and on the assumption stated above. Ratios of $^{238}Pu/^{239+240}Pu$ are also shown in the table.

Kray et al. reported that the $^{239}Pu/^{239+240}Pu$ ratio in the recent global fallout ranges from 0.5 to 0.8 by the use of mass spectrometer method.[6] Hisamatsu et al.[2], Nakanishi et al.[3] and Komura et al.[4] reported that the ratio in sediment and soil ranges from 0.4 to 0.6 by the use of α-X ray method and fission track method.

The ratio in the sediment sample determined in the present

study(~0.6)is in the range stated above. But, the ratio determined for the pit sample(~0.3) is somewhat smaller.

Sample	$^{239+240}Pu$ (Bq) $X10^{-2}$	^{238}Pu (Bq) $X10^{-2}$	$^{239}Pu/^{239+240}Pu$ (Bq/Bq)	$^{238}Pu/^{239+240}Pu$ (Bq/Bq)	^{137}Cs (Bq/kg)
P - 1	2.57 ± 0.07	0.17 ± 0.02	0.39 ± 0.18	0.066 ± 0.007	
P - 2	7.55 ± 0.08	0.70 ± 0.03	0.29 ± 0.05	0.093 ± 0.005	64.1 ± 3.3
P - 3	11.48 ± 0.07	1.00 ± 0.02	0.29 ± 0.08	0.087 ± 0.002	
S - 1	3.03 ± 0.05	0.07 ± 0.02	0.51 ± 0.14	0.023 ± 0.006	
S - 2	6.63 ± 0.05	0.13 ± 0.02	0.45 ± 0.08	0.020 ± 0.003	21.5 ± 3.3
S - 3	12.68 ± 0.10	0.33 ± 0.02	0.53 ± 0.15	0.026 ± 0.001	
RCC-1	93.72 ± 0.55	~ 0	0.91 ± 0.09	~ 0	
Rcc-2	164.98 ± 0.92	~ 0	1.03 ± 0.12	~ 0	------
RCC-3	274.97 ± 2.32	~ 0	0.98 ± 0.09	~ 0	
RCC-4	481.92 ± 3.08	~ 0	0.93 ± 0.08	~ 0	

Table 2 Activity ratios of $^{239}Pu/^{239+240}Pu$ and $^{239}Pu/^{239+240}Pu$ found in Nyu Bay sediment(S), pit sediment(P) and standard plutonium(PCC) samples.

The $^{238}Pu/^{239+240}Pu$ ratio found for the sediment sample(0.02 ~ 0.03) is relatively close to the ratio of 0.03 in global fallout[7]. Aginst this, the ratio found for the pit sample(~0.09) is considerably larger.

It appears that the main origin of the plutonium in the sediment sample is fallout from nuclear test explosions. Both of $^{239}Pu/^{239+240}Pu$ and $^{238}Pu/^{239+240}Pu$ ratios of the pit sample are different from those of global fallout. But, the main origin of the plutonium in the pit sample is also presumed to be nuclear debris. It is possible that the pit sample contain much of the old nuclear debris which were produced in Russian nuclear explosions carried out in 1961 to 1962. In our preceding study it was found that the $^{238}Pu/^{239+240}Pu$ ratios determined for fallout samples collected in 1961 to 1968 were rather high, ranging from 0.01 to 0.5[8].

It is desirable for raising accuracy of the present measurement method to get so much amount of electrodeposited plutonium that uranium contamination may be neglected. If the uranium contamination is as low as in the present study, at least 2 Bq of plutonium is necessary for getting sufficient accuracy.

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