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公開資料

Environmental Plutonium and Americium

—Knowledge and Analysis—

1996, 9

Tokai Works

POWER REACTOR AND NUCLEAR FUEL DEVEROPMENT CORPORATION

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Environmental Plutonium and Americium
— Knowledge and Analysis —

Masanao NAKANO※, Shuichi SUMIYA☆, Hitoshi WATANABE※
Hiromi KATAGIRI※, Yasuo AKATSU※
Kazunori TOBITA★

ABSTRACT

This report consists of two sections.

Section 1 shows the plutonium and americium's data of formation, property, atomic explosion test, spent fuel, effluent and environmental sample.

Section 2 explains analytical method for environmental samples. The outline of the analysis consists of 4 major procedures such as pretreatment, chemical separation, electrodeposition and alpha-ray spectrometry. Initially the principle of each procedure is explained. Secondly, practical analyzing procedures of plutonium-239,240 and americium-241 in the environmental samples are shown. Finally, calibration of Si-detector and measurement of radioactivity are presented.

※ Environmental Protection Section, Health and Safety Division, Tokai-works, PNC
☆ Safety Management Section, Safety Division, Head Office, PNC
★ Radiation Protection Section, Health and Safety Division, O-arai Engineering Center, PNC

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環境中のプルトニウムとアメリシウム
——知見と分析——

中野 政尚※ 住谷 秀一☆ 渡辺 均※
片桐 裕実※ 赤津 康夫※
飛田 和則★

要旨

この資料は、環境中プルトニウムとアメリシウムに関する知見とその分析法の二つの章から成る。

第１章ではプルトニウムとアメリシウムの生成、特徴、核実験、使用済み燃料、廃棄物、環境試料についてのデータを示す。

第２章では環境試料中の分析方法について説明している。分析の概略は大きく4つの工程、すなわち前処理、化学分離、電着、α線測定から成る。初めにそれぞれの工程の原理を説明している。次に実際の環境試料の分析手順を示す。最後にSi検出器の校正と放射能の測定について示す。

※東海事業所安全管理部環境安全課
☆本社安全部安全管理課
★大洗工学センター安全管理部放射線管理課

CONTENTS

Introduction

Most of plutonium(Pu) and americium(Am) isotopes are long-lived alpha emitting nuclides. With respect to the dose assessment and the accumulation to ecological system, it is important to investigate the behavior of the nuclides in the environment.

These nuclides in the environment of Japan are mainly originated from the atmospheric nuclear explosion tests. Due to the atmospheric nuclear explosion tests, Pu-239, 240 and 241 fell on the surface of the earth. From beta decay of Pu-241, Am-241 has been generated.

According to P.W.Krey et al.[1)], the activity ratio of Am-241/Pu-239,240 was 0.22 in 1974, and it is estimated to grow up to 0.42 in 2037.

The daughter nuclide of Am-241 is neptunium-237(Np-237). It is a long-life alpha emitting nuclide that would be a major concern with the geological disposal of nuclear waste. Therefore, it is important to understand the behavior of Am-241 as parent nuclide.

This is why the collection of knowledge relevant to the above nuclides and development of analytical method of Pu-239,240 and Am-241 have been performed.

1 Plutonium and Americium in the Environment

1.1 Formation and Nuclear Properties of Plutonium and Americium

In nuclear reactors Pu and Am are generated from Uranium(U) and/or Pu. Formation scheme of transuranic(TRU) nuclides in uranium reactor is shown in Figure 1[2].

Table 1 shows the oxidation-states of Pu and Am known at present. Usually, Pu would exist as Pu(Ⅲ), Pu(Ⅳ), Pu(Ⅴ), and Pu(Ⅵ) and Am would exist as Am(Ⅲ) in the normal condition on the surface of the earth[3].

Table 2[4] and Table 3[4] show the nuclear properties of Pu and Am isotopes, respectively.

Since Pu-239,240 and Am-241 are long half lived alpha emitting nuclides (2.4 $\times 10^4$years for Pu-239, 6.6$\times 10^3$years for Pu-240, and 432 years for Am-241) , these nuclides are important for the study of environmental radioactivity from the view point of dose assessment.

Alpha spectrometry method is generally applied to measure these nuclides in the environmental samples. The alpha energy of Pu-238 is 5.499MeV and that of Am-241 is 5.486MeV. Since these energies are too close to be discriminated by the usual alpha spectrometer, it is essential to separate chemically Pu from Am in the sample .

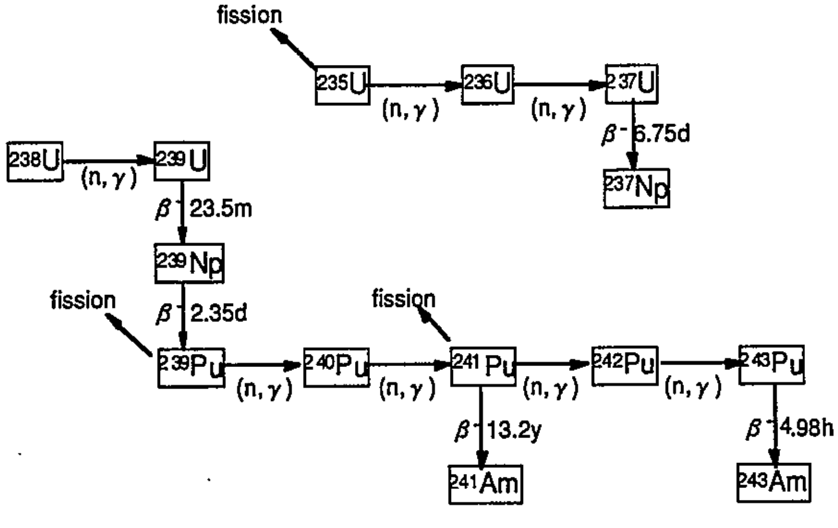


Figure 1 Main nuclear reactions in uranium fuel reactor

Table 1 oxidation states of Pu and Am

Element	Pu	Am
oxidation state		[2]
	(3)	(3)
	(4)	4
	(5)	5
	(6)	6
	7	7

( ):the common state
[ ]:only solid state

Table 2 Nuclear properties of Pu isotopes

Mass number	Half-life	Mode of decay	Main radiations (MeV)			Method of production
232	34min	EC>80%	α	6.60	62%	^{233}U(α,5n)
		α<20%		6.54	38%	
233	20.9min	EC 99.88%	α	6.30		^{233}U(α,4n)
		α 0.12%	γ	0.235		
234	8.8h	EC 94%	α	6.202	68%	^{233}U(α,3n)
		α 6%		6.151	32%	
235	25.6min	EC>99%	α	5.85		^{235}U(α,4n)
		α 3E-3%	γ	0.049		^{233}U(α,2n)
236	2.85yr	α	α	5.768	69%	^{235}U(α,3n)
	3.5E9yr	SF		5.721	31%	^{236}Np daughter
237	45.4d	EC>99%	α	5.65	21%	^{235}U(α,2n)
		α 3.3E-3%		5.36	79%	^{237}Np(d,2n)
			γ	0.059		
238	87.74yr	α	α	5.499	70.9%	^{242}Cm daughter
	4.8E10yr	SF		5.457	29.0%	^{238}Np daughter
239	2.41E4yr	α	α	5.155	73.3%	^{239}Np daughter
	5.5E15yr	SF		5.143	15.1%	n capture
			γ	0.129		
240	6.563E3yr	α	α	5.168	72.8%	multiple n capture
	1.34E11yr	SF		5.123	27.1%	
241	14.4yr	β^->99%	α	4.896	83.2%	multiple n capture
		α 2.41E-3%		4.853	21.1%	
			β^-	0.021		
			γ	0.149		
242	3.76E5yr	α	α	4.901	74%	multiple n capture
	6.8E10yr	SF		4.857	26%	
243	4.956h	β-	β^-	0.58		multiple n capture
			γ	0.084		
244	8.26E7yr	α	α	4.589	81%	multiple n capture
	6.6E10yr	SF		4.546	19%	
245	10.5h	β^-	β^-	1.28		^{244}Pu(n,γ)
			γ	0.327		
246	10.85d	β^-	β^-	0.374		^{245}Pu(n,γ)
			γ	0.224		

░ Nuclides concerned

▓ Nuclides used as chemical recovery tracer

Table 3 Nuclear properties of Am isotopes

Mass number	Half-life	Mode of decay	Main radiations (MeV)			Method of production
234	2.6min	EC				^{230}Th(^{10}B,6n)
						^{10}B,^{11}B on ^{233}U
237	1.22h	EC>99%	α	6.042		^{237}Np(α,4n)
		α 0.025%	γ	0.280	47%	^{237}Np(^{3}He,3n)
238	16.3h	EC>99%	α	5.94		^{237}Np(α,3n)
		α 1.0E-4%	γ	0.963	29%	
239	11.9h	EC>99%	α	5.776	84%	^{237}Np(α,2n)
				5.734	13.8%	
		α 0.010%	γ	0.278	15%	^{239}Pu(d,2n)
240	50.8h	EC>99%	α	5.378	87%	^{237}Np(α,n)
		α 1.9E-4%		5.337	12.0%	^{239}Pu(d,n)
			γ	0.988	73%	
241	432.7yr	α	α	5.486	84.0%	^{241}Pu daughter
	1.15E14yr	SF		5.443	13.1%	multiple n capture
			γ	0.059	35.7%	
242	16.01h	β^- 82.7%	β^-	0.667		^{241}Am(n,γ)
		EC17.3%	γ	0.042	weak	
242m	141yr	IT99.55%	α	5.207	89.0%	^{241}Am(n,γ)
	9.5E11yr	SF α 0.45%		5.141	6.0%	^{241}Am(n,γ)
			γ	0.049	41.0%	
243	7.38E3yr	α	α	5.277	88.0%	multiple
	2.0E14yr	SF		5.234	10.6%	n capture
			γ	0.075	68%	
244	10.1h	β^-	β^-	0.387		^{243}Am(n,γ)
			γ	0.746	67%	
244m	26min	β^->99%	β^-	1.50		^{243}Am(n,γ)
		EC0.041%				
245	2.05h	β^-	β^-	0.895		^{245}Pu daughter
			γ	0.253	6.1%	
246[a]	25.0min	β^-	β^-	2.38		^{246}Pu daughter
			γ	0.799	25%	
246[a]	39min	β^-	γ	0.679	52%	^{244}Pu(α,d)
						^{244}Pu(^{3}He,d)
247	24min	β^-	γ	0.285	23%	^{244}Pu(α,p)

[a] Not known whether ground-state nuclide or isomer.

▨ Nuclides concerned

■ Nuclides used as chemical recovery tracer

1.2 Plutonium and Americium from Atomic Explosion Tests

Estimated production amounts of TRU isotopes by atmospheric nuclear tests are shown in Table 4[5]. Pu-241 was generated much more than the other isotopes, but from the view point of radiological significance, Pu-239 is the most important nuclide.

Table 5[5] shows the integrated deposition density of Pu-238, Pu-239,240, Pu-241 and Am-241. The amount of fallout on the northern hemisphere is larger than that on the southern since atomic explosion tests had been done more in the northern hemisphere.

Table 4 Production of plutonium and transplutonium isotopes by atmospheric nuclear tests

(UNSCEAR 1982)

Isotope	Half-life(a)	Mass ratio relative to Pu-239 corresponding to production by nuclear tests	Generated amount (PBq)
Pu-238	87.7	0.00016	0.33
Pu-239	24100	1	7.8
Pu-240	6570	0.18	5.2
Pu-241	14.4	0.013	170
Pu-242	376000	0.0034	0.016
Am-242m	152	0.00000031	0.00037
Cm-244	18.1	0.000000025	0.00026

Table 5 Integrated deposition density of Pu-238, Pu239,240, Pu-241 and Am-241a)
(UNSCEAR 1982)

Location	Integrated deposition density(Bq/m^2)			
	Pu-238	Pu-239,240	Pu-241b)	Am-241c)
World	0.90	35	440	15
Northern Hemisphere	0.98	39	480	17
Southern Hemisphere	0.25	9.7	120	4.2
North temperate zone (40N-50N)	1.5	58	730	25
South temperate zone (40S-50S)	0.41	16	200	7.0

a) Through 1979 from nuclear explosions only. A satellite reentry in 1964 in the southern hemisphere caused additional widespread deposition of Pu-238.

b) Taking into account a delay of 10 months between production and deposition.

c) From Am-241 deposition plus Pu-241 decay.

Estimation of Pu-239 and Am-241 concentration in soil

According to Sh. SUMIYA et al.6), 80% of fallout nuclides existed in the surface layer of soil between 0-5cm in depth, and the density of dried soil is about 1.1g/cm^3 .

The volume of the soil with fallout nuclides is estimated 50000 cm^3/m^2.

The weight of the soil is calculated by multiplying the volume of the soil by the soil density. It becomes 55 kg dry/m^2

In the 55kg surface dry soil, there are 58 × 0.8 (Bq/m^2) for Pu-239,240 and 25 × 0.8 (Bq/m^2) for Am-241 at 40N-50N. Thus, the concentration of the nuclides in the surface soil at 40N-50N are,

Pu-239,240	$58 \times 0.8 \div 55 = 0.84$(Bq/kg dry)
Am-241	$25 \times 0.8 \div 55 = 0.36$(Bq/kg dry)

1.3 Plutonium and Americium in Spent Fuel and in Effluent

As shown in Figure 1, Pu and Am are generated from uranium via neutron capture. Table 6 shows the activities of the isotopes of Th, Pa, U, Np, Pu and Am under the calculation by ORIGEN 2. The calculation condition is that the PWR power is 38MW, that the burn-up is 38000MWd/tU, that the neutron flux is 2.93 $\times 10^{14}$ n/cm^2-sec and that cooling time is 10, 100, 1000 and 10000 years.

Even after 1000 years, Pu-239,240 and Am-241 will have very high activity .

Table 7[7)] shows the radionuclides released in effluents from fuel reprocessing plant in the world. According to the operational experience, Tokai Reprocessing Plant released relatively small amount of radionuclide compared with reprocessing plant in UK and France.

Table 6 Long-lived actinides in spent fuel

Year	Activity(GBq/tU) after				Year	Activity(GBq/tU) after			
	10	100	1000	10000		10	100	1000	10000
Th-229	1.70E-06	3.57E-05	5.27E-03	6.50E-01	Np-237	1.54E+01	1.90E+01	3.97E+01	4.60E+01
Th-230	1.81E-04	1.18E-02	3.27E-01	3.52E+00	Pu-236	1.95E+00	1.79E-05	1.78E-05	1.69E-05
Th-232	6.83E-09	5.99E-08	6.03E-07	6.92E-06	Pu-238	1.06E+05	5.22E+04	5.23E+01	1.52E-17
Pa-231	1.94E-04	1.59E-03	1.55E-02	1.51E-01	Pu-239	1.25E+04	1.25E+04	1.22E+04	9.47E+03
U-232	8.94E-01	4.10E-01	8.84E-05	1.69E-05	Pu-240	1.95E+04	1.95E+04	1.77E+04	6.82E+03
U-233	7.76E-04	7.50E-03	1.32E-01	1.89E+00	Pu-241	6.73E+01	8.84E-01	1.36E-04	6.52E-05
U-234	3.52E+00	2.29E+01	4.17E+01	4.10E+01	Pu-242	6.27E+01	6.27E+01	6.26E+01	6.16E+01
U-235	7.33E-01	7.34E-01	7.45E-01	8.41E-01	Am-241	6.03E+04	1.33E+05	3.17E+04	2.68E+00
U-236	1.19E+01	1.20E+01	1.25E+01	1.55E+01	Am-242m	2.35E+00	1.56E+00	2.58E-02	3.87E-20
U-238	1.17E+01	1.17E+01	1.17E+01	1.17E+01	Am-243	6.54E+02	6.48E+02	5.96E+02	2.56E+02

Table 7 Radionuclides released in effluents from fuel reprocessingplants,1985-1989 (UNSCEAR 1993)

Table 46
Radionuclides released in effluents from fuel reprocessing plants, 1985-1989
[B13, C17, N7, N13, S3, S9]

Reprocessing plant	*Year*	*Electrical energy generated (GW)*	*Release (TBq)*											
			Airborne effluents						*Liquid effluents*					
			^{3}H	^{14}C	^{85}Kr	^{129}I	^{131}I	^{137}Cs	^{3}H	^{14}C	^{90}Sr	^{106}Ru	^{129}I	^{137}Cs
France Cap de La Hague	1985	6.11	32		70300				2600	0.7	76	440	0.13	29
	1986	2.52	6		29000				2300	0.7	78	470	0.13	29
	1987	3.04	15		35000				3000		65			7.6
	1988	2.35	21		27000				2500		48			8.5
	1989	3.65	25		42000				3700		41			13
United Kingdom Sellafield	1985	1.70	268	7.0	23800	0.007	0.002	0.002	1062	1.3	52	81	< 0.1	325
	1986	3.81	171	5.4	53300	0.03	0.003	0.007	2150	2.6	18.3	28	0.12	17.9
	1987	2.43	78.3	9.5	34000	0.019	0.004	0.004	1375	2.1	15.0	22.1	0.1	11.8
	1988	2.84	185.6	3.6	39800	0.024	0.002	0.005	1724	3.0	10.1	23.6	0.13	13.3
	1989	3.69	677	3.9	51700	0.024	0.002	0.004	2144	2.0	9.2	25.0	0.17	28.6
Japan Tokai-Mura	1985	1.2	2.8		10000	0.0010	*		260		0.000002	*	*	0.00008
	1986	1.2	2.7		13000	0.0023	*		240		0.000025	*	*	0.00017
	1987	0.93	3.7		12000	0.00014	*		260		0.000009	*	*	0.00015
	1988	0.17	2.5		2700	0.00009	*		74		*	*	*	0.00009
	1989	1.1	3.7		9800	0.00024	*		240		*	*	*	0.00004
Total release (TBq)			1494	29.4	453400	0.108	0.013	0.022	23630	12.4	412.6	1090	0.880	483.7
Electrical energy generated (GW a)			36.74	14.47	36.74	19.07	19.07	14.47	36.74	23.1	36.74	27.70	27.70	36.74
Normalized release [TBq (GW a)$^{-1}$]			41	2.0	12300	0.0057	0.0007	0.0015	643	0.54	11	39	0.032	13

[*] Less than detection limit.

1.4 Plutonium and Americium in Environmental Samples

The concentrations of Pu and Am in Environmental samples, such as surface soil, seabed sediment and marine products, are shown in Table 8[8),9)], Table 9[10)] and Table 10[11)]. The concentrations of Pu and Am in Japanese environmental samples are much smaller than those in Sellafield. Figure 2[9)] and Figure 3[9)] show concentrations of Pu-239,240 and Am-241 in sea water around Tokai-mura. Decrease tendency for Pu-239,240 is found .

Table 8 Concentrations of each kind of samples for Pu-239,240 and Am-241 offshore Tokai

Nuclide	Sample	Unit	Concentration
Pu-239,240	Surface Soil	Bq/kg dry	0.21～0.7*
	Seabed Sediment	Bq/kg dry	0.054～1.5**
	Flat Fish	Bq/kg fresh	4.6×10^{-5}～4.2×10^{-4}*
	Seaweed	Bq/kg fresh	7.8×10^{-5}～1.1×10^{-2}*
	Sea water	Bq/l	4.3×10^{-6}～2.7×10^{-5}*
Am-241	Surface Soil	Bq/kg dry	0.058～0.26*
	Seabed Sediment	Bq/kg dry	0.043～0.67**
	Flat Fish	Bq/kg fresh	2.6×10^{-5}～3.0×10^{-4}*
	Seaweed	Bq/kg fresh	1.0×10^{-4}～5.2×10^{-3}*
	Sea water	Bq/l	5.0×10^{-7}～9.9×10^{-6}*

* PNC Technical review No.81[8)]
**Sumiya et.al[9)]

Table 9 Concentrations of Pu-239,240 and Am-241 in the coastal and estuarine sediments of the Irish Sea (Bq/kg±SD) (C.K.Kim et al.,1992)

Site	Pu-239,240	Am-241	Site	Pu-239,240	Am-241
1	28.0±2.5	29.4±1.2	13	37.8±1.8	53.2±3.9
2	58.4±3.3	84.1±5.2	14	163±7	249±15
3	2.3±0.3	2.61±0.32	15	657±27	1077±43
4	111±4	141±0	16	1589±50	1796±52
5	92.1±4.2	156±10	17	1396±38	1894±53
6	60.0±2.3	91.2±5.7	18	999±23	1229±46
7	193±7	266±10	19	55.0±3.0	69.9±5.1
8	119±5	159±8	20	14.6±1.1	14.5±1.1
9	240±9	334±18	21	85.1±3.7	118±8
10	64.0±3.0	101±6	22	71.4±3.0	101±7
11	33.2±1.8	41.4±3.1	23	141±6	227±10
12	81.0±3.4	114±7	24	102±4	163±9

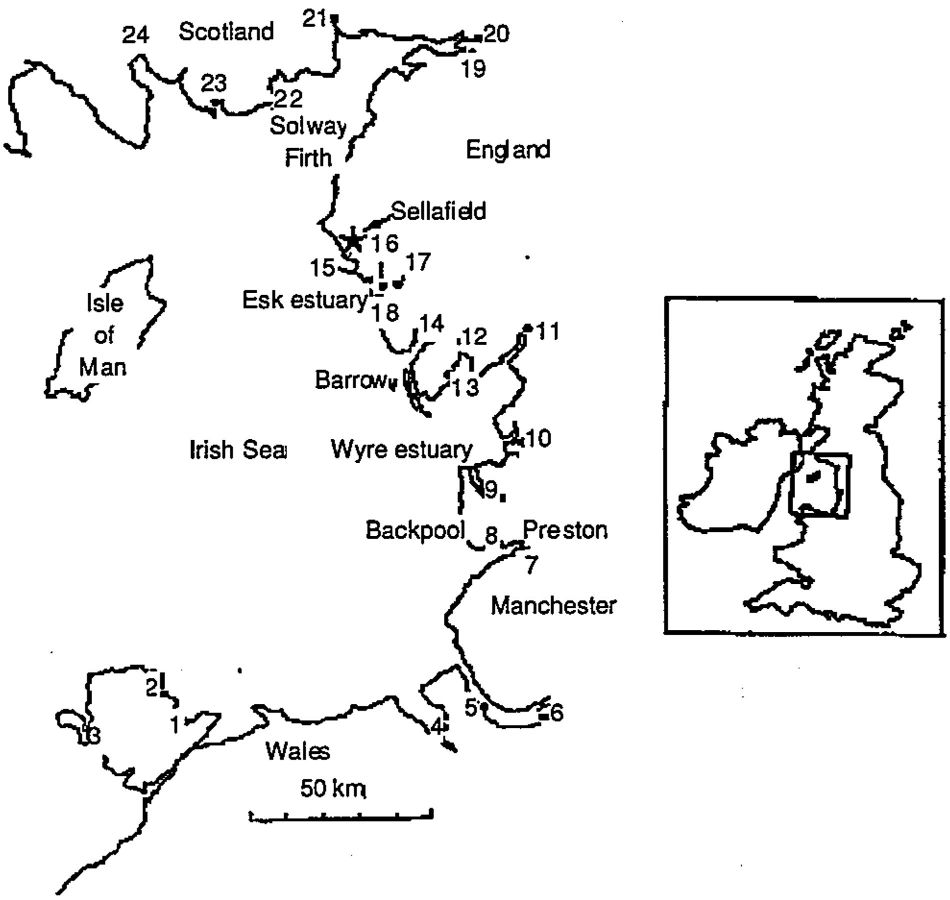


Figure Location map of the Northern Irish Sea, with sites of intertidal samples

Table 10 Transuranic activities in fish and shellfish in the Irish Sea and North Sea ,1983 (UNSCEAR 1988)

Sampling area/ landing points	sample	number of sampling observations	Mean activity concentration (wet), Bq kg^{-1}					
			^{238}Pu	^{239,240}Pu	^{241}Pu	^{241}Am	^{242}Cm	^{243,244}Cm
Sellafield shoreline area	Cod	1	0.0047	0.025		0.020	0.00044	0.00011
	Crabs	3	0.71	2.9	80	7.3	0.097	0.051
	Lobsters	3	0.54	2.2	63	14	0.059	0.062
	Winkles	2	6.6	27	710	37	0.45	0.17
Sellafield offshore area	Plaice	1	0.0085	0.034		0.038	n/d	n/d
	Cod	1	0.0057	0.026		0.030	0.00087	n/d
	Skate	1	0.011	0.044		0.045	0.00054	0.00027
	Whelks	1	1.7	7.3		15	n/d	n/d
Ravenglass a/	Cockles	1	14	54		75	1.5	0.47
	Mussels	2	9.9	41	1000	55	0.45	0.25
	Plaice	1	0.011	0.043		0.048	0.0012	0.00014
	Cod	1	0.0040	0.016		0.015	0.00088	0.00026
Northern North Sea	Cod	1	0.00067	0.0038		0.0051	n/d	0.00002
	Nephops	1	0.0019	0.0092		0.0074	0.00027	0.00008
Mid-North Sea	Nephrops	1	0.00075	0.0033		0.0025	n/d	n/d
	Mussels	1	0.0035	0.019		0.0045	n/d	n/d
Southern North Sea	Mussels	1	0.00077	0.0042		0.0013	n/d	n/d
	Cockles	1	0.0023	0.013		0.0054	n/d	n/d
Iceland area	Cod	1	0.000063	0.00027		0.00032	n/d	n/d

a/ Landing point.
n/d = Not detected.

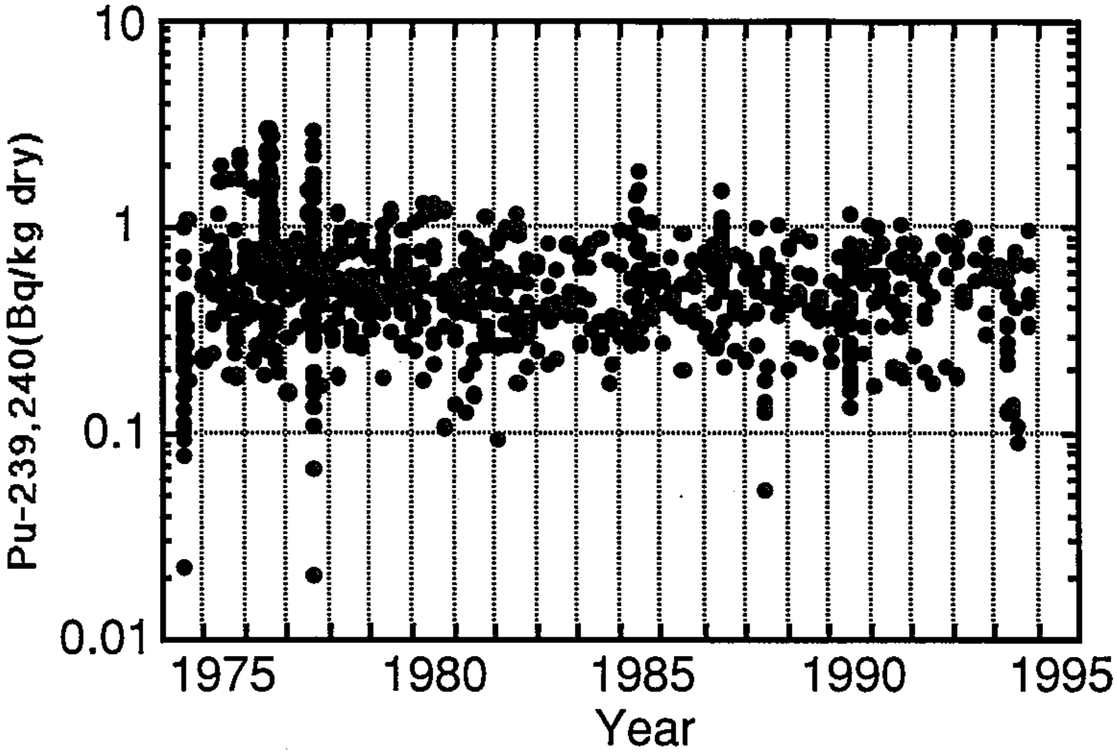


Figure 2 Concentration of Pu-239,240 in seabed sediment in Japan

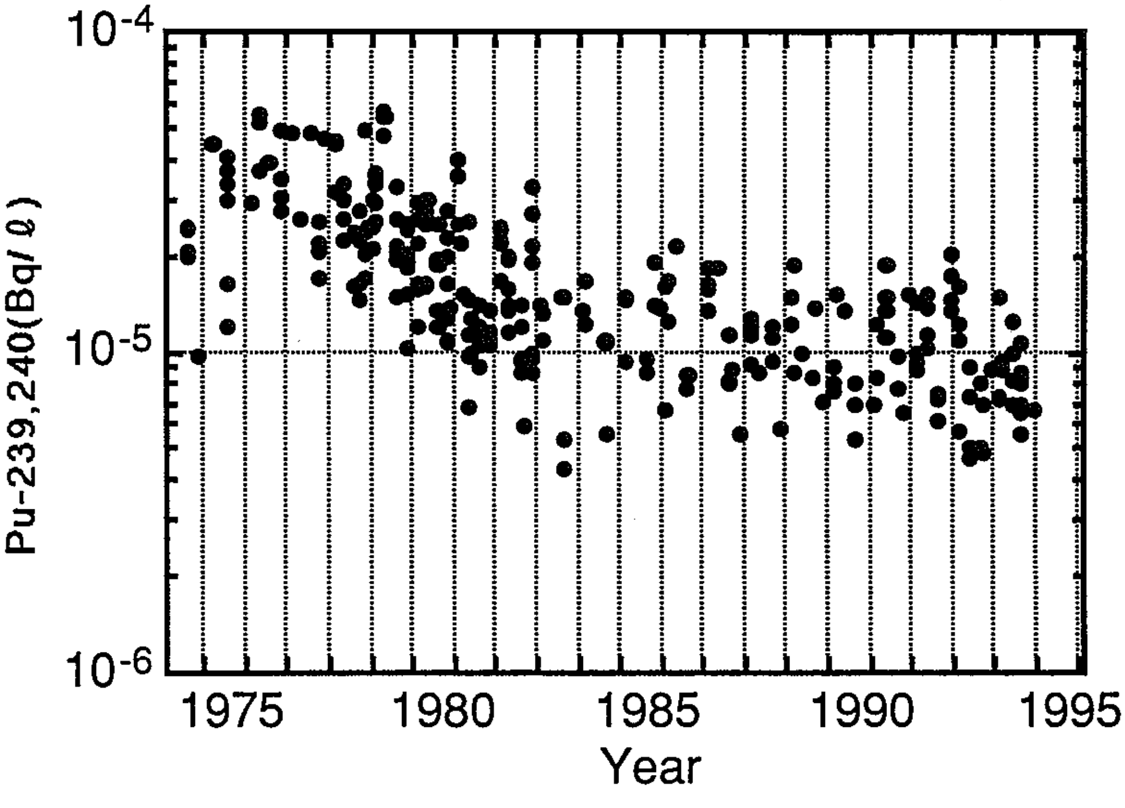


Figure 3 Concentration of Pu-239,240 in sea water in Japan

2. Analytical Method for Environmental Samples

2.1 Outline

The analyses of Pu and Am in the environmental samples contain four major procedures as represented in Figure 4. The principle of each procedure is explained from section 2.1.1 to 2.1.4.

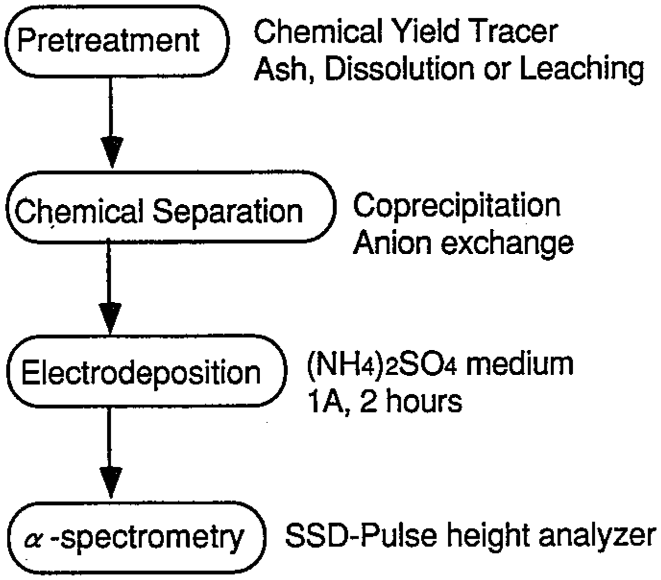


Figure 4 Block diagram of analytical procedures for Pu and Am in environmental samples

2.1.1 Pretreatment

(1) Extraction Rate for Plutonium by Acidic Leaching Methods

It is certain that all soil materials are decomposed by using HNO_3-HF, but it takes long time. Thus, simpler leaching methods using HNO_3 and HNO_3-H_2O_2 were compared. Extraction rates of both methods are shown in Table 11[12). The residual amounts for Pu in each residual soil were determined by decomposition method by HNO_3-HF. There is no big difference between them, and over 90% of Pu was extracted. You have to notice that this sample must contain only fallout Pu or waste liquid Pu. If it contains oxide nuclear fuel burned at high temperature, we have to dissolve it completely by using HNO_3-HF. The extraction amount for Pu by twice HNO_3 leaching is shown in Table 12[12). The extraction amount of the second extraction was about 3% of the first extraction.

Therefore, one-time HNO_3 leaching method was adopted to extract Pu from soil samples.

Table 11 Comparison of extraction rate for Pu by two types of leaching methods (STA, 1989)

	Leaching by HNO_3			Leaching by HNO_3+H_2O_2		
	Extracted Amount (Bq/kgdry)	Residual Amount (Bq/kgdry)	Extract Rate (%)	Extracted Amount (Bq/kgdry)	Residual Amount (Bq/kgdry)	Extract Rate (%)
1	6.2±0.5	0.67±0.09	91	7.4±0.6	0.52±0.07	94
2	8.5±0.7	0.67±0.09	93	6.7±0.6	0.52±0.08	93
3	7.0±0.6	0.70±0.08	91	7.4±0.6	0.52±0.08	94
4	7.4±0.6	0.52±0.08	94	7.4±0.6	0.48±0.07	94
5	7.8±0.6	0.56±0.07	93	6.7±0.5	0.67±0.07	91

Table 12 Extraction amount for Pu by twice HNO_3 leaching (STA, 1989)

	The First Extraction (Bq/kgdry)	The Second Extraction (Bq/kgdry)
1	7.4 ± 0.4	0.24 ±0.03
2	7.0 ± 0.4	0.14 ±0.02
3	7.8 ± 0.5	0.14 ±0.02
4	7.4 ± 0.4	0.11 ±0.02
5	6.3 ± 0.3	0.16 ±0.04

(2) Relationship between Heat Temperature of Soil Sample and Extraction Rate for Plutonium

Soil and seabed sediment contain much organic matter. When soil samples are leached by acid without decomposition of organic matter by heating, the organic matter may interfere for Pu analysis. So, it had better decomposite organic matter by electric furnace in advance. Table 13[12)] shows the chemical recovery of each temperature of electric furnace. No difference on concentration of Pu and on recovery rate in 500℃ and 700 ℃ is seen.

Therefore, heating treatment of 550 ℃ 4 hours was adopted in PNC Tokai Works.

Table 13 Chemical recoveries for Pu on heating treatment (STA, 1989)

Heat Condition	Pu-239,240(Bq/kgdry)	Recovery Rate(%)
Not heated	6.8±0.5	56
500℃-4hours	6.5±0.4	85
	7.2±0.4	85
	7.1±0.4	89
	6.6±0.4	83
	7.1±0.4	84
700℃-2hours	7.0±0.4	79
1000℃-2hours	1.1±0.1	16

(3) Chemical Yield Tracer

For the analysis of Pu-239,240, the solution of Pu-236 or Pu-242 is used as chemical yield tracer. They have some merits and defects, respectively. As the half life of Pu-236 is 2.85years, it is necessary to use this tracer early after purified. Since tracer solution of Pu-236 could contain a little Pu-238, it is essential to grasp the amount of Pu-238 in Pu-236 solution. The defect of Pu-242 as tracer is its low alpha energy. If the peak of Pu-239,240 in the sample is very big and wide, it is feared that the peak of Pu-239,240 covers over that of Pu-242 tracer.

For Am-241 analysis, curium-244(Cm-244) or Am-243 is used as chemical yield tracer. They are very good tracers for the analysis of Am-241 in environmental samples. However if analyzing sample contains Cm-244, Cm-244 cannot be used for the analysis of Am-241.

Though Am-243 is also used as tracer for the analysis of Am-241, Am-243 might contain a little Am-241. It is essential to grasp the amount of Am-241 in Am-243 solution. The alpha energies of Am-243 are lower than those of Am-241 and relatively close to them. If the peak of Am-241 in the sample is very big and wide, it is feared that the peak of Am-241 covers over that of Am-243 tracer like Pu-242 tracer.

These chemical yield tracers have some defects as mentioned above. It is necessary for selection of chemical yield tracers to consider the characteristics of these tracers and the concentration of concerned nuclides and interfering nuclides in analyzing samples for selection of chemical yield tracer.

2.1.2 Chemical Separation

There are several chemical separation methods or techniques in analyzing Pu or Am. The most representative one is anion exchange extraction. Figure 5[13)] and Figure 6[13)] represent the ion exchange processes for Pu and Am, respectively. Figure 7 and Figure 8 show distribution coefficient curves for different acidities of HNO_3 and HCl on anion exchange resin.

Figure 9 shows the sequential elution curves by the anion exchange methods with Figure 5[14)]. The anion exchange resin does not adsorb Cm^{3+} and weakly absorbs UO_2^{2+} in 8N HNO_3 solution. It does adsorb Th^{4+} and Pu^{4+} in this solution. As 8N HCl goes through the resin, Th^{4+} is eluted, but Pu is not. As 0.1N HI-8N HCl goes through the resin, Pu^{4+} is reducted to Pu^{3+} gradually, then eluted.

Table 14[15)] shows distribution coefficients of elements in mineral acid - methyl alcohol(MeOH) mixed solution. Usually, Am is never adsorbed with anion exchange resin. However Am is adsorbed with the resin in the mixed solution of acid and alcohol. All of Am, Cm and Rare Earth Element(REE) in HNO_3-MeOH solution are adsorbed with the resin. Then HCl-NH_4SCN-MeOH goes through the resin, only REE is eluted. Finally, HCl-MeOH goes through the resin, Am and Cm are eluted. Figure 10[14)] shows these sequential elution curves.

Coprecipitation is a very important chemical procedure, too. Figure 11[14)] shows the pH dependence of metal oxalate precipitate and coprecipitation of Am on $Ca(COO)_2$. When the pH of the solution is over 1.5, Am is precipitated with $Ca(COO)_2$ efficiently. The suitable pH for actinides and lanthanides analyses is selected about at 1.8 to separate other major ions such as Al, Fe, Ti, and phosphates.

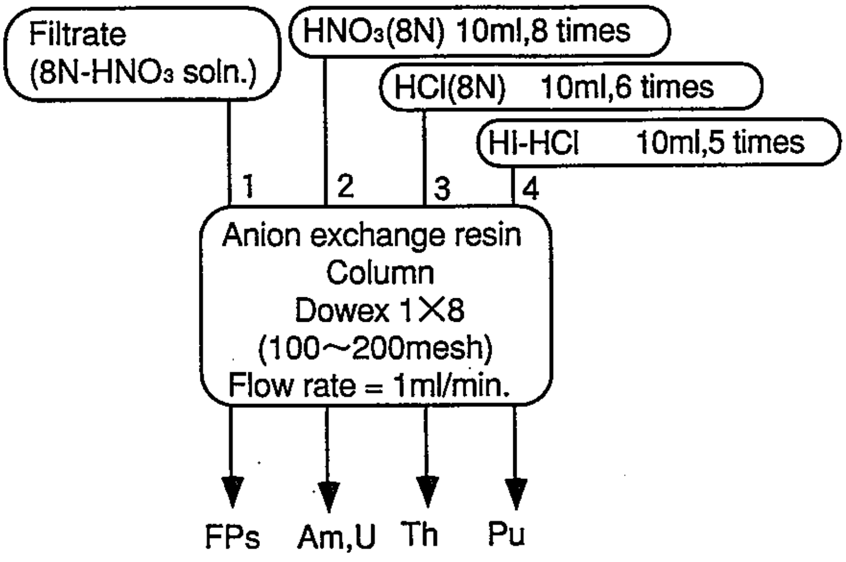


Figure 5 Ion exchange processes for Pu

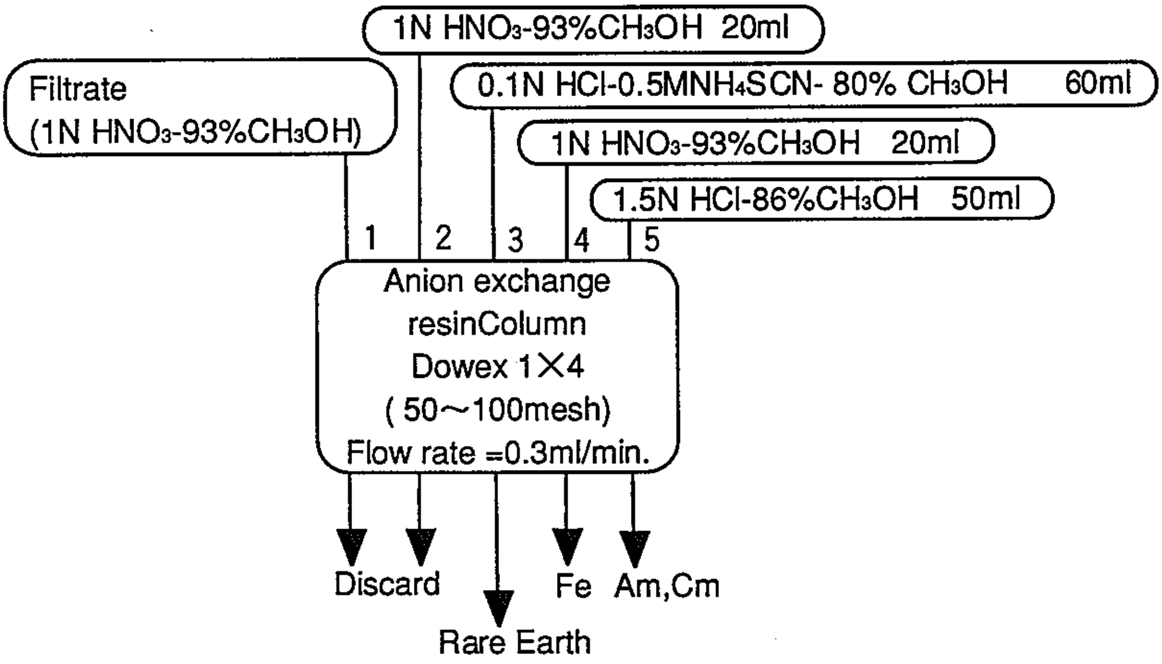


Figure 6 Ion exchange processes for Am

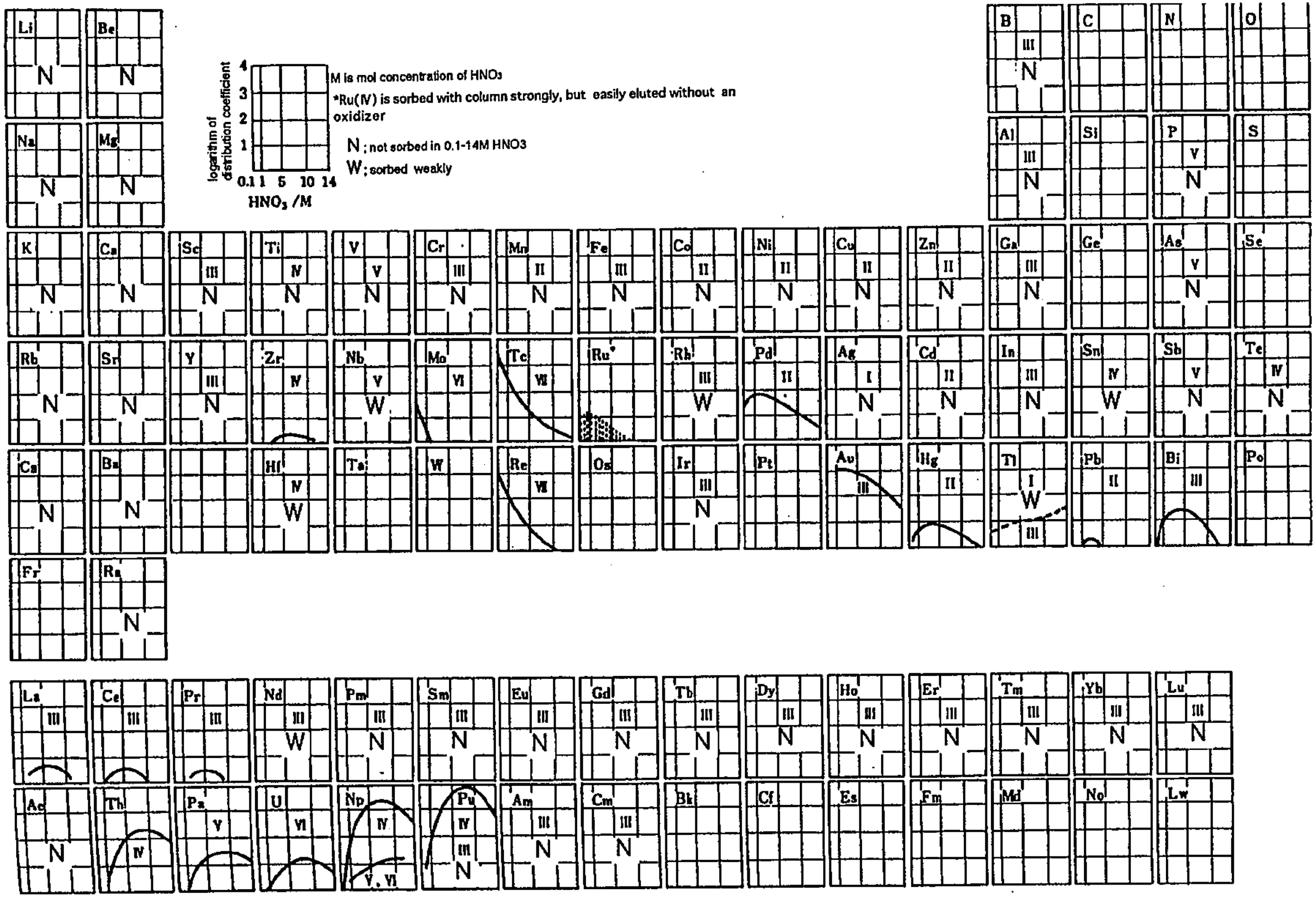


Figure 7 Distribution coefficients of the elements from HNO_3 acid on anion exchange resin

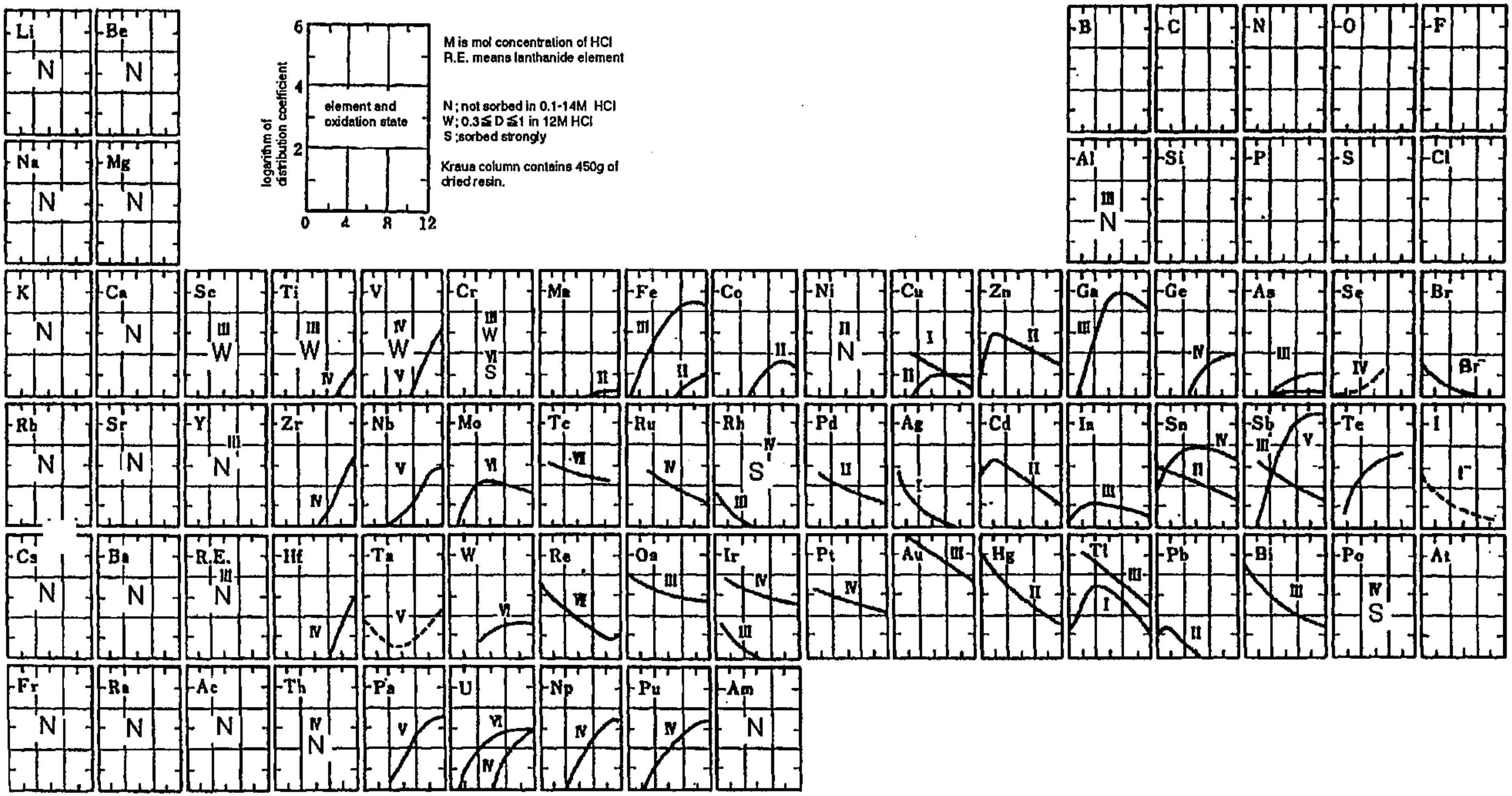


Figure 8 Distribution coefficients of the elements from HCl acid on anion exchange resin

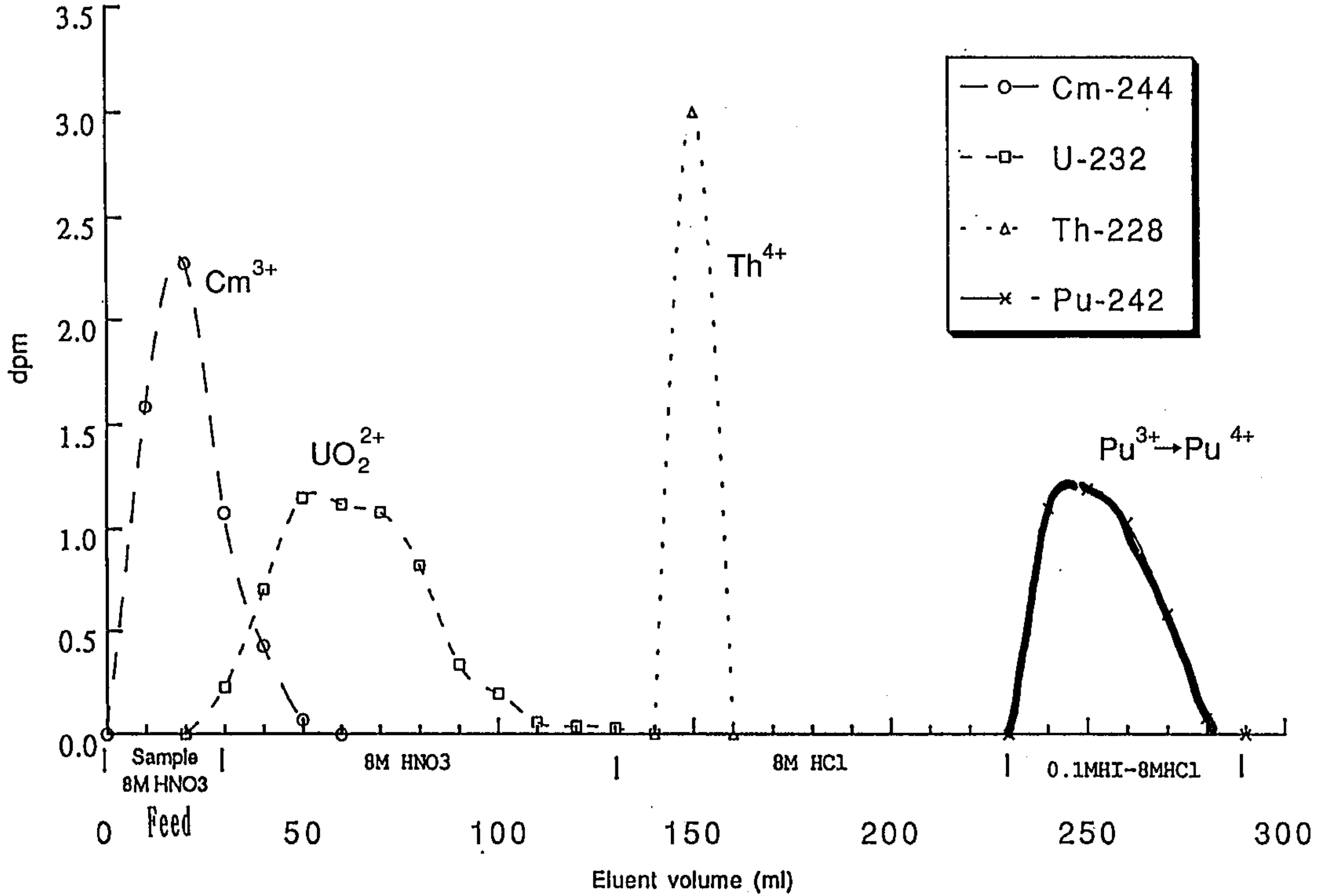


Figure 9 Sequential elution curves by anion exchange(Dowex 1×8)

Table 14 Distribution coefficients of actinides, lanthanides and metals on Dowex 1 anionic resin in different liquid mixtures of acid and alcohol

Dowex 1	Elements absorbed	Elements separated
HNO_3 MeOH	REE: Ce(III), 700^{+}, 800, $>1000^{*}$, $8000^{(*)}$ $La>10^{5(*)}$, Pr $4000^{(*)}$, Nd $2000^{(*)}$ Sn $305^{(*)}$, Eu 220, $150^{(*)}$ Lu, Yb, Tm; $25\text{-}30^{(*)}$ Er, Ho, Dy; $30\text{-}40^{(*)}$ Tb $55^{(*)}$, Gd $80^{(*)}$ ACT: Pu(IV)>4000, Th(IV) 12500^{*} Pb(II) 81, Bi(III) 3100, Po TPE: Cf(III) $218^{\times}$, Bk 40^{+}, 345, $19^{\times\times}$ Am 700^{**}, 1200^{++}, $1800^{\times}$, $56^{\times\times}$ Cm200^{**}, 450^{++}, $700^{\times}$, $27^{\times\times}$	Kd<1; Co(II), Ni(II), Mn(II), Cr(III), Mo(VI), V(V), Ti(IV), Al(III), Ga(III), In(III), Mg(II), Ca(II), Zn(II), Cd(II), Cu(II), Ag(I)$^{\circ}$, Zn$^{\circ}$, Y$^{\circ}$, Hf$^{\circ}$, Li$^{\circ}$, Na$^{\circ}$, K$^{\circ}$, Kd≧1; Cs 1°, 3, $3^{\times}$ Sr(II) 1, 33^{*} Zr(IV)<1, 1^{*}, $5.8^{\times}$ Fe(III)<1,2.5(*),$5.8^{\times}$ $UO_2 < 1^{\circ}$, 33^{*} Ru(?) $2.5^{\times}$
	(Note) S.S.Ahluwalia, J.Korkisch, Anal. Chim.Acta. 31, 552-556 (1964); 0.25N-95% $^{(*)}$ J.P.Feris, J.W.Warton, Anal.Chem.34, 1044-1080 (1962); 1N-90% $^{\circ}$ expressed as "not retained on resin" & * ; J.Korkisch, F.Tera, Anal. Chem.33, 1264-1266 (1961) 0.5N-90% ** V.A.Bochkarev; E.N,Voevodin, Radiokhimiya 7, 461-465 (1965); 1N-90% $^{+}$ L.I.Guseva, S.I. Grigoreva, Radiokhimiya 13, 778-780 (1971); 1N-80% $^{++}$ L.I.Guseva, G.S.Tikhomirova, S.I. Grigoreva, Radiokhimiya 15, 397-401 (1973); 1N-90% $^{\times}$ L.I.Guseva, G.S.Tikhomirova, Radiokhimiya 15, 401-404 (1973); 1N-90% $^{\times\times}$ I.A.Levedev, B.F.Myasoedov, L.I.Guseva, J.Radioanal.Chem.21, 259-266 (1974); 0.5N-80%	
HCl- NH_4SCN- MeOH	Am>Cm Cm $340^{\triangle}$, $31^{\triangle\triangle}$, >120	Ce<10 Eu $16^{\triangle}$, <12, $44^{\triangle\triangle}$
	(Note) I.A.Levedev, B.F.Myasoedov, L.I.Guseva, J.Radioanal.Chem.21, 259-266 (1974); 0.1N HCl- 0.5M NH_4SCN- 80% MeOH $^{\triangle}$ L.I.Guseva, G.S.Tikhomirova, Radiokhimiya 16, 152-156 (1974); 0.5M NH_4SCN- 80% MeOH $^{\triangle\triangle}$ ibid.; 0.1N HCl- 0.2M NH_4SCN- 80% MeOH	
HCl- MeOH	Zn(II) 400^{*} U $>10^{3+}$ Bi $>10^{5+}$ Pb Po	Th, 4.5^{+}, 71^{**} Am Cm
	* Y.Yoshino, Y.Kurimura, Bull.Chem.Soc.Japan.30, 563-566 (1959); 9.5N HCl- 80% MeOH ** D.H.Wilkins, G.E.Smith, Talanta 8, 132-138 (1961); 1.5N HCl- 86% MeOH + J.Korkisch, F.Tera, J.Chromitog, 6, 530-534 (1961), Z.Analyt.Chem.186, 290-295 (1962); 0.9N HCl- 85% MeOH	

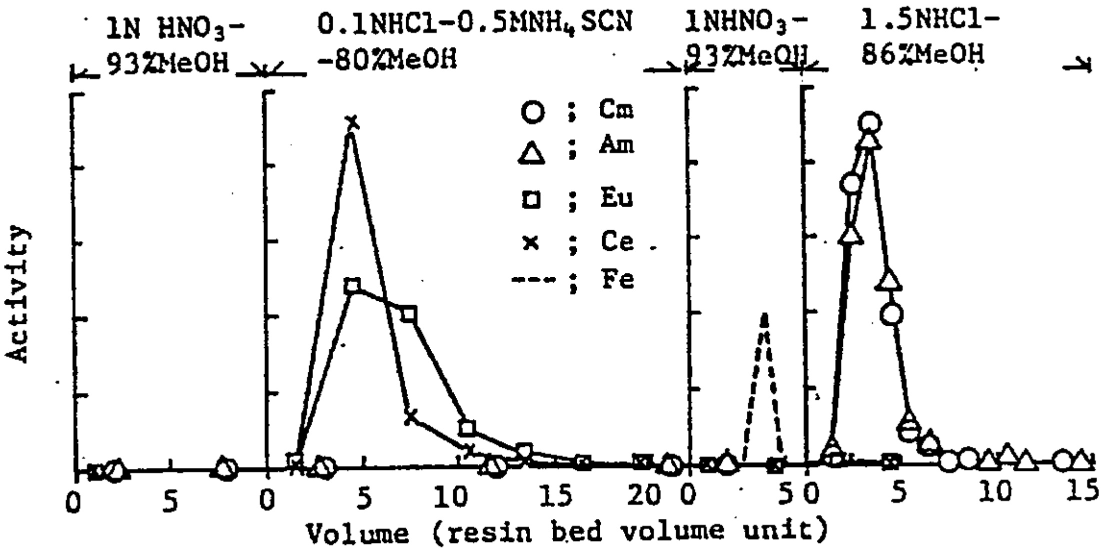


Figure 10 Sequential elution curves for Cm-244, Am-243, Eu-152 and Ce-144 (Dowex 1×4, 100-200mesh, 8mmf×12cm, 0.47ml/min.)

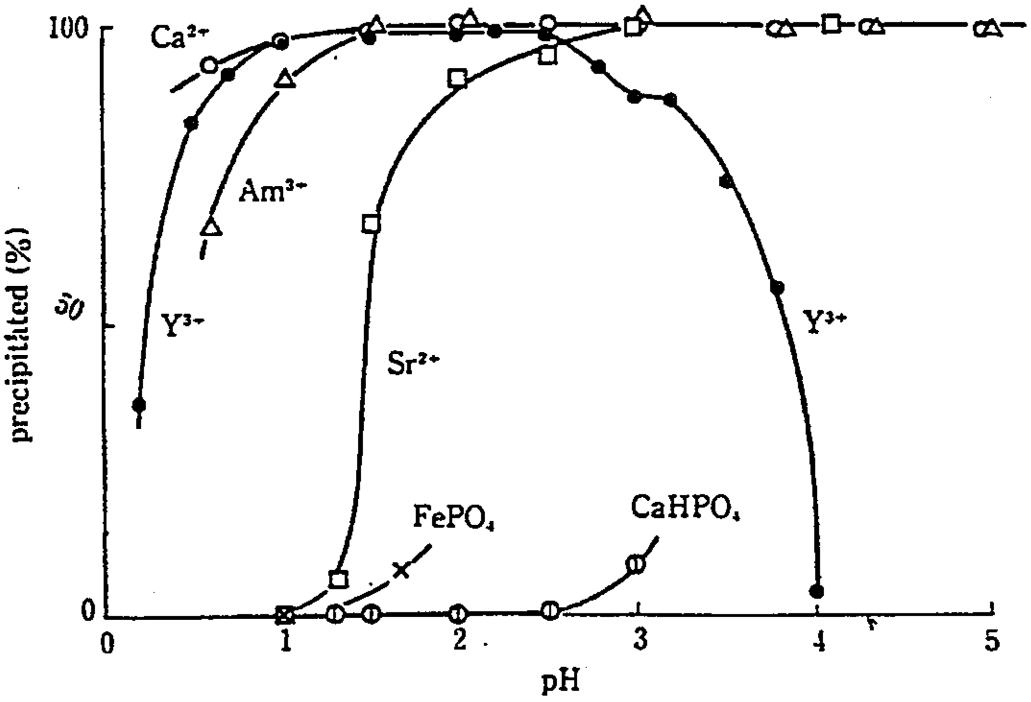


Figure 11 pH dependance of metal oxalate precipitate and coprecipitation of Am on Ca(COO)2

2.1.3 Electrodeposition

The recovery rate of electrodeposition depends on many factors such as current density, deposition time, pH of solution, interfering elements and surface condition of electrodeposition disk

The PNC's condition of electrodeposition is as follows

pH of solution is 2.
Current is 1.0A.
Deposition time is 120 minutes.

The relationship between deposition time and recovery rate is shown in Figure 12[12].

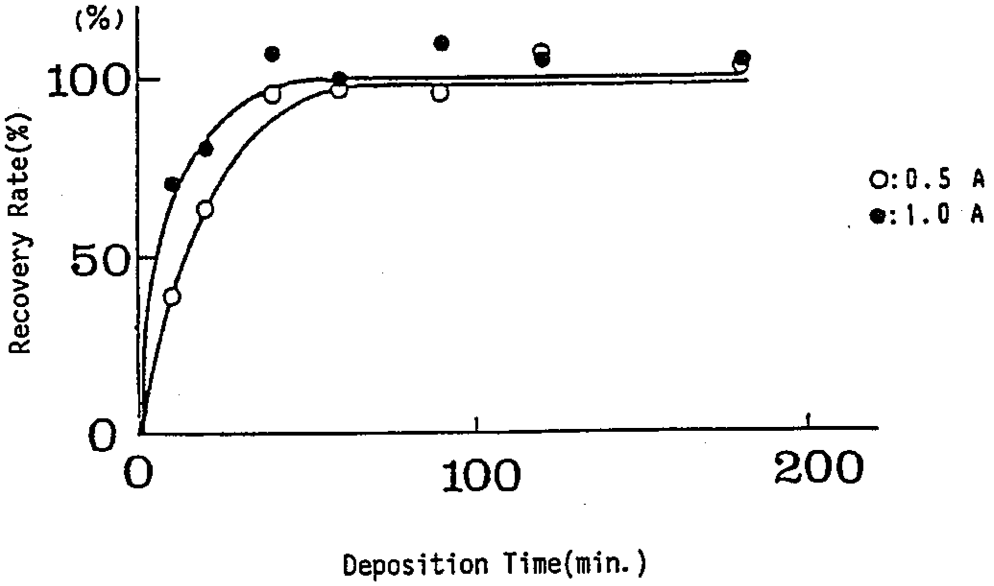


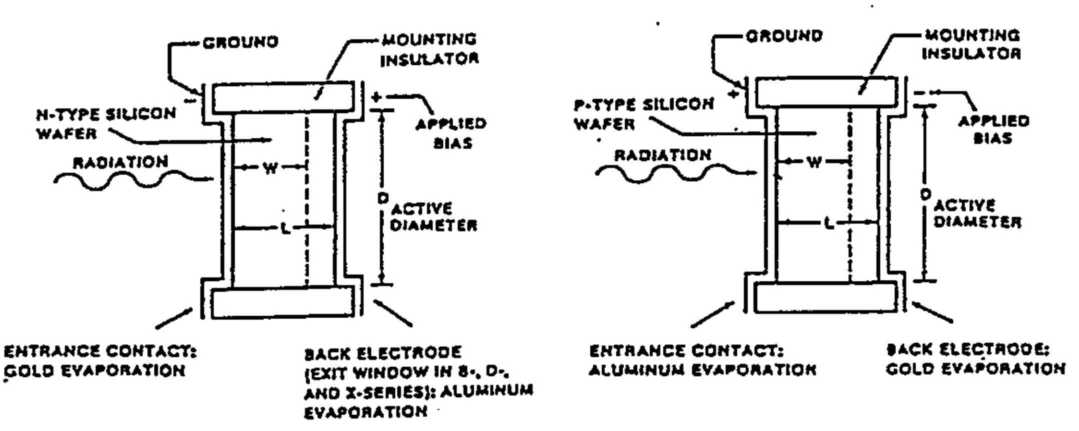
Figure 12 The relationship between deposition time and recovery rate
(H_2SO_4+NH_4OH solution) (STA, 1989)

2.1.4 Alpha-spectrometry

Structure of the Silicon Semiconductor Detector

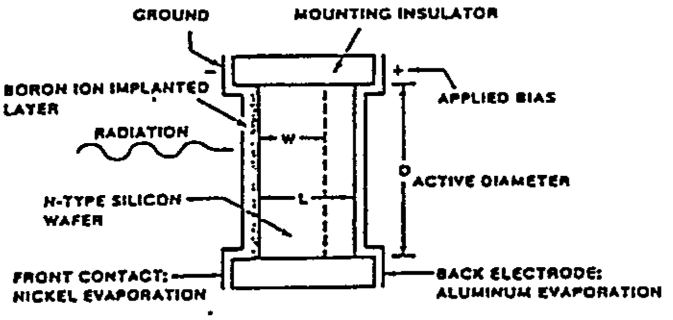
Figure 13(a) shows the basic structure of silicon semiconductor detector(SSD). The surface of the n-type silicon wafer is etched and then is covered with a piece of gold leaf so that the wafer may serve as the electrode. The SSD shown in Figure 13(b) is called reinforced type detector. In the case of the detector of this type, the p-type silicon wafer whose surface is covered with aluminum serves as the electrode. The surfaces of it are more resistive for damage than that of gold leaf-covered type, it is possible to clean the sensitive portions of the surface. And (c) type SSD has high counting efficiency for a charged particles. Though the counting efficiency of (b)type is about 25%, that of (c)type is over 35%.

And usually, the detectors with an effective area of 450mm^2 are used for measurement of environmental samples.



(a) A-, B-, C-, D-, E-, F-, and X-Series Detectors.

(b) R-Series (Ruggedized) Detectors. (ORTEC 576)



(c) I-Series (Ion Implanted) Detectors. (ORTEC 576A)

Figure 13 Designs of typical EG&G ORTEC charged particle detectors

2.2 Analytical Procedures of Plutonium-239,240 and Americium-241

The sequential analytical flow chart of Pu-239,240 and Am-241 is shown in Figure 14. It takes about 5 days for Pu analysis, and about 2 weeks for Am analysis. The details of these analyses are described in section 2.2.1 and 2.2.2.

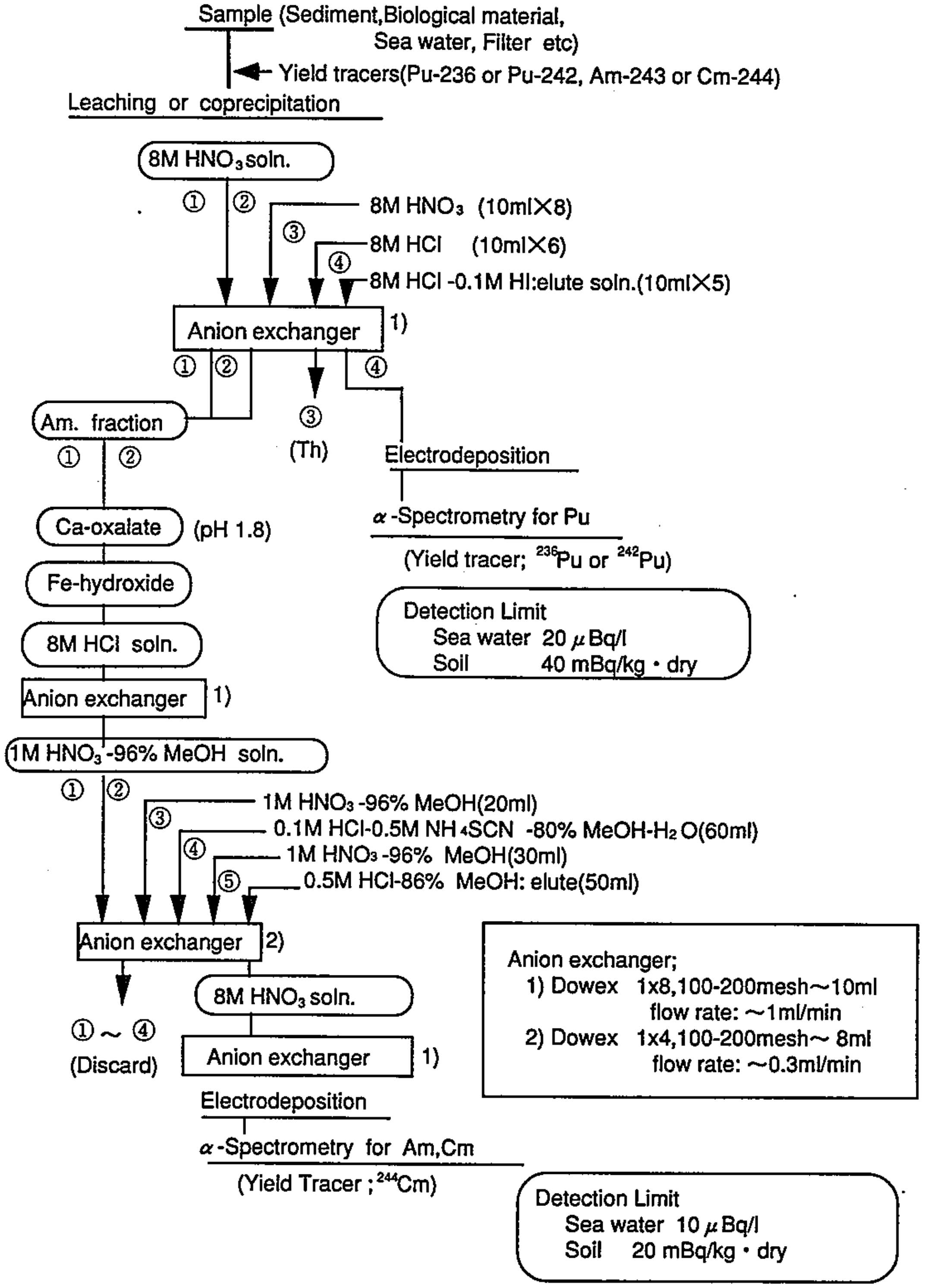


Figure 14 Sequential analytical flow chart of Pu-239,240 and Am-241

2.2.1 Analytical Method for Plutonium-239,240 in Marine and Agricultural Product Ashes

This analytical method is quoted from "manual of standard procedures for environmental sampling and analysis[13)]".

(1)Summary

Marine and agricultural product ashes undergo wet ashing with nitric acid and hydrogen peroxide. Then they are dissolved with 8N nitric acid. Next, the resultant solution are subjected to anion exchange separation. After being purified and eluted in the anion exchange processes, plutonium is electrodeposited on a stainless steel plate, and measured by silicon semi-conductor detector.

(2) Scope of Application

This method is applied to the analysis of Pu-239,240 in marine and agricultural product ashes. In case of 20g sample weight, 0.008cpm background, 80,000sec counting time, 25% counting efficiency and 60% recovery, then the detection limit is 2(mBq/sample).

(3)Flow Chart

Figure 15 shows the flow chart for Pu-239,240 in marine and agricultural product ashes.

(4) Reagents and Apparatuses

Reagents:

1) Pu-236 or Pu-242 standard solution: about 0.017Bq/ml
2) nitric acid HNO_3, nitric acid (8N)
3) hydrochloric acid HCl, hydrochloric acid (8N)
4) sulfuric acid H_2SO_4, sulfuric acid (1.8N)
5) perchloric acid $HClO_4$ (60%)
6) aqueous ammonia NH_4OH, aqueous ammonia (5N)
7) hydrogen peroxide solution H_2O_2 (30%)
8) octyl alcohol $CH_3(CH_2)_6CH_2OH$

9) thymol blue indicator; Dissolve 100mg thymol blue into 100ml alcohol.

10) elution solution for Pu - mixed solution based hydrochloric acid (HI: 0.1M, HCl: 8M); Mix 13.6ml HI (57%) and 708ml HCl, and dilute the mixture to 1 ℓ with water.

11) sodium hydroxide solution NaOH, sodium hydroxide solution (120g/ ℓ)

12) anion exchange resin: Perform following conditioning for Dowex 1 × 8 (100～200 mesh)

a) Put 500ml resin into 3 ℓ beaker, add 2 ℓ water, and stir up well. After the resin is settled, remove the supernatant by decantation. Repeat this procedure three times.

b) Add 1 ℓ sodium hydroxide solution (120gNaOH/ ℓ), stir up well, and remove the supernatant by decantation.

c) Add 1 ℓ water, stir up well, and remove the supernatant by decantation.

d) Add 2 ℓ hydrochloric acid (6N), stir up well, and remove the supernatant by decantation.

e) Rinse the resin with water in the same manner as in a).

f) Keep the resin under water.

g) Fill the column shown in Figure 16 with resin immediately before its use, and flow 10-CV (Column Volume) water and 10-CV nitric acid (8N) in this order through the column.

Apparatuses:

1) ion exchange column: Figure 16

2) electrodeposition cell: Figure 17 and Figure 18

3) electrodeposition plate: 1mm-thick, 1inch-dia. stainless steel plate (SUS-316)

4) electrodeposition device

5) alpha-ray spectrometer

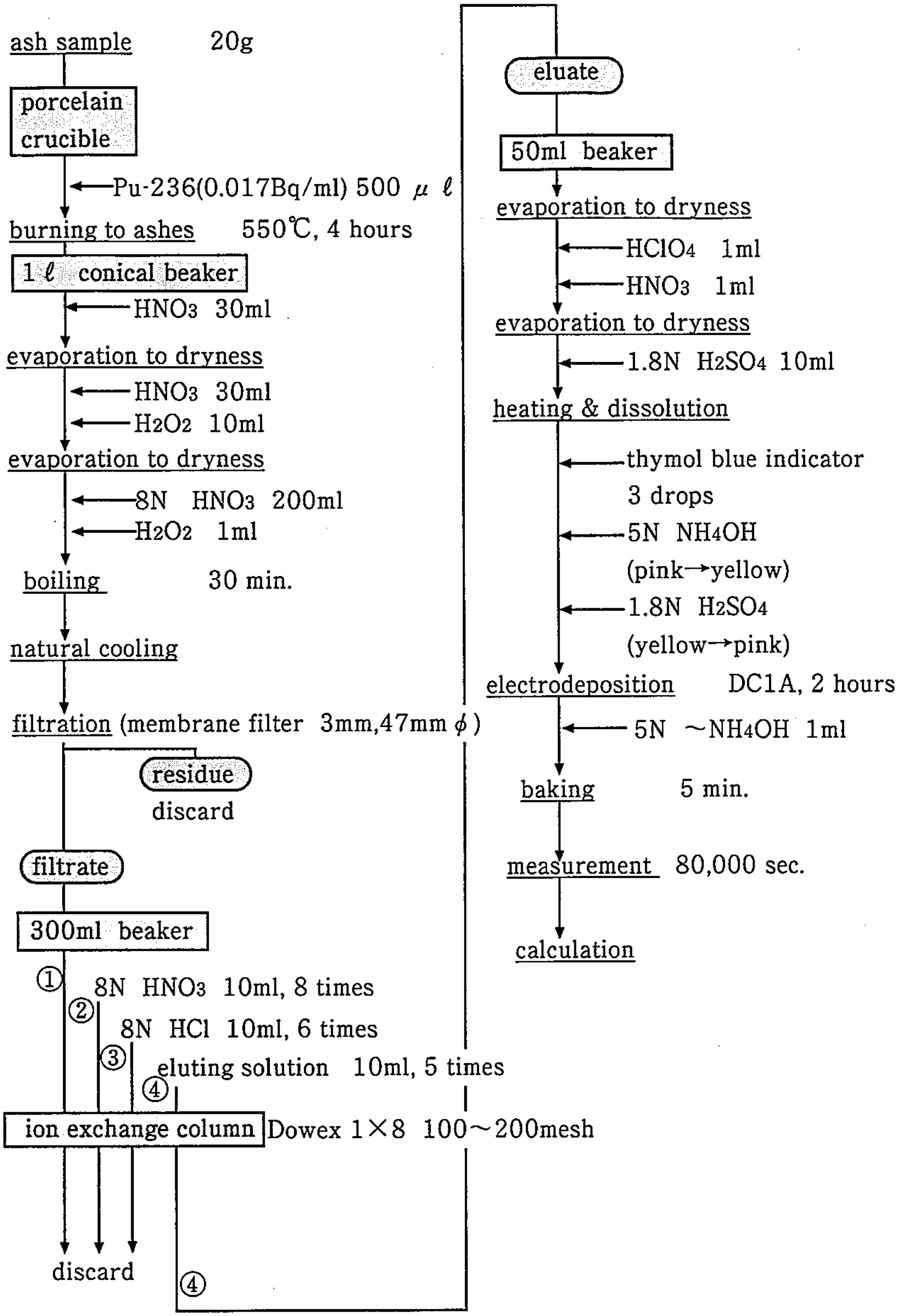


Figure 15 Flow chart for Pu-239,240 in marine and agricultural product ashes

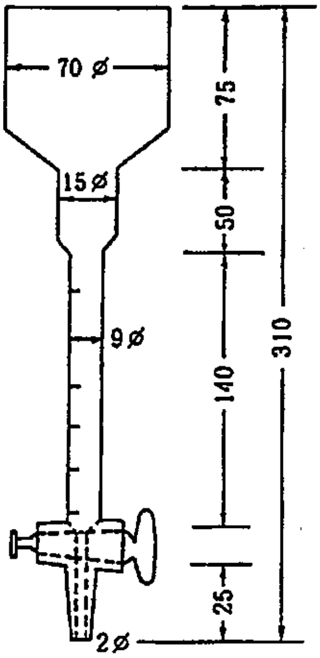


Figure 16 Ion exchange column

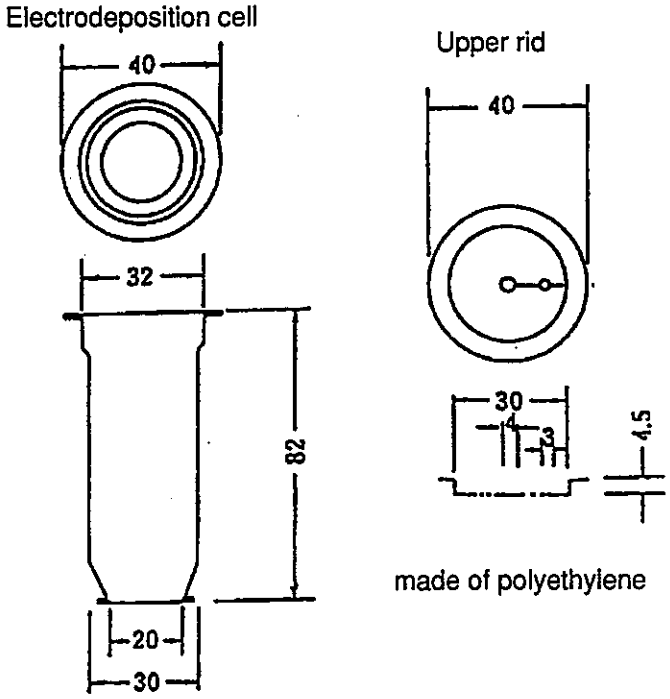


Figure 17 Electrodeposition cell

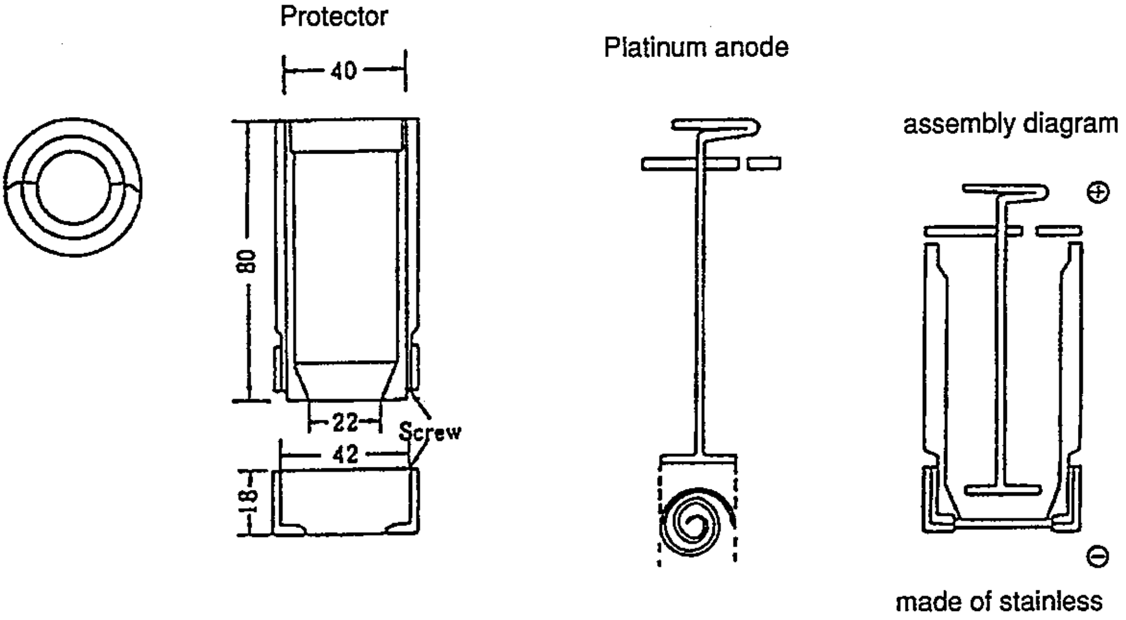


Figure 18 Electrodeposition cell, protector and platinum electrode

2.2.2 Analytical Method for Americium-241 in Seabed Sediment, Beach Sand and Soil

This analytical method is quoted from "manual of standard procedures for environmental sampling and analysis[13)]".

(1)Summary

Americium(Am) in a sample is leached out by nitric acid, separated from Pu with anion exchange resin with 8N nitric acid, followed by coprecipitated with calcium oxalate. The precipitate is subjected to ash and is dissolved in hydrochloric acid. The resultant solution is passed through anion exchange resin column with 8N hydrochloric acid in order to remove Fe, and then anion exchange resin column with mineral acid-methanol media in order to remove rare earth elements.

Finally, anion exchange with 8N nitric acid is applied to remove Th. Then Am is electrodeposited. Electrodeposited Am is then determined by the silicon semi-conductor detector.

(2)Scope of Applications

This method is applied to analyze Am-241 in seabed sediment, beach sand and soil. In case of 50g sample weight, 0.008cpm background, 200,000 second counting time, 25% counting efficiency, 60% recovery, then the detection limit is 1(mBq/sample).

(3)Flow Chart

Figure 19 shows the flow chart for Am-241 contained in seabed sediment, beach sand and soil.

(4) Reagents and Apparatuses

Reagents:

1) Cm-244 standard solution: about 0.017Bq/ml
2) nitric acid HNO_3, nitric acid (8N)
3) hydrochloric acid HCl, hydrochloric acid (8N)

4) sulfuric acid H_2SO_4, sulfuric acid (1.8N)

5) perchloric acid $HClO_4$ (60%)

6) aqueous ammonia NH_4OH, aqueous ammonia(5N), aqueous ammonia(0.05N)

7) hydrogen peroxide solution H_2O_2(30%)

8) sodium hydroxide solution NaOH, sodium hydroxide solution (120g/ ℓ)

9) thymol blue indicator; Dissolve 100mg thymol blue indicator in 100ml alcohol.

10) ammonium chloride NH_4Cl

11) oxalic anhydrous $H_2C_2O_4$

12) ammonium oxalate mono hydrate $(NH_4)_2C_2O_4 H_2O$

13) Fe carrier solution (20mgFe/ml); Dissolve 97.6g Iron(Ⅱ) chloride 6-hydrate ($FeCl_2$ $6H_2O$) in 50ml hydrochloric acid, and dilute with water to 1 ℓ .

14) Ca carrier solution (100mgCa/ml); Dissolve 294.6g extra pure calcium nitrate tetrahydrate ($Ca(NO_3)_2$ $4H_2O$) in water and make up the volume to 500ml.

15) ammonium thiocyanate NH_4SCN (Do not use metal spoons.)

16) methanol (extra pure) CH_3OH

17) mineral acid - methanol mixed solution

Prepare the following 3 kinds of solutions and keep in a sealed brown bottle.

(A) 1M HNO_3 - 93% CH_3OH

Put 100ml methanol into a 1 ℓ volumetric flask, add then 75ml nitric acid and stir up. Add methanol to the marked line, and stir up well.

(B) 0.1M HCl - 0.5M NH_4SCN - 80% CH_3OH

Put 40g ammonium thiocyanate into a 200ml beaker with a plastic spoon. Measure 800ml methanol, add it in the beaker by 100mls to dissolve ammonium thiocyanate, and transfer the resultant solution to hydrochloric acid, dilute the solution with 1 ℓ volumetric flask. Add 8.6ml hydrochloric acid, dilute the solution with water to the marked line.

(C) 1.5M HCl - 86%CH_3OH (Am eluting solution)

Measure 860ml methanol, add a portion of 100ml into a 1 ℓ volumetric flask, and add 130ml hydrochloric acid. Add the remaining methanol, and dilute the solution to 1 ℓ with water.

18) Anion exchange resin

(1) Dowex 1 × 8 (100 ~ 200mesh) for nitric-acid-type and hydrochloric-acid-type ion exchange resin.

Perform conditioning as follows:

a) Put 500ml resin in a 3 ℓ beaker, add 2 ℓ water, and stir up well. After settling, remove the supernatant by decantation. Repeat these operations three times.

b) Add 1 ℓ sodium hydroxide solution (120g/ ℓ) to the beaker, and stir up sufficiently. Remove the supernatant by decantation.

c) Add 1 ℓ water and stir up sufficiently. Remove the supernatant by decantation.

d) Add 2 ℓ hydrochloric acid (8N) and stir up well. Remove the supernatant by decantation.

e) Wash the resin with water in the same manner as described in a).

f) Store the resin in water.

g) Fill the column shown in Figure 22(A) or (C) with the resin immediately before use, and flow 10-C.V. (column volume) water. Flow 10-C.V. nitric acid (8N) for nitric-acid-type resin and 5-C.V. hydrochloric acid (8N) for hydrochloric-acid- type resin, respectively.

(2) Dowex 1 × 4 (50 ~ 100mesh) for mineral acid-methanol-type ion exchange resin

Perform conditioning as follows:

a) Put 500ml resin in a 3 ℓ beaker, add 2 ℓ water, and stir up well. After settling, remove the supernatant by decantation. Repeat these procedures three times.

b) Add 1 ℓ 1M nitric acid, and stir up sufficiently. After settling, remove the supernatant by decantation. Repeat these procedures three times.

c) Add 1 ℓ 1M nitric acid - 93% methanol mixture, and stir up sufficiently. After settling, remove the supernatant by decantation. Repeat these procedures three times.

d) Fill the column shown in Figure 22(B)with resin immediately before use, fill the upper zone of the resin with 5ml 1M nitric acid- 93% methanol mixture, and cover the column with a watch glass.

Apparatuses:

1) ion exchange column: Figure 22

2) electrodeposition cell: Figure 20 and Figure 21

3) electrodeposition plate: 1mm-thick, 1inch-dia. stainless steel plate (SUS-316)

4) electrodeposition device

5) alpha-ray spectrometer

6) microtube pump: flow rate 0.1～0.5ml/min. tube: inside dia. 1.5mm, outside dia.3.2 mm : Figure 23.

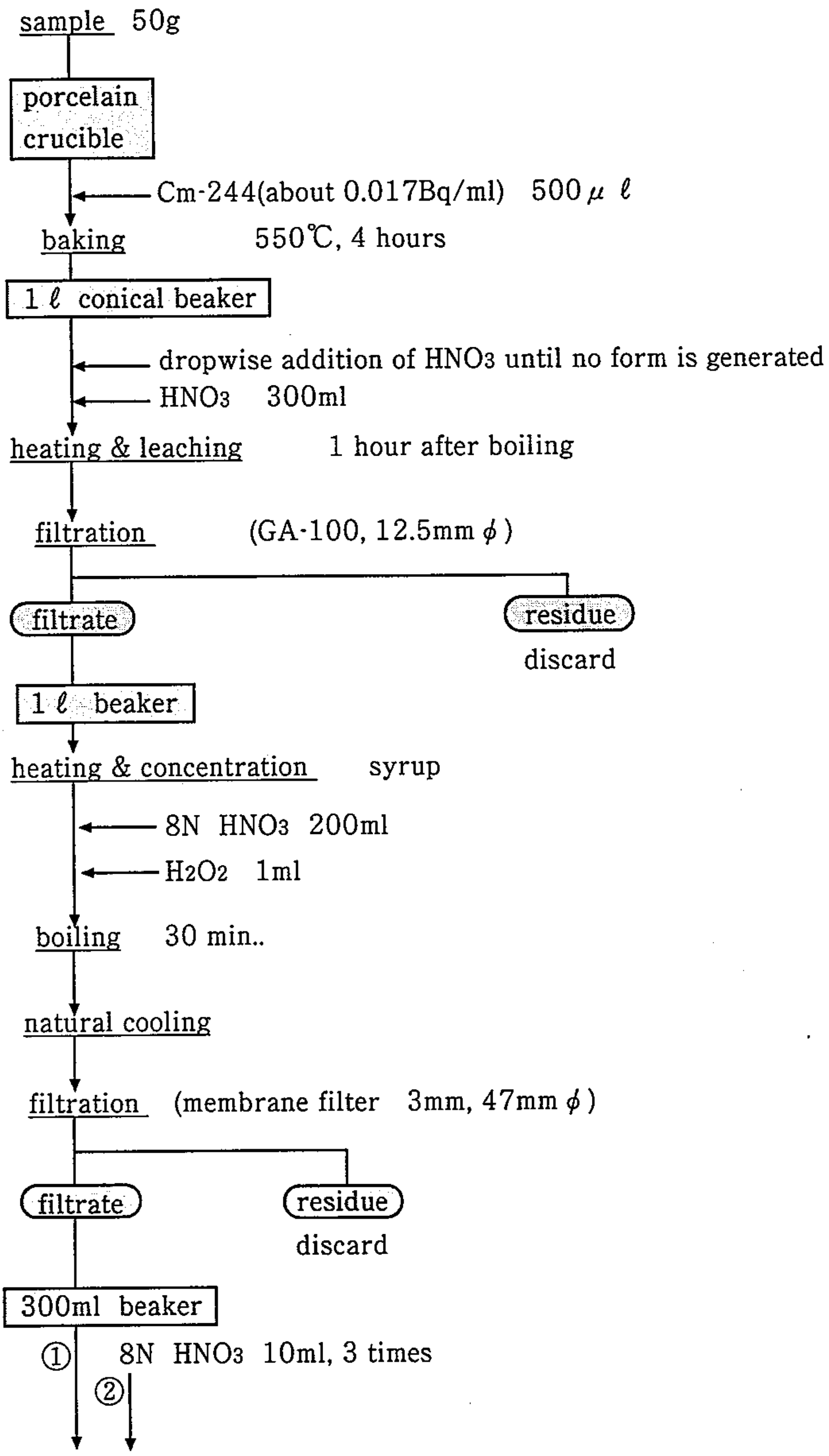


Figure 19 Flow chart for Am-241 in seabed sediment, beach sand and soil

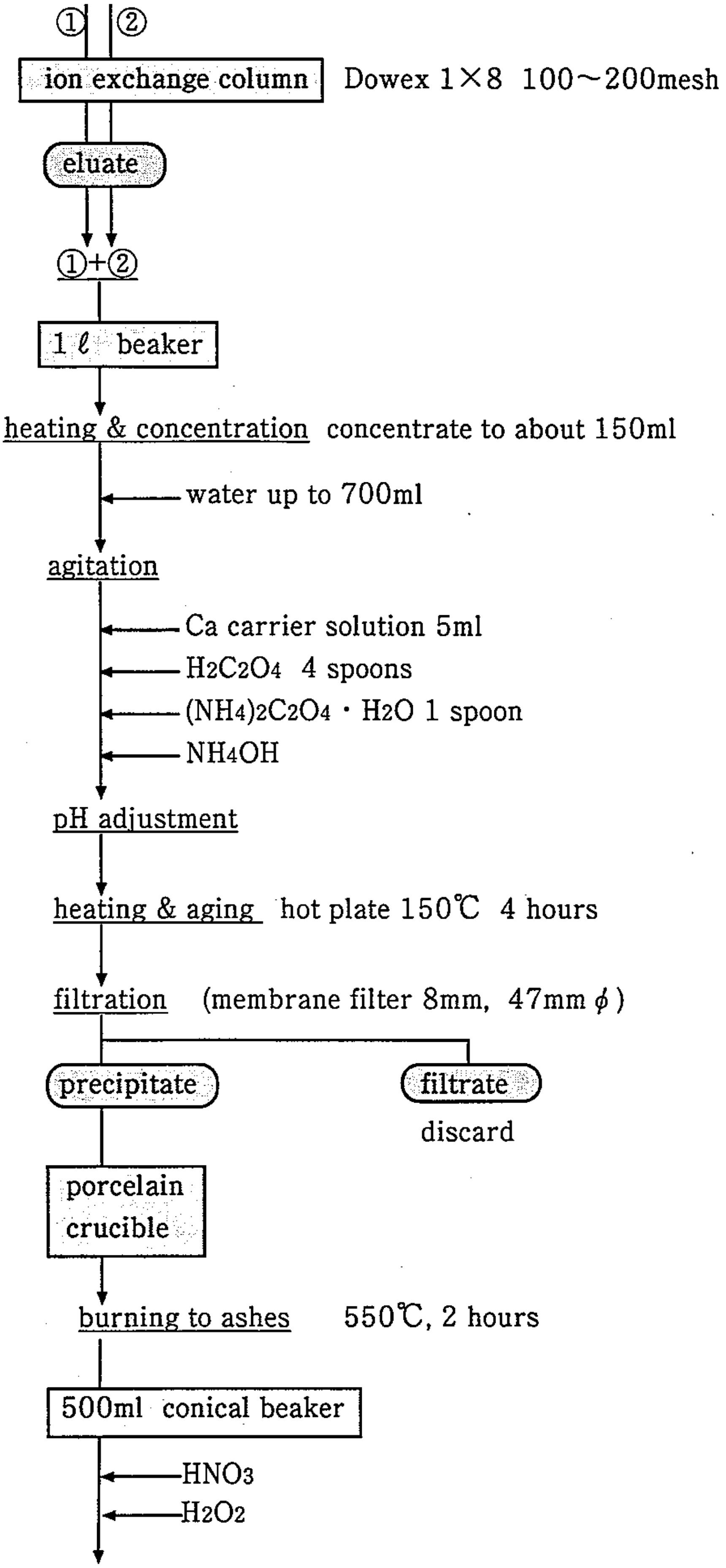


Figure 19 Flow chart for Am-241 in seabed sediment, beach sand and soil (Cont.)

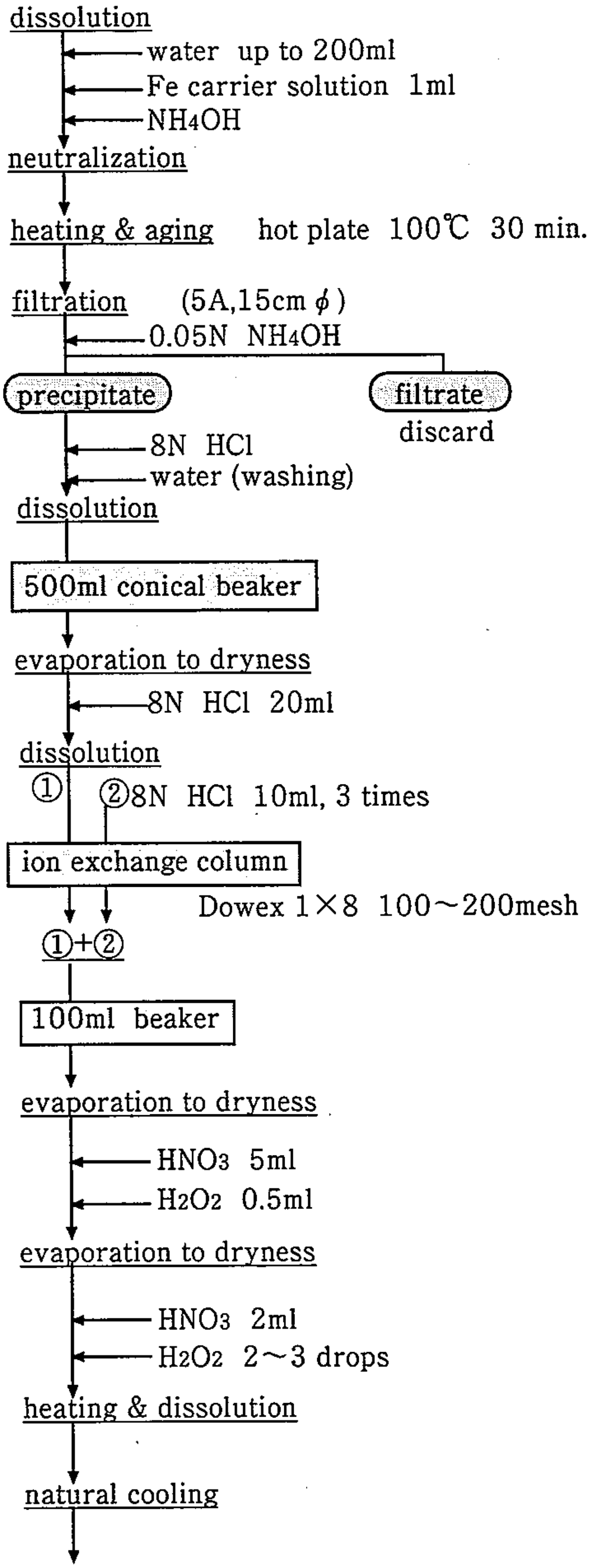


Figure 19 Flow chart for Am-241 in seabed sediment, beach sand and soil (Cont.)

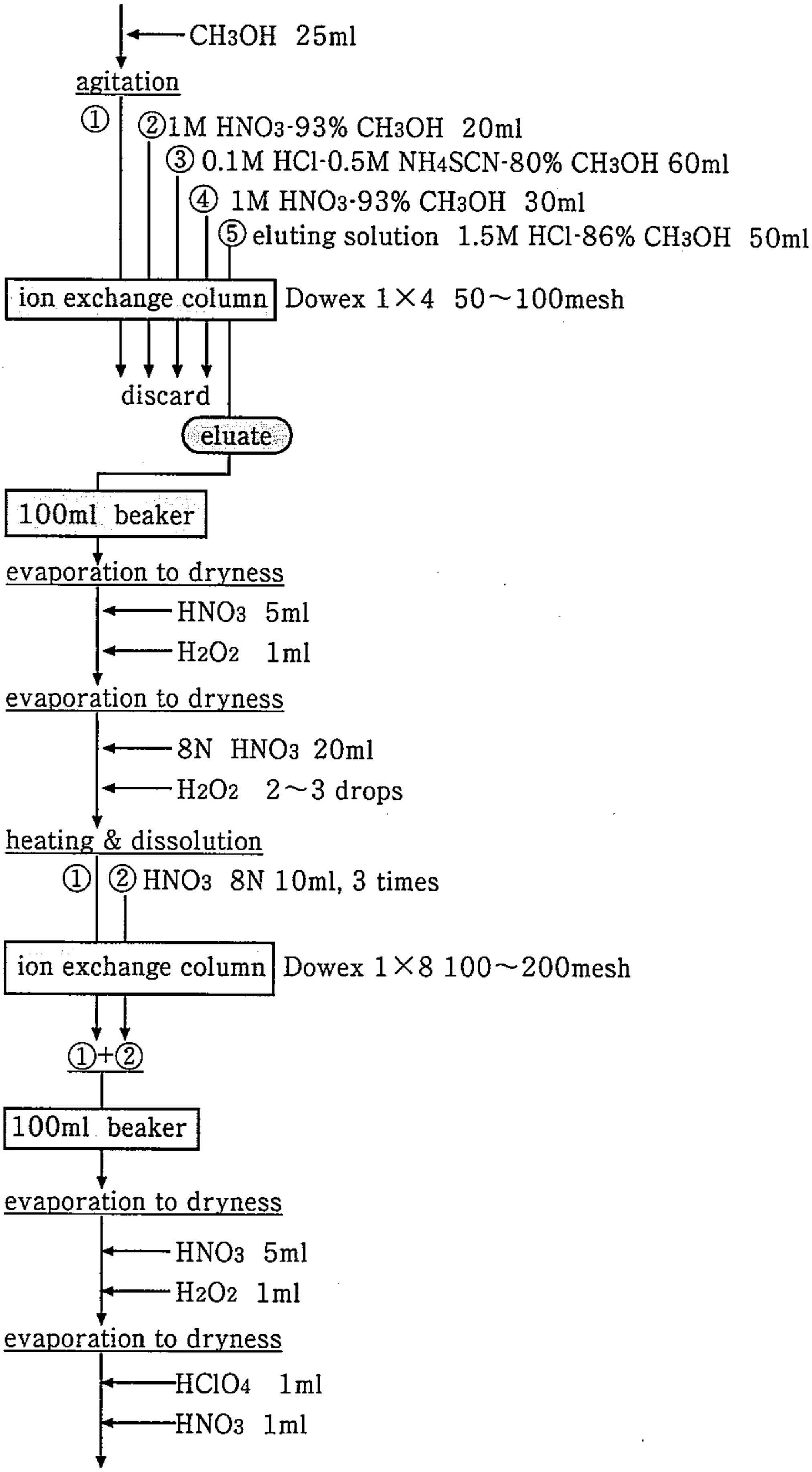


Figure 19 Flow chart for Am-241 in seabed sediment, beach sand and soil (Cont.)

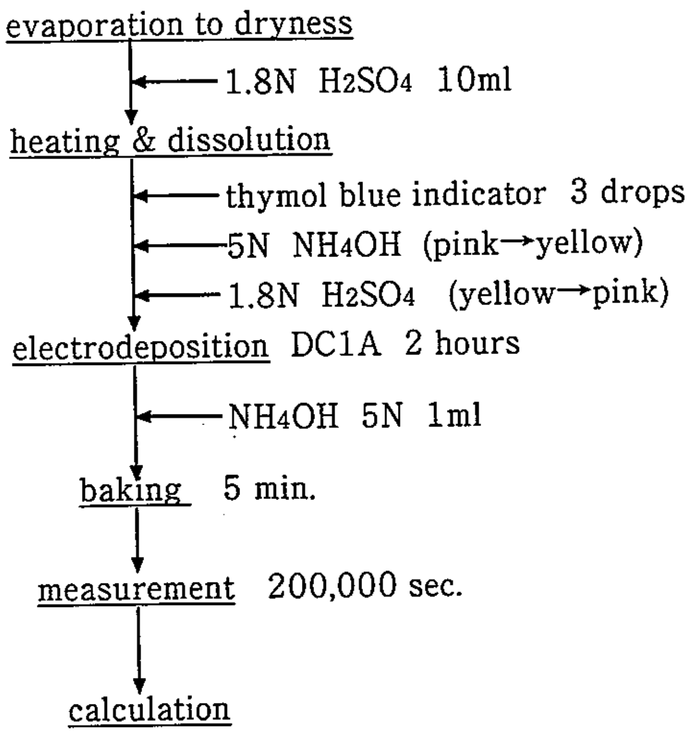


Figure 19 Flow chart for Am-241 in seabed sediment, beach sand and soil (Cont.)

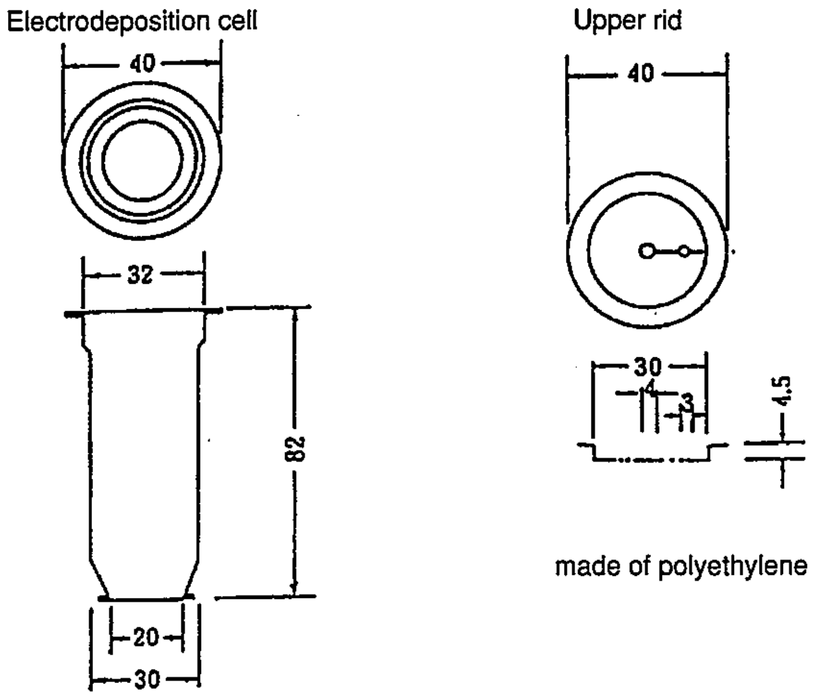


Figure 20 Electrodeposition cell

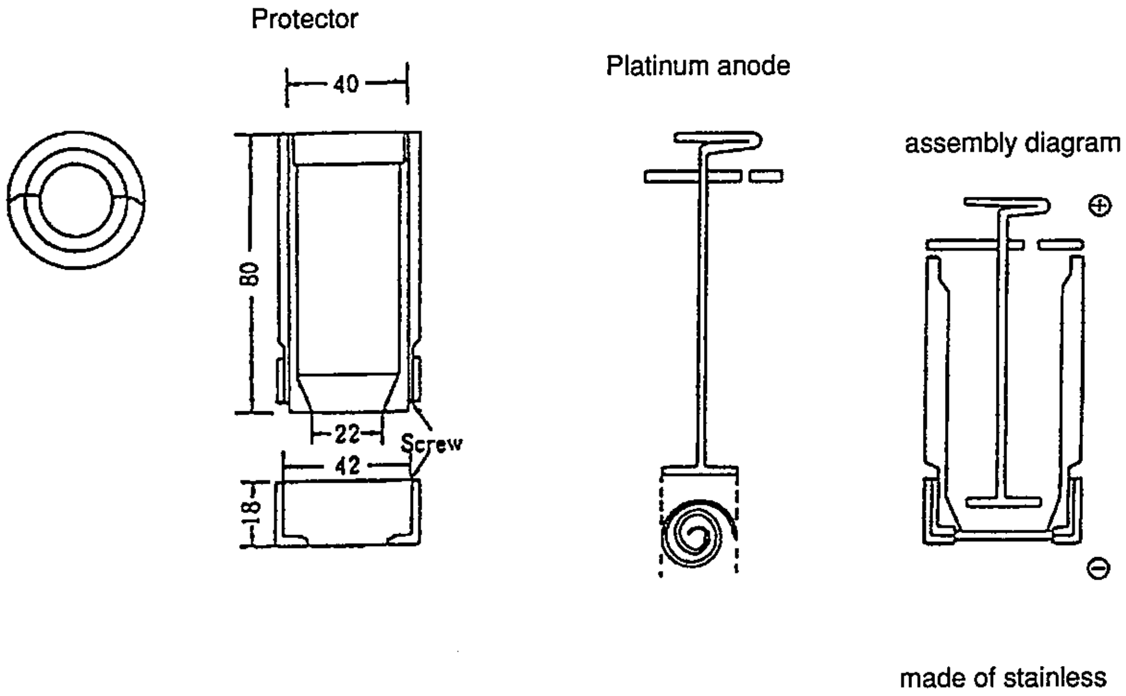
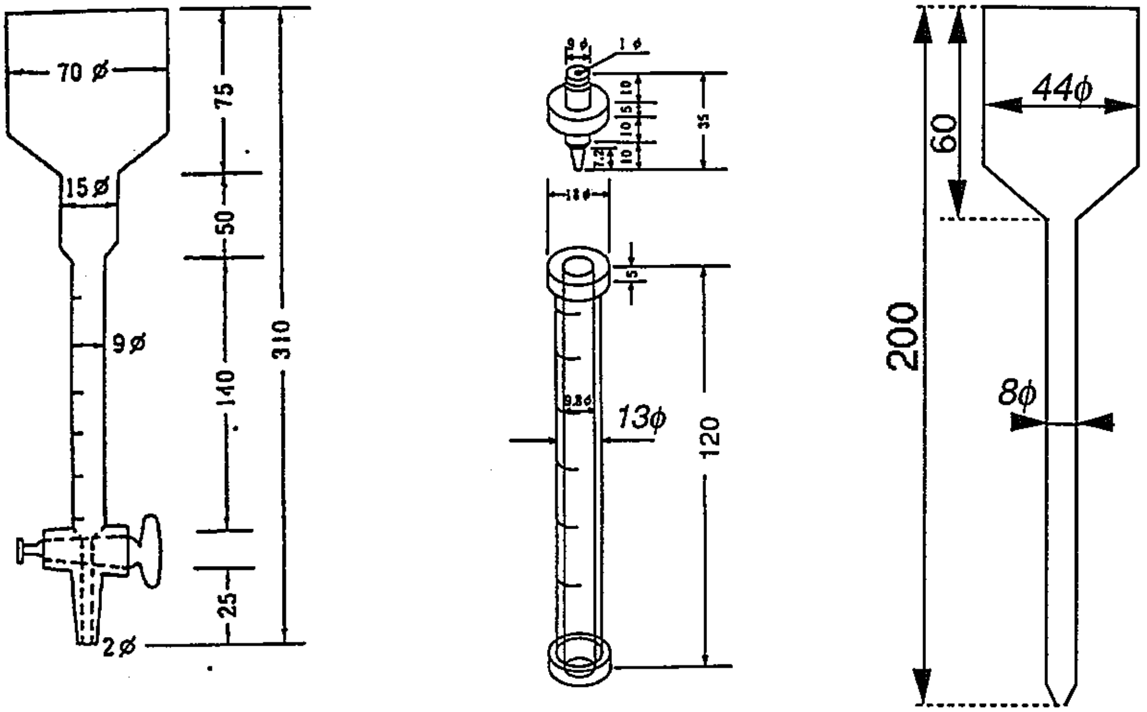


Figure 21 Electrodeposition cell, protector and platinum electrode



(A)

(B)

(c)

Figure 22 Ion exchange column

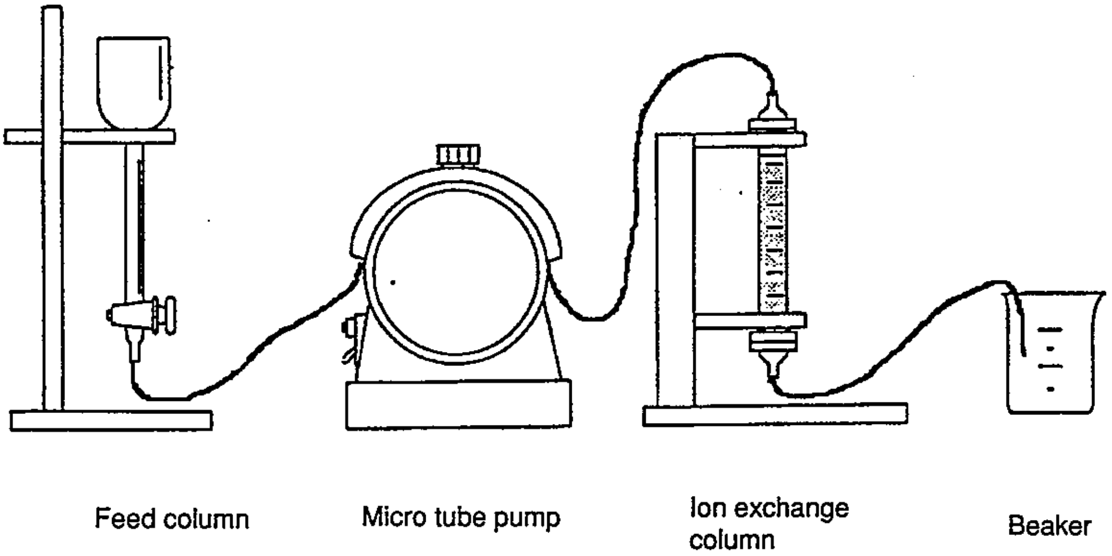


Figure 23 Alcohol ion exchange

2.3 Calibration and Measurement of Radioactivity

2.3.1 Energy Calibration

1) Pu-239, Am-241 and Cm-244 standard sources are measured.

2) Based on the spectra, the relationships among the peak center channels of Pu-239, Am-241 and Cm-244 and their alpha-particle energies are plotted in X-Y diagram.

Pu-239....5.157MeV
Am-241....5.486MeV
Cm-244....5.805MeV

3) A linear calibration equation is obtained from the diagram.

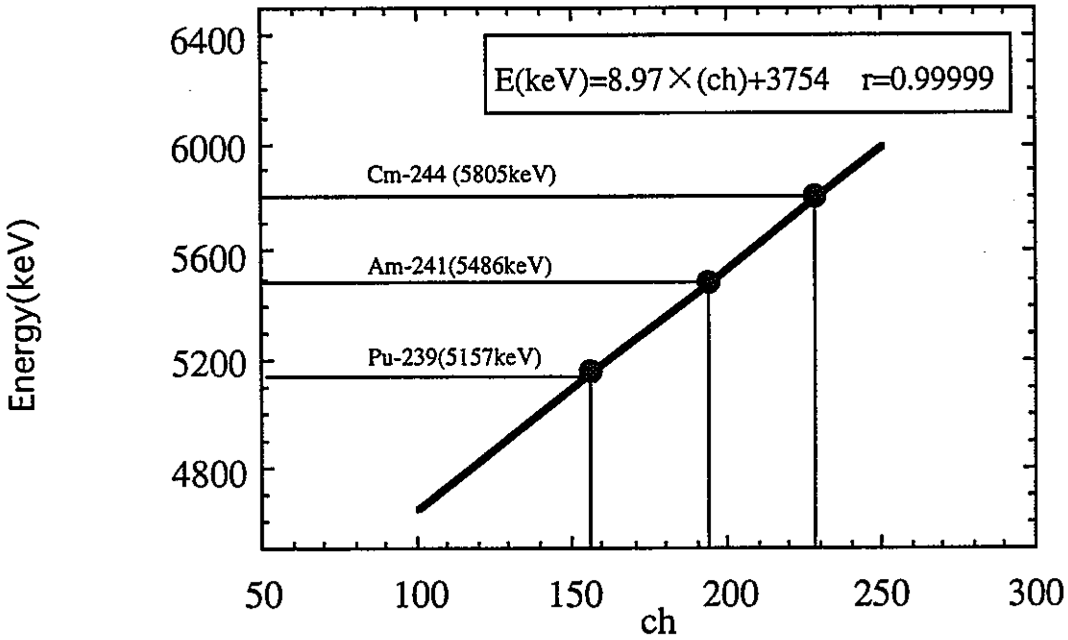


Figure 24 Energy calibration curve

2.3.2 Resolution (FWHM)

1) Alpha-ray spectrum using Am-241 standard source disk is measured for about 4,000-5,000 seconds (until the peak-channel counts reach approximately 5,000).

2) Measured counts in each channel are printed out.

3) The mono-energetic peak spectrum is plotted in a linear graph. Channels and the counts are plotted in X-Y diagram. A Gaussian spectrum is obtained.

4) A horizontal line through a half of the maximum counts of the spectrum is written in the graph. Two vertical lines through the intersections of the horizontal line and the spectrum are written. The intersections of those vertical lines and the axis of abscissa show their channel number.

5) The energies of those two channel numbers are determined using the energy calibration curve.

6) The difference between the both energies shows the degree of resolution and is named as full width at half maximum(FWHM).

7) Actual FWHM value depends on the performance of detectors to be used and usually ranges from 20 to 30 keV.

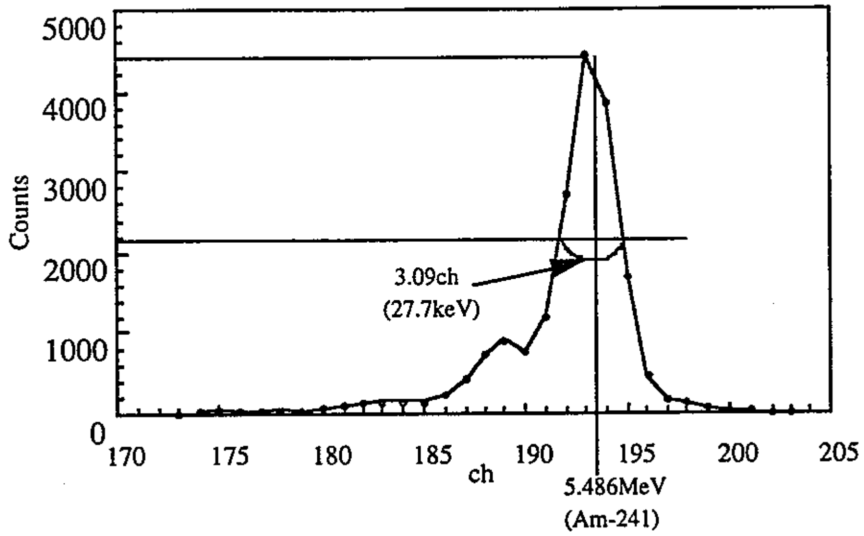


Figure 25 FWHM of Am-241

1): FWHM (Full Width at Half Maximum) is the peak full width at a half of the peak height. The value of FWHM indicates the width of the peak as well as the performance of the detector. It can also be used to check the degree of deterioration of the detector and the appropriateness of instrument regulation.

2.3.3 Counting Efficiency

The counting efficiency of the detector is determined by measuring standard reference sources. It should be noted, however, that the number of available alpha-ray sources is limited.

(1) Standard Alpha-ray Source

A standard alpha-ray source disk should be the same shape as the measurement sample, if possible, the same radionuclide as the one to be analyzed should be used as the standard source. If such source is not available, the one made by the use of a nuclide with almost the same alpha particle energy should be adopted as the standard source.

(2) Measurement and Calculation

a) Standard source and the background are measured. Measurement for standard source disk should be continued until the total counts of the peak area exceed 10,000.

b) The value of the background counting rate is subtracted from the value of the counting rate of the detected radioactivity of the standard source disk to find the value of the counting efficiency.

$$F = \frac{\left(\frac{S}{t} - \frac{B}{t_b}\right)}{S_o} \times 100$$

F :Counting efficiency (%)
S :Peak area detected in the standard source (counts)
B :Background area of the same channels as the peak of the standard source(counts)
t :Measurement time of the standard source (sec.)
t_b :Measurement time of the background (sec.)
S_o :Radioactivity of the standard source disk(Bq)

2.3.4 Calculation of Radioactivity

The radioactivity is calculated by comparing the background-subtracted peak counting rate of a chemical yield tracer with the background-subtracted peak counting rate of the radionuclide to be analyzed in the sample.

(1) Counting Rate of the Sample (including the background)

Identify the peak of the added tracer and the concerning nuclides by analysis the measurement sample spectra. It should be noted that, even with the same detector, the shape of peak and the central channel differ from one sample to another .

Determine the leftmost and rightmost channels of each peak area by analysis each peak broadening in the spectrum. As unsatisfactory chemical separation or electrodeposition will cause tailing, fractions below 1% of the counts of the entire peak area should be omitted.

Find the counts of each channel for the leftmost though the rightmost channel of the peak area and then find the total counts of each peak area (including the background) . Find the counting rate of the sample (including the background) by dividing the total counts by the measurement time.

$$\begin{array}{c}\{\text{the counting rate of the sample}\}\\ \text{(including the background)}\end{array}\ \text{(cps)}=\frac{N\pm\sqrt{N}}{t}$$

N : Total counts (including the background)(counts)

t : Measurement time (sec.)

(2)Background Counting Rate

$$\text{(Background counting rate) (cps)}=\frac{N_b\pm\sqrt{N_b}}{t_b}$$

N_b : Total counts (only the background)(counts)

t_b : Background measurement time (sec.)

(3) Net Counting Rate

Find the net counting rate by subtracting the background counting rate from the sample.

$$(\text{net counting rate})\ (\text{cps}) = \left(\frac{N}{t}-\frac{N_b}{t_b}\right) \pm \sqrt{\frac{N}{t^2}+\frac{N_b}{t_b^2}}$$

(4) Radioactivity in Samples

The radioactivity of added tracer's concentration is corrected up to the starting date of measurement for radioactive decay.

The radioactivity in the sample is found by comparing the net counting rate of the added tracer and the net counting rate of the nuclide to be analyzed.

The radioactivity in the unit weight of the sample is the above-mentioned radioactivity divided by the weight or volume.

$$A \pm \sigma A = \frac{D}{W}\left\{\frac{Cs}{Ct} \pm \frac{Cs}{Ct}\sqrt{\left(\frac{\sigma s}{Cs}\right)^2+\left(\frac{\sigma t}{Ct}\right)^2+\left(\frac{\sigma d}{D}\right)^2}\right\}$$

$A \pm \sigma A$:Radioactivity per unit weight or volume in sample
(Bq/kg or Bq/ℓ Supplied for analysis)
D :Radioactivity of added tracer after radioactive decay correction (Bq)
σd :Error of radioactivity of added tracer after radioactive decay correction (Bq)
Cs :Net counting rate of sample(cps)
σs :Counting error for net counting rate of sample (cps)
Ct :Net counting rate of tracer(cps)
σt :Counting error for net counting rate of tracer (cps)
W :Sample weight or volume (kg, ℓ)

(5) Chemical Recovery

The chemical recovery is not used in calculation conducted to obtain the results of measurement. With it, however, it is possible to ascertain whether chemical separation work is carried out correctly.

$$Y = \frac{Ct}{DF} \times 100$$

Y :Chemical recovery (%)
F :Counting efficiency of detector

When the recovery is low (less than 60%), you should analyze the same sample from the beginning.

3 Summary

In view of long-lived alpha emitters and dose assessment for public, Pu-239,240 and Am-241 are very important nuclides.

This report summarized general knowledge and analytical methods for Pu and Am. The analytical methods contained several chemical procedures and alpha ray measurement with some basic principles.

Our environmental monitoring data around Tokai reprocessing plant for Pu-239,240 and Am-241 show the same levels as those of background areas. Therefore these nuclides around Tokai reprocessing plant can be regarded as originated from global fallout of atmospheric nuclear explosion tests.

The authors hope this report is useful to analyze Pu-239,240 and Am-241 in environmental samples.

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