

# Sequential Isotopic Determination of Plutonium, Americium, Uranium, and Strontium in Soil Sample

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## Abstract

A procedure is developed to provide sequential analysis of  $^{238}\text{Pu}$ ,  $^{239/240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{238}\text{U}$ , and  $^{90}\text{Sr}$  in soil sample. Tracers and/or carriers ( $^{242}\text{Pu}$ ,  $^{243}\text{Am}$ ,  $^{232}\text{U}$ , and stable strontium) are added into the soil sample as chemical yield monitors, and then digested and extracted with nitric acid. Plutonium, strontium, americium, and uranium are sequential separated and purified by Dowex ion-exchange resin, EiChroM Sr-resin, EiChroM TRU-resin, and Chelate-100 resin, respectively.  $^{90}\text{Sr}$  is measured by Cerenkov counting. The actinides are determined by alpha-particle spectrometer. This method is verified by analyzed the U.S. National Institute of Standard and Technology NRIP2000 soil samples. The analytical results of  $^{241}\text{Am}$ ,  $^{238}\text{Pu}$ ,  $^{239/240}\text{Pu}$ ,  $^{238}\text{U}$ , and  $^{90}\text{Sr}$  agree with the NIST values within +5.7%, +2.4%, +1.6%, -2.2%, and -2.7%, respectively. According to the traceability limits defined in ANSI N42.22 criteria and measurement traceability to NIST for low-level radionuclides has been demonstrated to be better than 23%, 22%, 22%, 22% and 34% for  $^{241}\text{Am}$ ,  $^{238}\text{Pu}$ ,  $^{239/240}\text{Pu}$ ,  $^{238}\text{U}$ , and  $^{90}\text{Sr}$ , respectively.

Keywords: sequential analysis, NIST, traceability

## Introduction

$^{238}\text{Pu}$ ,  $^{239/240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{238}\text{U}$ , and  $^{90}\text{Sr}$  in the environmental samples, such as soil, vegetation, and water, are frequently analyzed for both emergency and routine radiation monitoring. Health physicists use the analytical results of environmental samples to estimate the amount of radioactive material present in the environment, calculating its burden for the radiological workers and the general public. For this study, a procedure is developed to provide sequential analysis of  $^{238}\text{Pu}$ ,  $^{239/240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{238}\text{U}$ , and  $^{90}\text{Sr}$  in soil sample, and a set of NIST NRIP 2000 soil samples were used to test the validation of this sequential analysis method.

## Experimental

### Sample preparation

The NRIP2000 soil samples were dry ashed at 450°C for 24 hours. The ashed soil samples were cooled to room temperature, transferred to a 1000 ml beaker, and tracers ( $^{242}\text{Pu}$ ,  $^{243}\text{Am}$ ,  $^{232}\text{U}$ ) and carriers ( $\text{Sr}^{2+}$ ,  $\text{Ca}^{2+}$ ) were added to the samples before digested with

concentrated nitric acid and hydrogen peroxide. The digested soil samples were finally dissolved in 8N nitric acid, filtered through glass-fiber filter papers, and heated to incipient dryness. The sequential analysis flowchart is shown in Figure 1.

#### Plutonium separation and purification <sup>1</sup>

Each sample was dissolved in 3:2 nitric acid (100ml) and heated to approximately boiled prior to addition of 1ml hydrogen peroxide so as to ensure oxidation of Plutonium (III) to Plutonium (IV). Gentle boiling was continued to remove the excess hydrogen peroxide, and each solution was cooled to room temperature before its introduction to an anion exchange column of 7cm length and 2cm internal diameter (Dowex 1x8, Cl-form, 100-200 mesh, SIGMA Chemical Co., U.S.A.) to affect isolation of plutonium. The column was rinsed with a further aliquot of 3:2 nitric acid (120ml) and the combined effluent retained for strontium, americium and uranium isolation. The anion exchange column was washed with 5:1 hydrochloric acid (300ml) to remove naturally occurring thorium species; plutonium was then removed from the column by reduction with a freshly prepared mixture of 29 Vol. % ammonium iodide (5 w/v %)-conc. hydrochloric acid. This solution was heated to incipient dryness, treated with nitric, perchloric acid, and then electrolyzed for 2 hours with platinum as an anode and a stainless steel plate as a cathode. The stainless steel plate was alpha-counted with an Ortec surface barrier detector assembly (EG&G Ortec, U.S.A.).

#### Strontium separation and purification <sup>2</sup>

Oxalic acid (20 g) was added to the retained strontium, americium and uranium fraction, and the pH of the solution was adjusted to 4.2 to form the precipitation of oxalate. The oxalate was filtered through Whatman 42 filter papers, and the filtrate retained for americium and uranium isolation. The oxalate was dissolved in 8M nitric acid (10 ml), transfer to a Sr.Resin extraction chromatography column (EiChroM Industries Inc., U.S.A.), and then rinsed with a further aliquot of 8M nitric acid (15 ml). Twenty milliliter of water was added to the column to afford strontium removal. The effluent was collected and heated to dryness, and the strontium nitrate was then weighed to calculate the recovery of strontium. The strontium nitrate was dissolved in 8M nitric acid (20 ml), transfer to a polyethylene low-diffusion liquid-scintillation vial, and, after 14 days later, subjected to Cerenkov counting on a Packard 2260TR liquid-scintillation counter (Packard Instrument Company, U.S.A.).

#### Americium separation and purification <sup>3,4</sup>

Carrier ( $\text{Ca}^{2+}$ , 100 mg), Oxalic acid (5 g) and ammonium oxalate (5 g) was added to the retained americium and uranium fraction, the pH of the solution was adjusted to 1.5, and the precipitation of oxalate is formed. The oxalate was filtered through Whatman 42 filter papers, and the filtrate retained for uranium isolation. The oxalate combined with the filter paper was

ashed at 550°C for 4 hours. The ashed samples were cooled to room temperature, dissolved in the solution of 1 M Al(NO<sub>3</sub>)<sub>3</sub> in 3 M HNO<sub>3</sub> (10 ml) after the addition of ascorbic acid (100 mg), and then transfer to a TRU.Resin extraction chromatography column (EiChroM Industries Inc., U.S.A.). Sequentially rinse the column with 2 M nitric acid (5 ml) and 0.5 M nitric acid (5 ml), and the effluent was discarded. Stripped the americium with 9 M hydrochloric acid (3 ml) and 4 M hydrochloric acid (15 ml). Collected this eluate and heated to dryness, and an alpha spectrometry source prepared according to the procedure for plutonium (see above).

#### Uranium separation and purification <sup>5</sup>

The retained uranium fraction was heated to boiled for 10 minutes, and cooled to room temperature. Adjusted the pH value of solution to 3.0, and 10 ml of chelating resin (Chelex-100, Na-form; 50-100 mesh, SIGMA Chemical Co., U.S.A.) was added to the solution. Stirred for 2 hours, the uranium was adsorbed on the resin. The resin was filtered through Whatman 42 filter papers, and the uranium was then desorbed by ammonium carbonate solution (3 w/v %). This solution was heated to dryness, and an alpha spectrometry source prepared according to the procedure for plutonium (see above).

### Results

The results of measurement and traceability for NRIP 2000 soil samples is listed in Table 1. A comparison of reported mean values shows that the <sup>241</sup>Am, <sup>238</sup>Pu, <sup>240</sup>Pu, <sup>238</sup>U and <sup>90</sup>Sr values agree with the NIST value within +5.7%, +2.4%, +1.6%, -2.2% and -2.7%, respectively. According to the traceability limits defined in ANSI N42.22, the <sup>241</sup>Am, <sup>238</sup>Pu, <sup>240</sup>Pu, <sup>238</sup>U and <sup>90</sup>Sr values have met the ANSI N42.22 criteria and measurement traceability to NIST for low-level radionuclides has been demonstrated to be better than 23%, 22%, 22%, 22% and 34% for <sup>241</sup>Am, <sup>238</sup>Pu, <sup>240</sup>Pu, <sup>238</sup>U and <sup>90</sup>Sr, respectively.

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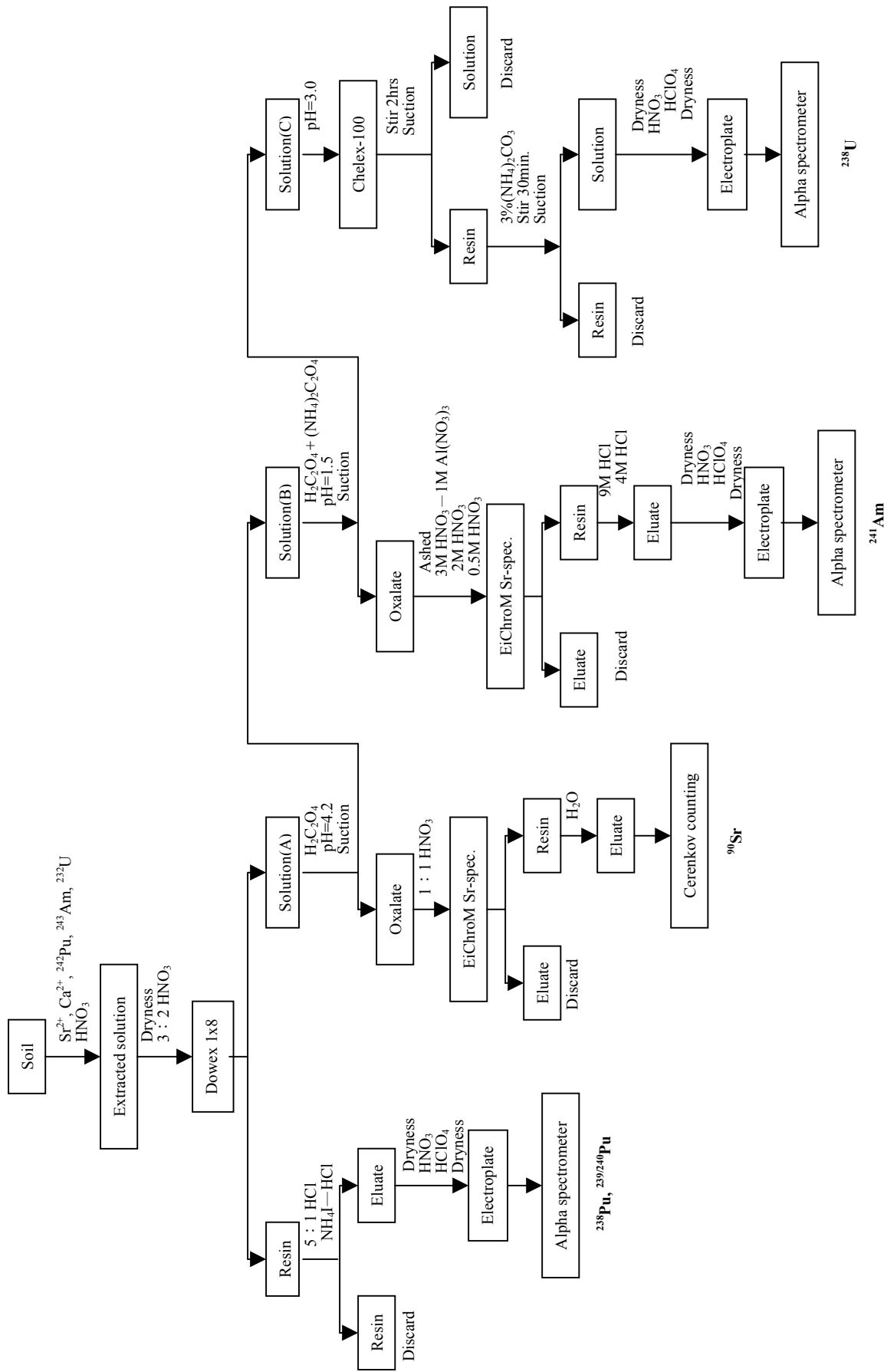


Figure 1. The flowchart of sequential procedure for isotopic analysis of plutonium, americium, uranium and strontium.

Table 1. The results of measurement and traceability for NRIP 2000 soil samples.

Nuclide	Measurement Results						
	INER Value		NIST Value		Difference (±%)	Traceability Limit (±%)	ANSI N42.22 Traceable
	Massic Activity Bq/g	Expanded Uncertainty (%,k=2)	Massic Activity Bq/g	Expanded Uncertainty (%,k=2)			
<sup>241</sup> Am	0.7545	14.6	0.715	0.64	+5.7	23	Yes
<sup>238</sup> Pu	0.7472	14.2	0.730	1.10	+2.4	22	Yes
<sup>239/240</sup> Pu	0.7248	14.2	0.713	0.78	+1.6	22	Yes
<sup>238</sup> U	0.6876	15.0	0.703	0.60	-2.2	22	Yes
<sup>90</sup> Sr	0.7267	23.22	0.747	0.74	-2.7	34	Yes