



Alpha spectrometry applications with mass separated samples



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HIGHLIGHTS

- High resolution alpha samples created using a commercial ICP-MS.
- Mass-based isolation of a plutonium isotope (^{238}Pu) from a mixed plutonium standard.
- Fit technique with alpha spectroscopy quantifies the enhancement of ^{238}Pu signature.

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ABSTRACT

^{241}Am has been deposited using a novel technique that employs a commercial inductively coupled plasma mass spectrometer. This work presents results of high-resolution alpha spectrometry on the ^{241}Am samples using a small area passivated implanted planar silicon detector. We have also investigated the mass-based separation capability by developing a ^{238}Pu sample, present as a minor constituent in a ^{244}Pu standard, and performed subsequent radiometric counting. With this new sample development method, the ^{241}Am samples achieved the intrinsic energy resolution of the detector used for these measurements. There was no detectable trace of any other isotopes contained in the ^{238}Pu implant demonstrating the mass-based separation (or enhancement) attainable with this technique.

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1. Introduction

Alpha spectrometry is a common analytical technique for the detection of alpha emitting radionuclides. The main reasons for this are low backgrounds, high detection efficiency and relatively low-cost. Other parameters drive the choice of the specific detector. The summary prepared by Vajda et al. (2012) provides a well-written overview of alpha spectrometry applications, detector options, etc.

Room temperature silicon detectors with ion implanted contacts (“windows”) are the most common choice amongst detectors used in alpha spectrometry due to their thin dead layers (50–100 nm) which minimize energy straggling. Furthermore, they are offered in a variety of sizes, provide good energy resolution, rugged contacts that can be easily cleaned and high detection efficiency. The energy resolution of the detector improves with decreasing the active area at the cost of geometrical (detection) efficiency. High-resolution alpha spectrometry involves the

production of samples which are thin and free of contaminants and a small-area PIPS detector that attains <15 keV full-width-at-half-maximum (FWHM) energy resolution.

Pacific Northwest National Laboratory (PNNL) has internally funded a project to investigate the feasibility of using the inherent mass-based separation capabilities of a commercial inductively coupled plasma mass spectrometer (ICP-MS) to reduce some of the tedious chemical separation procedures needed for certain radiometric techniques. This work discusses new results of the Atomic Mass Separation for Enhanced Radiation Detection Measurements (MS-RAD) project. We previously reported (Dion et al., 2014) the results of two ^{241}Am samples (“implants”) and the results of energy resolution improvement using low-resolution alpha spectrometry. New results using a high-resolution alpha spectrometer substantiate our previous results and also allow comparison of the two MS-RAD samples. Furthermore, a mass 238 (AMU) implant was derived from a ^{244}Pu NIST standard where ^{238}Pu was a minor constituent. An analysis was performed to determine the enhancement of the ^{238}Pu signature of the MS-RAD implant compared to an electrodeposited sample created from the unseparated NIST standard.

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2. Experimental methods

2.1. ICP-MS implants

The details of the implant apparatus used in the quadrupole ICP-MS were previously described in detail (Dion et al., 2014; Liezers et al., 2015). A brief description of the method is supplied here. The ICP-MS was a modified model PQ Excell (VG-Elemental) with an S-option interface where the sample introduction area, ICP and plasma sampling interface were enclosed by a custom fume hood designed specifically for the analysis of radiological samples. The system has a nominal ion transmission efficiency of 0.024%. The standard electron multiplier of the ICP-MS was removed and replaced with a custom standoff. A copper metal block was centered on the standoff and positioned 1 mm behind the quadrupole exit ion lens. A 15.9 mm diameter stainless steel planchette was mounted on the copper block with conductive silver epoxy where the ion of interest could be implanted.

Two ^{241}Am MS-RAD samples were developed for the high-resolution alpha spectrometry derived from a commercial gamma standard solution with an activity of $\sim 3.7 \text{ E4 Bq g}^{-1}$. The implanted samples were targeted to have an activity of $\approx 0.1 \text{ Bq}$. The difference between the two samples was the potential difference applied to the planchette and exit ion lens during implantation. For the “low-bias” sample, the exit ion lens was held at +30 V and the planchette was held at ground. In this configuration, the implanted ion current could be monitored through a feedthrough via an external electrometer. For the “high-bias” sample, the exit ion lens was held at ground potential and the implant was held at -1500 V ; the ion current could not be monitored since this potential difference was too large for the electrometer. The stock ^{241}Am solution was diluted to a concentration of 1.04 ppb in a 10 mL volume using 0.3 M HCl for the low-bias sample, and the high-bias sample used a diluted solution of 11.6 ppb to reduce the implant time to 18 min. The ICP-MS parameters for the ^{241}Am implants are summarized in Table 1.

To investigate the mass-based separation capability of the technique, a mass 238 (AMU) implant was created from a dilution of a ^{244}Pu standard (National Bureau Standards Standard Reference

Table 1

A summary of the ICP-MS operating conditions used for the ^{241}Am and ^{238}Pu implants.

Parameter	^{238}Pu	^{241}Am	Unit
ICP RF power	1350	1350	Watts
Argon cool flow	12	13	L min ⁻¹
Argon auxiliary flow	1.2	0.8	L min ⁻¹
Argon nebulizer flow	1.0	0.95	L min ⁻¹
Solution uptake	0.2	0.2	mL min ⁻¹
Spray chamber temp.	+2	+3	°C
Spray chamber type	Impact	Impact	
	Bead	Bead	
Nebulizer	Quartz	Quartz	
(AR35-1-FM02E)	Concentric	Concentric	
Sample cone aperture	1.0	1.0	mm
Skimmer cone aperture	0.7	0.7	mm
Expansion chamber press.	1.9	1.9	mbar
Analyzer press.	9.0×10^{-7}	9.0×10^{-7}	mbar
Extraction lens potential	-550	-550	V
Ion lens (L1)	+6	5	V
Ion lens (L2)	-40	-40	V
Ion lens (L3)	-200	-200	V
Hexapole bias	-5	-3	V
Focus	+22	+28	V
Differential aperture	-100	-75	V
Ion Lens (D1)	-35	-39	V
Ion Lens (D2)	-110	-168	V
Quadrupole bias	-2.0	-1.0	V

Table 2

The time-decayed isotopic atomic abundance [%] and alpha activity of the ^{244}Pu standard used in this work as of September 2014. The table only contains isotopes and/or daughters with an atomic abundance greater than 0.001 %. The alpha activity is based on 570 pg of the ^{244}Pu standard.

Isotope	Atoms (%)	Alpha activity (mBq)
^{234}U	0.00119 ± 0.002	$1.56 \times 10^{-3} \pm 0.31 \times 10^{-3}$
^{236}U	0.00245 ± 0.00002	$3.35 \times 10^{-5} \pm 0.02 \times 10^{-5}$
^{237}Np	0.00255 ± 0.002	$3.78 \times 10^{-4} \pm 0.08 \times 10^{-4}$
^{238}Pu	0.00381 ± 0.001	13.8 ± 2.8
^{239}Pu	0.0340 ± 0.001	0.445 ± 0.013
^{240}Pu	0.675 ± 0.004	32.4 ± 0.2
^{241}Pu	0.0174 ± 0.0003	-
^{241}Am	0.0721 ± 0.002	52.2 ± 1.1
^{242}Pu	1.32 ± 0.004	1.11 ± 0.01
^{244}Pu	97.87 ± 0.008	0.374 ± 0.001

Material 996, NIST, 2014). The time-decayed isotopic atomic abundance (as of September 2014) of the ^{244}Pu standard is shown in Table 2 (Robinson et al., 2014), half-life values and uncertainties were taken from ENDF (Chadwick et al., 2011). As seen in Table 2, several plutonium isotopes at varying abundances are present, however, mass 238 was selected as the implant species. Room temperature alpha spectroscopy cannot fully deconvolve the alpha particles of ^{239}Pu and ^{240}Pu or ^{238}Pu and ^{241}Am . Therefore, if one wants to quantify (without deconvolution) the amount of ^{238}Pu in a sample that contains ^{241}Am , a chemical separation would be performed to remove the americium. We selected mass 238 because there was enough activity in $\approx 600 \text{ pg}$ of the standard to make a detection after implantation and the influence of the ^{238}Pu to the ^{241}Am energy region would be observable in an unseparated sample (i.e., the activities of ^{238}Pu and ^{241}Am are within an order of magnitude). In contrary, the amount of ^{239}Pu in the standard would have eliminated this opportunity.

The stock concentration of SRM-996 was 3 ppm in 0.5 M HCl with initial activity and isotopic ratios measured on May 17, 1980. A 10 ml volume of the standard was prepared to a concentration of 50 ppb in 0.4 M HCL. The implant ion current was experimentally determined with a 500 ppb solution of Lu. The Lu solution corresponded to an implant ion current of 26 pA on a Keithley 6517B electrometer; this equates to an incident ion flux of $3.25 \times 10^8 \text{ }^{175}\text{Lu}^+$ ions $\text{s}^{-1} \text{ ppm}^{-1}$. The ICP-MS operating parameters are given in Table 1.

The ^{244}Pu diluted standard solution was aspirated through the ICP-MS for 5 h and 42 min with the quadrupole set to admit mass 238.05. The solution waste from the spray chamber was recycled back into the starting solution to allow the extended implant time as actual solution consumption rate is only $\approx 0.025 \text{ g min}^{-1}$. At the end of the implant period 0.5 ml of the solution remained.

2.2. Comparative samples

A stippled sample was created from the ^{241}Am gamma standard for energy resolution comparison to the MS-RAD samples. The standard was diluted in 3 M HNO_3 to 39.8 Bq mL^{-1} . The stippled sample was realized by aliquoting $\approx 5 \mu\text{L}$ of the solution to the center of a 15.9 mm diameter stainless steel disk and allowed to air-dry. The stipple was targeted to have approximately the same activity of ^{241}Am as the ICP-MS implants, $\approx 0.1 \text{ Bq}$.

For analysis of the mass-based separation capability of the MS-RAD method, an electrodeposited plutonium sample was created from the stock ^{244}Pu standard. A serial dilution of the $3 \mu\text{g g}^{-1} \text{ }^{244}\text{Pu}$ stock solution in 3 M HNO_3 was prepared, from which an aliquot (570.5 pg) of the dilution was transferred to a teflon vial. The aliquot was quantitatively transferred from the vial into a 50 mL glass beaker using three 1 mL aliquot rinses of Optima

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