Alpha Spectrometry Applications with Mass Separated Samples

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Abstract

²⁴¹Am 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 ²⁴¹Am samples using a small area passivated implanted planar silicon detector. We have also investigated the mass-based separation capability by developing a ²³⁸Pu sample, present as a minor constituent in a ²⁴⁴Pu standard, and performed subsequent radiometric counting. With this new sample development method, the ²⁴¹Am 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 ²³⁸Pu implant demonstrating the mass-based separation (or enhancement) attainable with this technique.

Keywords: alpha spectrometry, ICP-MS, actinide, Am-241

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. (Vajda et al., 1987) 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

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14 active area at the cost of geometrical (detection) efficiency. High-resolution al-
15 pha spectrometry involves the production of samples which are thin and free of
16 contaminants and a small-area PIPS detector that attains < 15 keV full-width-
17 at-half-maximum (FWHM) energy resolution.

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

33 2. Experimental Methods

34 2.1. ICP-MS Implants

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

47 Two ^{241}Am MS-RAD samples were developed for the high-resolution alpha
48 spectrometry derived from a commercial gamma standard solution with an ac-
49 tivity of $\sim 3.7 \text{ E4 Bq g}^{-1}$. The implanted samples were targeted to have an
50 activity of $\approx 0.1 \text{ Bq}$. The difference between the two samples was the potential
51 difference applied to the planchette and exit ion lens during implantation. For
52 the “low-bias” sample, the exit ion lens was held at 30 V and the planchette was
53 held at ground. In this configuration, the implanted ion current could be mon-
54 itored through a feedthrough via an external electrometer. For the “high-bias”
55 sample, the exit ion lens was held at ground potential and the implant was held
56 at -1500 V; the ion current could not be monitored since this potential differ-
57 ence 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 Molar 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 Material 996 (NIST, 2014)). The time-decayed isotopic atomic abundance (as of Sept. 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 ≈ 600 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 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 3.25×10^8 $^{175}\text{Lu}^+$ ions sec^{-1} 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 hrs 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 ≈ 0.025 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 Molar HNO_3 to 39.8 Bq mL^{-1} . The stippled sample was realized by aliquoting ≈ 5 μL 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, i.e., 0.1 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 $\mu\text{g g}^{-1}$ ^{244}Pu stock solution in 3 M HNO_3 was prepared, from which an aliquot (570.5 pg) of the dilution was transferred to

Table 1: A summary of the ICP-MS operating conditions used for the ^{241}Am and ^{238}Pu implants.

	^{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	$^{\circ}\text{C}$
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

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 grade 3 M HNO₃. Two mL of 0.36 M NaHSO₄ was added to the sample and subsequently dried on a hot plate. Following the drying procedure, 5 mL of Optima grade concentrated nitric acid was added and then the mixture was wet ashed (i.e., heated on a hot plate and concentrated hydrogen peroxide was added drop-wise) then brought to dryness again. The sample was transferred to the electrodeposition cell using 0.75 M H₂SO₄ (initially with 5 mL then three 2 mL rinses). Once the sample was pH adjusted, a platinum wire anode was inserted in the cell about 1 cm from the disc. The electrodeposition was performed for 1.5 hr. Subsequently, the cell was quenched using 1.5 M NH₄OH then rinsed with 0.1 M NH₄OH two times and deionized water three times. The cell was disassembled and the disc was rinsed with deionized water and set to air dry.

Table 2: The time-decayed isotopic atomic abundance [%] and alpha activity of the ²⁴⁴Pu standard used in this work as of Sept. 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 ²⁴⁴Pu standard.

Isotope	Atoms [%]	Alpha Activity [mBq]
²³⁴ U	0.00119 ± 0.002	1.56×10 ⁻³ ± 0.31×10 ⁻³
²³⁶ U	0.00245 ± 0.00002	3.35×10 ⁻⁵ ± 0.02×10 ⁻⁵
²³⁷ Np	0.00255 ± 0.002	3.78×10 ⁻⁴ ± 0.08×10 ⁻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 ± .008	0.374 ± 0.001

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116 2.3. Alpha Spectroscopy

117 The measurements of the alpha energy spectrum were performed in a Can-
118 berra Alpha Spectrometer-Model 7401. The 7401 was mainly used as a vacuum
119 chamber with vacuum gauge because control over analog shaping time and out-
120 put pulse shape was needed to obtain ideal energy resolution. Therefore, an
121 Ortec 142A charge-sensitive preamplifier was connected to a right angle adapter
122 which minimized the input cable length from the detector to preamplifier and
123 bypassed the internal electronics of the 7401. The output of the 142A was fed
124 to an Ortec 572A spectroscopy amplifier. The bipolar output of the 572A was
125 analyzed by an Amptek MCA-8000A Pocket Multichannel Analyzer (MCA) and
126 energy spectra were recorded by a laptop PC. The 7401 vacuum chamber was
127 evacuated by a dry scroll pump and achieved ultimate vacuum of a couple mtorr.

128 The ²⁴¹Am samples developed using the ICP-MS implant method achieved
129 the intrinsic energy resolution of the large-area silicon detector (20 keV FWHM)

130 shown in our previous results (Dion et al., 2014). A high-resolution spectrometer
 131 was assembled to further investigate if any differences between the two ICP-MS
 132 prepared samples (i.e., applied bias) affected the alpha spectroscopy energy
 133 resolution. The detector used for these measurements was a Planar Implanted
 134 Planar Silicon (i.e., PIPS[®] by Canberra) model PD25-10-500AM; 25 mm² ac-
 135 tive area, 500 μ m thick and 10 keV FWHM energy resolution at 5486 keV
 136 with 0.5 μ s shaping time. A comparison of the alpha energy spectrum of the
 137 high-bias MS-RAD sample is compared to the stipple sample in Figure 1 at a
 source-to-detector distance (SDD) of 4.7 mm. Compared to the MS-RAD sam-

Figure 1: Alpha energy spectra of the high-bias MS-RAD implant (blue solid line) and the
 stippled ²⁴¹Am sample (red dotted line). The spectra have been peak height normalized. It
 is apparent that the energy straggling imparted by the MS-RAD technique is minimal while
 the stipple sample shows significant broadening of the 5485.6 keV alpha of ²⁴¹Am most likely
 due to the carrier matrix of the stipple.

138 ple, the stipple sample peak has shifted 4 channels to a lower energy and was
 139 broadened by 3 keV FWHM. The peak widths of several radiometric measure-
 140 ments are summarized in Table 3. The high-bias MS-RAD sample was counted
 141 at SDDs of 3.2, 4.7, 7.2 and 11.2 mm and the spectra obtained did not exhibit
 142 any change in energy resolution suggesting the observed energy resolution is due
 143 to the intrinsic resolution of the detector. Within the variance of the measure-
 144 ments, both MS-RAD samples achieve the lower-limit energy resolution of the
 145 small area PIPS detector used in these measurements while the stippled sample
 146 exhibited broadened line width. There is a slight gain (6 %) in energy resolution
 147 measured in the low-bias sample in comparison to the high-bias sample that is
 148

149 statistically significant. This could be an implication that the ^{241}Am ions in the
 150 low-bias sample are not “buried” into the planchette as much as the high-bias
 151 sample.

152 We investigated the ion implant depth and the effect of peak width broad-
 153 ening by Monte-Carlo modeling. The experimental geometry was defined and
 154 included the detector dead layer, SDD, silicon active area, vacuum pressure and
 vacuum chamber dimensions. The simulations were performed with Geant4

Table 3: Statistical averages and standard deviation of the FWHM energy resolution of the low- and high-bias MS-RAD samples and the stippled sample of ^{241}Am .

Sample	FWHM [keV]
low-bias	9.69 ± 0.12
high-bias	10.31 ± 0.06
stipple	14.17 ± 0.04

155 (Agostinelli et al., 2003) and the ^{241}Am decay was handled using the Radioac-
 156 tive Decay Module. The source term was 1×10^6 decays of ^{241}Am of an isotropic
 157 point source aligned about the center axis of the detector (i.e., silicon). The
 158 total energy deposited per event in the silicon was stored in a ROOT (Brun and
 159 Rademakers, 1997) histogram. This histogram was Gaussian smeared to return
 160 the same FWHM energy resolution as measured with the low-bias sample. The
 161 ion implant depth from the high-bias sample was approximated by trial and
 162 error simulations by varying the thickness of a disc of stainless steel located
 163 0.02 mm above the source term. It was found that a thickness 75 Å of stainless
 164 steel above the isotropic point source reproduced the experimentally observed
 165 difference in energy resolution between the two MS-RAD samples.

166 The implant depth is an area of concern for MS-RAD. The radiometric
 167 counting of a particular isotope could be heavily influenced by the attenua-
 168 tion/straggling imposed by the implant method and would require further effi-
 169 ciency corrections. Biasing the implant was examined as a way to “steer” ions
 170 to a multi-implant target which would allow multiple implants from one input
 171 solution. Further studies have to be performed that quantify the depth of the
 172 implanted ion as a function of implant bias.

173 Creating alpha spectroscopy samples using a modified ICP-MS produced a
 174 clean, thin matrix unattainable with the traditional sample preparation meth-
 175 ods compared in this work. **However, it should be noted that other**
 176 **techniques have been developed that can attain thin sample matrices**
 177 **and FWHM energy resolution equal to what is reported in this**
 178 **work Becerril-Vilchis et al. (1996); Tsoupko-Sitnikov et al. (2000).**
 179 MS-RAD could prove a valuable tool as an alternative separation procedure for
 180 isotopes of plutonium that have alpha energy overlaps or separating isotopes
 181 that have similar chemical behavior.
 182

2.4. Mass-Based Separation of ^{238}Pu

A PIPS PD150-12-500AM (150 mm² active area, 12 keV FWHM energy resolution at 5486 keV and 500 μm thick) was used for the alpha spectroscopy measurements to compare the MS-RAD implant to the electrodeposited (ED) sample created from the ^{244}Pu standard. The larger detector was used for increased detection efficiency. Both samples were counted with a source-to-detector distance of 4.7 mm and to obtain counting statistics uncertainty < 5 %; the mass implant was counted for 12 days and the ED sample for 5 days.

The background subtracted spectra are shown in Figure 2. The data is

Figure 2: Alpha energy spectra of the MS-RAD implant (dotted red curve) and the ED sample (black curve). Due to the unresolvable separation of ^{238}Pu and ^{241}Am with room temperature silicon detectors, the ^{238}Pu alpha particle energy is convoluted in the ^{241}Am peak of the ED sample. See text for description of plot inlay. Background has been subtracted from both energy spectra.

shown as uncalibrated to eliminate any ambiguity that may occur due to the variances in sample construction that could lead to energy straggling, however, alpha particle energy has been confirmed with a certified three isotope alpha calibration source. It is clear from comparison of the two energy spectra that all other isotopes of plutonium (239, 240, 242 and 244) have been removed from the MS-RAD implant. This would suggest that mass 241 has also been removed (because of the mass window imposed by the plutonium species) and the only contribution to the energy spectrum could be mass 238 (^{238}Pu). Having prior knowledge of ICP-MS performance, the most likely mass to tail into the

²³⁸Pu implant would be mass 239. From Table 2, the activity ratio of ²³⁸Pu to ²³⁹Pu is 31:1. Therefore, if ²³⁹Pu had tailed into the mass 238 implant, ≈ 46 counts would be expected in the energy region of channel 2050–2120 in Figure 2. However, two counts are present in this energy region which indicates >20 times removal of mass 239 to 238.

The inset in Figure 2 is a linear y-axis plot focused on channels 2210–2270 of the ED and MS-RAD spectra. This is the alpha particle region of ²⁴¹Am and ²³⁸Pu. The ED sample (solid black curve) in this channel region has a dominant alpha particle peak **interfered by** a higher energy alpha particle. The dashed red curve is the MS-RAD mass 238 implant. To qualitatively confirm that the high-side asymmetric shape of the main alpha peak of the ED sample in this region of the inset is attributable to ²³⁸Pu in the ²⁴⁴Pu standard, the MS-RAD spectrum has been scaled based on the ratio of peak height of the lower energy alpha contribution of ²³⁸Pu (channel 2226) in the ED sample to the peak height in the MS-RAD implant (blue dash-dotted curve) and clearly the scaled MS-RAD implant overlaps this high-side asymmetry very well. Furthermore, the higher energy peaks beyond channel 2250 in the ED sample are alpha-conversion electron coincidences and higher energy low-yield alpha particles from the decay of ²⁴¹Am. Again, the omission of these peaks from the MS-RAD implant further confirms the removal of mass 241 (this is more visible in the log y-axis plot).

The level of signal enhancement (or separation) can be quantified if the activity of ²³⁸Pu in the ED sample can be deconvoluted from the ²⁴¹Am alpha distribution. This avoids the dependency on the SRM-996 data sheet and also corrects for any discrepancies during the electrodeposit sample preparation. The energy spectrum of the ED sample was analyzed using the Gold (Morháč and Matoušek, 2011; Gold, 1964) deconvolution algorithm implemented via ROOT's TSpectrum class. The deconvolution function takes a detector “response” and “source” spectrum and calculates the resulting deconvoluted spectrum. The response of the silicon detector was modeled as a convolution of a Gaussian with one exponential (Bortels and Collaers, 1987) as:

$$f(x) = \frac{A}{2 \cdot \tau} e^{\left(\frac{x-\mu}{\tau} + \frac{\sigma^2}{2\tau^2}\right)} \operatorname{erfc}\left(\frac{1}{\sqrt{2}}\left(\frac{x-\mu}{\sigma} + \frac{\sigma}{\tau}\right)\right). \quad (1)$$

The shape parameters are the width of the Gaussian σ , and τ to describe the exponential low-energy peak tailing, A is the peak area (number of counts) and μ is the peak position. The deconvolution algorithm returned the center location of each peak which was used as an initial guess. The constant “amplitude” term A was adjusted for each peak to roughly describe the energy spectrum, σ and τ were set to 2.00 (chn) and 3.08, respectively.

There are seven peaks of interest in the alpha spectrum between channels 1990–2270: two main alpha peaks for each isotope (5499.0 and 5456.3 keV (99.89 %) for ²³⁸Pu and 5485.6 and 5442.8 keV (97.9 %) for ²⁴¹Am), ²⁴¹Am has two peaks at higher energy that are low intensity alpha particles and coincident events (Siiskonen and Pöllänen, 2006) and a low-energy alpha particle at 5388 keV (1.66 %). A function to describe the seven peaks was defined based on

Equation 1 (i.e. $f(x)_1 + f(x)_2 + \dots$). The shape parameters of the function were fixed as previously described and the peak centers were initially set to the result of the Gold algorithm but were allowed to change. The function was fit to the energy spectrum and resulted in the dash-dotted (magenta) curve in Figure 3. The parameters from the final fit were back substituted into individual functions and are plotted in Figure 3 to show the contribution of each isotope; ^{241}Am peaks are solid red and ^{238}Pu peaks are dashed green.

The uncertainties stated in this analysis are the standard deviation uncertainties of the fitted peaks returned from MINUIT James (2014) implemented in ROOT. The ratio of activity based on the described fit analysis of ^{238}Pu and ^{241}Am determined by the area term in Equation 1 is 0.27 ± 0.04 and 0.73 ± 0.03 , respectively compared to 0.209 ± 0.043 and 0.791 ± 0.036 derived from Table 2. We find reasonable agreement when considering the assumptions given above and error propagation between the relative alpha activity found by deconvolution and peak fitting and the reported time-decayed values of the standard. Therefore, using the values found for total counts from the convoluted ^{238}Pu

Figure 3: The experimental alpha spectrum of the ED sample (gray filled histogram). The individual contributions from ^{241}Am (solid red curves) and ^{238}Pu (dashed green curve) are plotted along with the sum function (dash-dotted magenta curve).

and ^{241}Am alpha region from the ED sample, we can estimate the level of mass

238 enhancement (or removal) achieved on the ^{244}Pu standard used in this work. Based on count times, the estimated activity from the ^{238}Pu implant and the ED sample are given in Table 4. Therefore, the implant procedure used in this work has enhanced the ^{238}Pu signal from the ^{244}Pu isotopic standard by 159 ± 27 based on alpha activity.

Table 4: The estimated activity of the ED and ^{238}Pu implant developed in this work based on alpha spectroscopy and the peak fitting in Figure 3. These are used to estimate the level of separation achieved in this work. *The “other” contribution for the ^{238}Pu implant is the integral of the energy spectrum excluding the contribution of ^{238}Pu ; no other peaks are visible in the spectrum.

^{244}Pu ED:	
Isotope	Count Rate [s^{-1}]
^{241}Am	20.2 ± 0.3
^{238}Pu	7.6 ± 0.3
$^{239,240,242,244}\text{Pu}$	13.8 ± 0.2
^{238}Pu Implant:	
Isotope	Count Rate [s^{-1}]
^{238}Pu	1.33 ± 0.04
Other*	0.037 ± 0.006

264

265 3. Conclusions

266 A high-resolution alpha spectrometer was assembled to compare two MS-
 267 RAD ^{241}Am implants that were developed with different accelerating potentials
 268 and to compare to a conventional stipple source. There was a slight improvement
 269 in energy resolution achieved with the low-bias implant compared to the high-
 270 bias while a 32 % and 27 % improvement was found between the stipple and
 271 low- and high-bias implants, respectively. The difference in energy resolution be-
 272 tween the MS-RAD implants could be due to the influence of the implant bias
 273 on the actual depth that the ions are implanted into the planchette. This feature
 274 requires a more thorough investigation but preliminary simulations indicate a
 275 75 Å implant depth reproduces the energy resolution difference between the two
 276 implants. Each of the MS-RAD ICP-MS implanted ^{241}Am samples achieved the
 277 intrinsic energy resolution of the detector used in this work even at the closest
 278 SDD and no improvement was found by increasing the SDD. This achievement
 279 is likely due to the elimination of any carrier matrix in the sample develop-
 280 ment. The efficiency of the ICP-MS (0.02 %) limits the applications that can be
 281 targeted by this method but still allows for the preliminary quantitative steps
 282 necessary for this technique.

283 The mass-based separation and subsequent implant capability using a modi-
 284 fied ICP-MS for radioisotopes was tested with a NIST SRM-996 ^{244}Pu standard.

This test has shown the removal of all other plutonium (and americium) isotopes from a mass 238 (AMU) implant. This ^{238}Pu implant was analyzed via alpha spectroscopy and compared to an ED sample created from the unseparated standard. An unfolding fit technique was applied to the convoluted energy region of ^{241}Am and ^{238}Pu of the ED sample and it was shown that the MS-RAD method was able to enhance the ^{238}Pu signature of the ^{244}Pu standard by 159 ± 27 . Therefore, MS-RAD has the ability to either eliminate or reduce the amount of chemical separation necessary in samples that contain radioisotopes of the same (or different) elements which exhibit radiometric overlap.

Future work needs to develop a method of quantification starting from the front-end injected sample to radiometric counting. Reducing analytical timelines by the elimination of chemical separations requires improved efficiency of the implant technique. The efficiency (0.02 %) of the current instrument would only be applicable to original samples with high sample activity since most of the isotope of interest would not be implanted. This low efficiency reduces the activity of the implant, which increases the count time needed to reach statistical significance, and therefore lengthens the analysis timeline. PNNL is pursuing the purchase of a high-resolution multi-collector ICP-MS (with radioactive isotope capability) which will enable consecutive implants and an increase (10–50 times) in efficiency.

4. Acknowledgements

We wish to thank Janet Cloutier (PNNL) for her work as project manager, Dawn Lucas and May-Lin Thomas (PNNL) for developing the electrodeposited samples, Shannon Morley (PNNL) for developing the stipple samples and Charles Barinaga (PNNL) for useful discussions and insight. The research described in this paper was conducted under the Ultra-Sensitive Nuclear Measurements Initiative, a Laboratory Directed Research and Development Program at Pacific Northwest National Laboratory, a multi-program national laboratory operated by Battelle for the U.S. Department of Energy.

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