Alpha and Conversion Electron Spectroscopy of $^{238,239}\text{Pu}$ and ^{241}Am and Alpha-Conversion Electron Coincidence Measurements

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Abstract

A technique to determine the isotopic constituents of a mixed actinide sample has been proposed by a coincident alpha-conversion electron measurement. This presents a unique signature to allow the unfolding of isotopes that possess overlapping alpha particle energy and reduce backgrounds of an unseparated sample. The work presented here are results of conversion electron spectroscopy of ^{241}Am , ^{238}Pu and ^{239}Pu using a dual-stage peltier-cooled 25 mm² silicon drift detector and alpha spectroscopy with a passivated ion implanted planar silicon detector. The conversion electron spectra were evaluated from 20–55 keV based on fits to the dominant conversion electron emissions, which allowed the relative conversion electron emission intensities to be determined. These measurements provide crucial singles spectral information and calibration to aid in the coincident measurement approach. Furthermore, an alpha-conversion electron spectrometer was assembled using the silicon based detectors described and results of a coincident spectrum analysis is reported for ^{241}Am .

Keywords: conversion electron, alpha spectrometry, silicon drift detector, charged-particle coincidence detection.

1. Introduction

Silicon-based alpha spectrometry has intrinsic advantages of relative low-cost, very low backgrounds ($\approx 10^{-5}$ counts per second), provides detection limits of ≈ 1 mBq **for MeV alpha emitting nuclides** and near 100 % intrinsic detection efficiency. This is advantageous for the detection of actinide radioisotopes because most actinides have high alpha decay branching ratios. The energy resolution of room temperature ion implanted silicon detectors are dependent on thickness and area. For detectors that are commercially available and routinely

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9 used for alpha spectroscopy this ranges from 10–22 keV FWHM. These char-
 10 acteristics make alpha spectrometry a very popular radioanalytical method for
 11 isotopic identification of actinide samples. The method has downfalls including
 12 unresolvable alpha particle energies for nuclides of interest to the safeguards
 13 and forensics community, and chemistry procedures for sample preparation. As
 14 an example of the energy interference, the main alpha particles emitted from
 15 $^{239}\text{Pu}/^{240}\text{Pu}$ and $^{238}\text{Pu}/^{241}\text{Am}$ are separated by 11.6 keV and 13.4 keV [1], re-
 16 spectively. Hence, if a sample contains ^{239}Pu and ^{240}Pu , alpha spectrometry
 17 may be used in **combination** with mass spectrometry to determine the indi-
 18 vidual isotopics of the sample. Similarly, chemical processes can be performed
 19 to separate the plutonium from the americium and the individual samples can
 20 be analyzed with alpha spectrometry since mass spectrometry has difficulties
 21 with ^{238}Pu because of natural uranium background.

22 In some circumstances, isotopic information on a sample in a reduced time
 23 frame would be highly valued. We are investigating a coincident detection tech-
 24 nique that could perform quantitative measurements of a mixed isotopic sample
 25 with minimal sample preparation chemistry. The basis for the measurement is
 26 time coincident detection of an alpha particle and conversion (or Auger) elec-
 27 tron. Preliminary theory of the measurement and simulations with a general
 28 qualitative comparison of nuclear detector options has been summarized in Ref.
 29 [2]. The majority of alpha-decaying nuclides populate either the ground state
 30 or a low excited level of the daughter nucleus. These low energy levels inher-
 31 ently have large internal conversion coefficients (ICCs) which dominate gamma
 32 emission. There has been work on the applicability of alpha-gamma (photon)
 33 coincident detection [3, 4] but this requires two different types of detectors; one
 34 for high alpha detection efficiency and one for high gamma efficiency. Also, the
 35 photon yield of many actinides is quite low (^{241}Am is one exception) so the
 36 overall counting time of a system to achieve acceptable statistics of this type
 37 may be quite large. Conversely, low-energy (20–50 keV) electrons interact in
 38 matter much the same as alpha particles (over certain energy windows) and
 39 hence both particles could be detected with one segmented detector or two de-
 40 tectors of the same type. A large unknown is the low-energy spectrum of the
 41 actinides of interest. Conversion electron yields, i.e., ICCs, are mainly based on
 42 theoretical calculations which are dependent on the measured photon emission
 43 [5]. Therefore, a measurement of the low-energy spectrum of these isotopes is a
 44 crucial step in understanding the proposed coincident measurement.

45 There have been investigations into the use of conversion electron spec-
 46 troscopy for the isotopic analysis of actinides [6, 7, 8, 9]. Refs. [6, 7, 8] explored
 47 the use of Si(Li) windowless, cryogenically cooled detectors to perform isotopic
 48 analysis of mixed plutonium samples and curium isotope samples purely based
 49 on conversion electron spectroscopy. Other groups have explored tiling small
 50 area Si-PIN diodes for the detection of conversion electrons from radioxenon
 51 isotopes [10]. Recent work has also explored the use of silicon drift detector
 52 (SDD) technology for conversion electron measurements [9], which focused on
 53 the feasibility of conversion electron spectroscopy particularly for the ratio anal-
 54 ysis of $^{240}\text{Pu}/^{239}\text{Pu}$, a similar concept as Ref. [6].

55 This work reports conversion electron and alpha spectroscopy of ^{241}Am ,
56 ^{238}Pu and ^{239}Pu and also coincident alpha-conversion electron measurements
57 of ^{241}Am . Alpha spectroscopy was performed for source confirmation and
58 performed with a passivated implanted planar silicon detector, i.e., Canberra
59 PIPS[®]. The pulse-reset functionality of the SDD preamplifier eliminated the
60 possibility of alpha particle measurements with the off-the-shelf electronics,
61 therefore, the SDD was only used for low-energy measurements. The conver-
62 sion electron spectra were analyzed based on the relative peak area after an un-
63 folding technique of each conversion electron peak and compared to theoretical
64 and experimental yields. This comparison provides a thorough understanding of
65 the experimental response and yield of the isotopes to be used in the proposed
66 coincident measurement and calibration of each detector. The coincident mea-
67 surement was made using the combination of the PIPS and SDD detector. The
68 detection efficiency of this setup was not optimal since a detection could only
69 be made with an alpha particle measured in the PIPS detector and a conversion
70 electron in the SDD.

71 2. Apparatus

72 Alpha and conversion electron energy spectra were recorded for ^{241}Am (29.59
73 ± 0.95 Bq), ^{238}Pu (40.52 ± 1.30 Bq) and ^{239}Pu (43.25 ± 1.34 Bq). The
74 commercial electrodeposited sources were diffusion bonded oxide and had an
75 active diameter of 5 mm with a platinum backing.

76 2.1. Alpha Measurements

77 Alpha spectroscopy was performed with a partially depleted silicon PIPS de-
78 tector (PD150-12-500AM) in a Canberra alpha spectrometer model 7401 which
79 was evacuated to ≈ 5 mTorr with a dry scroll vacuum pump. The PIPS detec-
80 tor had an active area of 150 mm^2 , was $500\text{ }\mu\text{m}$ thick and can achieve 12 keV
81 FWHM at 5486 keV. The 7401 is a double-wide NIM module that provides
82 rough vacuum measurements, ease of changing between different PIPS detec-
83 tors, high-voltage application and analog pulse processing optimized for alpha
84 spectrometry. However, the internal electronic processing and high voltage of the
85 7401 were bypassed to allow a user-specified charge sensitive preamplifier and
86 analog shaping electronics. Therefore, an Ortec 142A charge sensitive preampli-
87 fier followed by an Ortec 572 shaping amplifier with a shaping time of $3\text{ }\mu\text{s}$ was
88 used for analog pulse shaping while an Amptek PocketMCA provided ADC and
89 energy spectrum data. The source to detector distance for the measurements
90 was ≈ 13 mm with count times for ^{241}Am , ^{238}Pu and ^{239}Pu of 41 h. The detec-
91 tor and electronics were energy calibrated with a 4 Bq electrodeposited alpha
92 calibration source (Eckert and Ziegler Multi-nuclide alpha calibration source)
93 before each measurement.

94 2.2. Conversion Electron Measurements

95 The idea of a semiconductor drift detector was first proposed by Emilio Gatti
96 and Pavel Rehak [11]. The novel concept was based on decoupling the charge

carrier electric field from the depletion field; hence, a thin crystal with large active area could be depleted by a small area contact. This leads to a large reduction in detector input capacitance and therefore lends itself to low-energy x-ray spectroscopy. This is in fact the basis of recent SDDs; a small charge collection contact coupled with FET and detector peltier cooling and a charge-reset style preamplifier further reduce leakage current. All these aspects of construction reduce the electronic noise contributions of the system significantly allowing full-width at half-maximum (FWHM) measurements of ^{55}Fe x-rays of 125 eV at 5.9 keV.

An Amptek model AXR detector, PA-230 preamplifier and DP5/PC5 was acquired with a silicon nitride (“C2”) window. Preliminary measurements and energy calibrations (e.g., spectrum in Figure 1) were performed with the window in place. However, for the low-energy conversion electron spectra the window was removed. The SDD had a 25 mm² active area, was 500 μm thick detector and the field-effect transistor (FET) and SDD are cooled by a dual-stage peltier. The detector entrance junction is implanted boron through an oxide layer into the silicon. The boron creates the electrical contact and the oxide layer is 300–400 Å thick. There is an estimated incomplete charge collection region created by the contact of ≈ 1500 Å. The SDD was mounted in a conflat vacuum chamber. A custom vacuum feedthrough was manufactured on a 3/4-16 UNF-2A threaded face seal fitting (PAVE Technology Co.) and allowed the 10-pin FJ-style interconnect wire between the SDD and electronics. This feedthrough was mounted on a machined 2.75” conflat blank flange. The chamber was evacuated to 10^{-6} torr with a turbo pump system to eliminate the possibility of condensation on the detector through subsequent cooling in vacuum. It is critical to attain reasonable vacuum levels to prevent condensation on the entrance window of the SDD before the cooling the detector. To reduce external particulate contamination, a 10 μm filter was installed on the vent line of the chamber.

The system was calibrated with ^{55}Fe (5.9 and 6.5 keV), ^{109}Cd (22 and 24.9 keV) and ^{133}Ba (30.625, 30.973, 34.92, 34.987 and 35.81 keV) photon sources. An energy spectrum of ^{55}Fe at a temperature of 234 K is shown in Figure 1. The FWHM energy resolution of the SDD at 234 K is 141 eV at 5.9 keV. The peaks at 5.9 and 6.5 keV are x-rays from the ^{55}Fe decay, the peak at 4.16 keV is the silicon escape (5.9 keV - 1.74 keV) and the double pile-up x-ray peaks are also present. Some of the pulse processing parameters of the DP5 for the results presented in this work are summarized in Table 1.

3. Measurements and Results

3.1. Alpha Spectroscopy

Alpha spectra of the three sources are shown in Figure 2. The line shapes of the alpha peaks are noticeably asymmetric as expected [12]. The low-energy tailing is due to the distribution of electronic and non-electronic energy losses from the alpha particle in the silicon medium. Analytical functions used to describe alpha peaks are a linear combination of exponential functions (to describe

Figure 1: Energy spectrum of ^{55}Fe taken at 234 K with the SDD equipped with the silicon nitride window. The FWHM energy resolution is 141 eV for the 5.9 keV peak.

Table 1: A summary of the DP5 parameters, including trapezoidal trigger parameters and detector bias.

	Setting
Sampling	20 Mhz
Peaking Time	9.600 μs
Flat Top	0.200 μs
Fast Peaking	400 ns
Detector Bias	-105 V

140 the tailing) and a Gaussian [13]. There appears to be ^{238}Pu and/or ^{241}Am con-
 141 tamination in the ^{239}Pu source. This contribution is visible in the ^{239}Pu energy
 142 spectrum (dotted red curve) as two peaks, the main peak being at ≈ 5500 keV.
 143 The additional peak in the ^{239}Pu spectrum centered at 5306.2 keV is ^{210}Po con-
 144 tamination, which is a ^{214}Rn daughter. Also, there appears to be ^{239}Pu and/or
 145 ^{240}Pu present in the ^{238}Pu source (solid blue curve). This contamination is the
 146 peak centered at ≈ 5150 keV in the ^{238}Pu spectrum. There are other alpha emit-
 147 ting isotopes that could also contribute to source contamination in these energy
 148 regions. However, the nuclides that would lie in the ^{241}Am energy region (e.g.,
 149 $^{222,223,224}\text{Rn}$ or ^{228}Th) have half-lives on the order of days eliminating them as
 150 candidates and in the $^{239,240}\text{Pu}$ region there is no direct energy overlap except
 151 for ^{208}Po which is a very unlikely candidate. Table 2 contains the alpha particle
 152 emissions, decay mode/s and intensity of the nuclides discussed.

Table 2: Decay mode, branching ratio and alpha emission for the **nuclides** studied in this work. Only alpha particles with intensity >1 % are listed. Data is taken from [14].

Isotope	Decay mode [Branching %]	Alpha [keV]	Intensity [%]
^{238}Pu	α [100]	5499.0	70.91
		5456.3	28.98
^{239}Pu	α [100]	5156.6	70.77
		5144.3	17.11
		5105.5	11.94
^{240}Pu	α [100]	5168.2	72.80
		5123.7	27.10
^{241}Am	α [100]	5486.6	84.8
		5442.8	13.1
		5388	1.66

Low-resolution alpha **spectrometry** with commercial sources cannot fully resolve the alpha particles of ^{239}Pu from ^{240}Pu or ^{241}Am from ^{238}Pu . Deconvolution techniques can be employed for this task and obtain ratio uncertainties [15] of $\leq 10\%$ when ample counting statistics are allowed, high-resolution spectrometers are used and a thin, “clean” source is constructed. However, for convenience we report and compare the relative contamination of the plutonium sources based on the ratios of region of interest (ROI) as summarized in Table 3. **Two ROIs were determined for each plutonium spectrum.. First, the dominant alpha peak for each region of interest was fit with a constrained Gaussian to determine the centroid and sigma (σ). For example, three peaks for the ^{238}Pu curve in Figure 2 were fit; the dominant ^{238}Pu peak at 5499 keV, the next major alpha emission of ^{238}Pu at ≈ 5450 keV and the peak present at ≈ 5150 keV (the minor contaminant). ROI₁ contained the counts from the dominant isotope (i.e., ^{238}Pu in the ^{238}Pu spectrum and $^{239}\text{Pu} + ^{240}\text{Pu}$ in the ^{239}Pu spectrum) and ROI₂ contained the counts from the minor isotope (i.e., $^{239}\text{Pu} + ^{240}\text{Pu}$ in the ^{238}Pu spectrum and ^{238}Pu in the ^{239}Pu spectrum). ROI₁ was defined as the centroid of the main alpha peak plus half of its FWHM to the centroid of the lower energy second alpha peak minus five sigma. ROI₂ ranged from the centroid of the minor alpha peak plus half of its FWHM then was set to be of equal width as ROI₁. The spectrum was then integrated over each ROI and the ratio (ROI₂/ROI₁) was calculated.**

3.2. Conversion Electron Spectroscopy

The SDD is sensitive to alpha particles, photons (x-ray and γ -ray) and the conversion and Auger electrons emitted from the nuclides investigated. However,

Figure 2: Alpha energy spectra of the three electrodeposited sources investigated in this work as labeled in the legend. The isotopic contamination of the two plutonium sources is visible and quantified in Table 3.

for the conversion electron measurements, the dynamic range of the SDD is 1–55 keV (i.e., it is not capable of detecting the alpha particles). It should be noted that the coincident measurement approach being explored is primarily focused on the energy region from 20–55 keV, where the majority of conversion electrons reside and “ignores” the low-energy x-ray (13–14 keV) region.

For the SDD measurements presented in this work, the source-to-detector distance was 5 mm and the count times were arbitrary but a requirement to achieve at least 100 counts in a low-yield conversion electron peak of each isotope was set. The count times were 21.4 h for ^{241}Am and ^{239}Pu and 11.7 hours for ^{238}Pu . A background spectrum was acquired for 6.6 h, scaled and subtracted from each spectrum, respectively. The MCA energy spectra were analyzed in ROOT [16] and are shown in Figure 3 as solid curves. Furthermore, a continuum subtraction algorithm was applied to each spectrum [17]; the continuum are the dotted (red) curves in Figure 3.

The response of the SDD to photons is Gaussian as seen in Figure 3 for the $\text{La}1$ x-ray line of all nuclides. The detector response to conversion electrons contains low-energy tailing. The response function of silicon detectors to electrons has been studied previously, for example [18, 12]. The response is defined by an *absorption peak* and an *escape component*; where in the energy range measured

Table 3: A comparison of the relative isotopic composition of the plutonium sources investigated in this work based on the experimental alpha measurements. **See text for a description of the ROI definitions.**

Source	ROI ₁ [keV]	(Nuclide) [counts]	ROI ₂ [keV]	(Nuclide/s) [counts]	ROI ₂ /ROI ₁
²³⁸ Pu	5426–5505	(²³⁸ Pu) 271408	5084–5162	(²³⁹ Pu+ ²⁴⁰ Pu) 819	0.0030 ± 0.0001
²³⁹ Pu	5077–5163	(²³⁹ Pu) 449514	5421–5506	(²³⁸ Pu+ ²⁴¹ Am) 3175	0.0071 ± 0.0001

Figure 3: Energy spectra using the SDD for the **nuclides** investigated in this work. The continuum is shown as the dotted (red) curve in each spectrum. The dominant peaks of interest are labeled for each spectrum; photons are shown as daughter isotope and shell while the conversion electron peaks are labeled with the energy level of the daughter and subsequent shell of emission.

in this work the escape component would be due to electron backscattering, energy straggling in the source construction and energy loss in the silicon dead layer [19, 20].

The conversion electron peaks in this work were fit using an exponential to describe the low-energy tailing and a Gaussian to describe the absorption portion of the peaks as [12],

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

Here A is the peak area, μ is the centroid of the peak, σ is the width of the Gaussian and τ describes the exponential tail. A function was defined based on Eq. 1 for each energy spectrum (isotope) to include a fit to each conversion electron peak of significant statistics, the pair of low-energy x-rays and a constant term to describe the baseline of each spectrum. The Minuit2 [21] library and Migrad algorithm were used for error minimization through a least-squares fit in ROOT. A section of the fit result is shown in Figure 4 for ^{238}Pu . The exponential function describes the tailing reasonably well and allows the area of each conversion electron peak to be extracted if there is peak overlap. The FWHM energy resolution of the SDD for the x-rays of the nuclides found from the fitting routine are given in Table 4. The calculated values for energy and conversion electron subshell ratios from the fitting routine are summarized in Table 5. The uncertainties reported contain full correlation covariance matrix calculations and are propagated errors of the uncertainty returned by Minuit on the peak area parameter of Eq. 1; uncertainties imposed by the background subtraction algorithm are ignored because of the minimal influence in the energy region of interest. The measured subshell ratios found in this work are

Figure 4: A portion of the ^{238}Pu energy spectrum showing the fit to the data. The total fit function is the solid (blue) curve, the L-shell conversion electron individual fits are dashed-dotted (black) and the low-energy x-rays are dotted (magenta). The x-ray peaks are fit with a Gaussian and the conversion electron peaks are fit with the function given in Eq. 1.

compared to BrICC [5] and experimental measurements when available. The

Table 4: The energy **resolution** of the SDD for **x-ray signatures of the nuclides investigated and** measurements performed in this work. The detector performs consistently across all three nuclides; an indication of similar source construction and experimental systematic effects. The energy reported in this table is from the fit to the data and is an indication of the level of accuracy of the energy calibration (the published values are given in parenthesis [1]) used to convert the x-axis of the energy spectra. The statistical uncertainty in the energy calibration is 10 eV.

Isotope	Energy [keV] ([1])	FWHM [eV]
^{241}Am	13.961 (13.946)	224 ± 3
	17.770 (17.751)	252 ± 4
^{239}Pu	13.632 (13.618)	230 ± 5
	17.263 (17.222)	234 ± 6
^{238}Pu	13.635 (13.618)	222 ± 4
	17.250 (17.222)	205 ± 4

222 results for ^{241}Am 59.5-L1,L2 and L3 subshells correlate well with the measure-
 223 ments of [22] but deviate from BrICC significantly. The 59.5-M1 and M2 are
 224 not consistent with [22] or BrICC while [22] and BrICC agree. ^{239}Pu is only
 225 compared to BrICC since previous experimental measurements of the subshell
 226 coefficients could not be found. Overall, the results of this work are consistent
 227 with BrICC; the largest outlier is the 38.7-L2/38.7-L3 ratio and this could be
 228 due to the 38.7-L2 conversion electron being heavily convoluted with uranium x-
 229 rays in this energy region. The results of the L-subshells of ^{238}Pu are interesting
 230 as good agreement is found between this work with [23] and BrICC for the 43.5-
 231 L2/43.5-L3 ratio but obviously the 43.5-L1 emission has been **overestimated**
 232 **in this work** by $\approx$ a factor of 2. This is unexpected as the spectrum and fit
 233 results shown in Figure 4 would not imply crude over or under estimations of
 234 either the 43.5-L1 or the 43.5-L2,L3 yields.

235 The ^{239}Pu spectrum has several interesting features. The spectrum has a low-
 236 energy (<20 keV) continuum that is most likely due to the β -decay of ^{241}Pu
 237 (Q-value 20.8 keV) present in the source. Also, there is evidence of ^{240}Pu activity
 238 that could not be observed from the alpha spectroscopy shown in Figure 2 due to
 239 insufficient energy resolution. The ^{240}Pu 45.2 keV L2 and L3 conversion electrons
 240 have been labeled in Figure 3 and were fit for analysis. The 45.2 keV M2 and M3
 241 conversion electrons are also visible at 40 keV but were not used for analysis due
 242 to low statistics. The fractional amount of ^{240}Pu present in the ^{239}Pu source was
 243 estimated using the ICCs calculated by the BrICC database for the 45.2 keV L2,
 244 3 shell of ^{236}U , 225 and 197, respectively and the peak area returned from the fit
 245 to the spectrum. The ICCs can be used to calculate the probability of conversion
 246 electron emission per decay: 45.2 keV L2 and L3 for ^{240}Pu , 10.06 and 8.81 %,
 247 respectively, and the 51.6 keV L2 and L3 for ^{239}Pu , 3.35 and 2.83 %, respectively.
 248 Using this information and ratios of the peak areas from the fit to the data

Table 5: A summary of the parameters found from deconvolution using the analytical function given in Eq. 1 to the data given in Figure 3. The $\sum$ all is the sum of all measured subshell emission for the particular isotope and energy level. For example, $\sum$ all for ^{239}Pu -51.6 = (51.6-L2 + 51.6-L3 + 51.6-M2 + 51.6-M3 + 51.6-Ntot). *The author was not able to find any experimental data on subshell conversion coefficients for ^{235}U .

Shell	Energy [keV]	Subshell Ratio	Exp. this work	Exp. others	Theory BrICC(FO)
^{241}Am					
59.5-L1	37.118 ± 0.009	L1/ $\sum$ all	0.284 ± 0.012	0.235 ± 0.066 [22]	0.304
59.5-L2	37.892 ± 0.010	L2/ $\sum$ all	0.436 ± 0.013	0.458 ± 0.129 [22]	0.281
59.5-L3	41.886 ± 0.011	L3/ $\sum$ all	0.114 ± 0.004	0.128 ± 0.036 [22]	0.283
59.5-M1,2	54.064 ± 0.018	M1,2/ $\sum$ all	0.0287 ± 0.0047	0.178 ± 0.050 [22]	0.132
^{239}Pu					
38.7-L2	17.490 ± 0.029	L2/L3	1.30 ± 0.01	-*	1.16
38.7-L3	21.520 ± 0.012	-	-	-*	-
51.6-L2	30.700 ± 0.006	L2/ $\sum$ all	0.407 ± 0.010	-*	0.402
51.6-L3	34.437 ± 0.009	L3/ $\sum$ all	0.333 ± 0.009	-*	0.340
51.6-M2	46.426 ± 0.017	M2/ $\sum$ all	0.115 ± 0.006	-*	0.107
51.6-M3	47.643 ± 0.037	M3/ $\sum$ all	0.0873 ± 0.0051	-*	0.095
51.6-Ntot	50.460 ± 0.027	Ntot/ $\sum$ all	0.0544 ± 0.0032	-*	0.057
^{238}Pu					
43.5-L1	20.282 ± 0.032	L1/L2	0.0739 ± 0.0057	0.0345 ± 0.0019 [23]	0.0341
43.5-L2	22.549 ± 0.004	L1/L3	0.0935 ± 0.0058	0.0399 ± 0.0022 [23]	0.0385
43.5-L3	26.327 ± 0.004	L2/L3	1.27 ± 0.03	1.147 ± 0.020 [23]	1.13
43.5-M2	38.354 ± 0.010	M2/M3	1.37 ± 0.07	1.04 ± 0.05 [23]	1.08
43.5-M3	39.167 ± 0.016	M3/ $\sum$ all	0.0871 ± 0.0035	-	0.0960
43.5-Ntot	42.266 ± 0.020	Ntot/ $\sum$ all	0.0458 ± 0.0018	-	0.0557

249 suggests an activity ratio of $^{239}\text{Pu}/^{240}\text{Pu}$ of 16.14 ± 0.81 and 14.40 ± 0.78 for
250 the L2 and L3 conversion electron emission, respectively and an average of 15.27
251 ± 1.1 . Furthermore, using the information derived by alpha spectroscopy in
252 Table 3, the activity of the ^{239}Pu source (43.25 Bq) attributable to ^{239}Pu , ^{240}Pu
253 and $^{241}\text{Am}+^{238}\text{Pu}$ as: 40.13 ± 1.6 , 2.81 ± 0.23 , 0.30 ± 0.01 Bq, respectively.
254 The dominant conversion electrons for ^{238}Pu and ^{241}Am are not overlapped in
255 energy with ^{239}Pu or ^{240}Pu and have similar emission probabilities as ^{240}Pu as
256 shown in Table 6. Considering the data presented here and if we assume the
257 detection efficiency and background over the entire conversion electron energy
258 range constant, one can infer that the lower limit of detection of ^{238}Pu and
259 ^{241}Am is greater than 0.30 Bq and below 2.81 Bq.

260 3.3. Alpha-Conversion Electron Coincidence

261 Using the described detectors an alpha-conversion electron coincidence spec-
262 trometer has been assembled and tested. A measurement of ^{241}Am has been per-
263 formed and we briefly report the results here. The detectors were assembled in a
264 conflat vacuum chamber and angled in reference to the source to increase the ge-

Table 6: The energies and emission probabilities of the L2 and L3 conversion electrons of ^{241}Am and ^{238}Pu as calculated from BrICC.

Shell	Energy [keV]	Prob. per decay [%]
^{241}Am		
59.5-L2	37.94	9.44
59.5-L3	41.93	9.50
^{238}Pu		
43.5-L2	22.55	10.62
43.5-L3	26.33	9.41

ometrical efficiency. A schematic of the detector mounting is shown in Figure 5. The PIPS detector was used for alpha particle detection and the SDD was only sensitive to the low-energy spectrum. The electronic signals were recorded by an XIA Pixie-4 [24] and coincidence analysis was performed offline. The ^{241}Am source used in this measurement had an activity of ≈ 3700 Bq; a different source than used above with the SDD and for clarity is referred to as $^{241}\text{Am-a}$. The assembled apparatus shown in Figure 5 was installed into a 6" conflat chamber. The aluminum base ([4] in the image) was constructed of two pieces; the top part secured the detectors and the bottom half had a shelf for the source. The bottom half of the base was in contact with the interior wall of the conflat chamber and provided means of a heat sink for the SDD peltier by application of vacuum compatible thermal conductive grease (Nusil CV-9042). The chamber was evacuated by a turbo pump assembly (i.e., Pfeiffer HiCube Eco) and attained $\approx 5 \times 10^{-6}$ torr. The coincident results using the alpha-conversion elec-

Figure 5: A sketch of the coincident apparatus using the PIPS and SDD detector previously described. The items identified are: [1] PIPS detector, [2] SDD and preamplifier board, [3] ^{241}Am source position, [4] aluminum base/heat sink, [a] alpha particle trajectory into PIPS and [b] low-energy particle trajectory into SDD.

279 tron spectrometer with the ^{241}Am -a source are shown in Figure 6. The results
 280 here are based on ≈ 6900 coincident events out of a dataset of 5.95×10^5 total
 281 events. The coincident events had a normally distributed time difference centered
 282 at $\approx 0.5 \mu\text{s}$ with a width of $0.3 \mu\text{s}$. Energy spectra of the coincident events
 in the PIPS (a) and SDD (b) are shown in Figure 7. The angled configuration

Figure 6: A plot of the coincident events measured with the alpha-conversion electron spectrometer for ^{241}Am . The events are shown as the low-energy events measured with the SDD versus the alpha particles measured with the PIPS detector. Both energy spectra have been re-binned to highlight the coincident lines of interest.

283 of the detectors has increased the line width of the detector response due to
 284 (most likely) increased energy straggling in the source itself and the dead layer
 285 of the silicon detector/s. The most intense coincident peak above 15 keV is the
 286 contribution of the 59.5 keV-L1,L2 conversion electron of ^{241}Am with energy of
 287 37.1 keV (10 % emission probability) and 37.9 keV (9.4 % emission probability),
 288 respectively. The contributions of the different conversion electrons to the
 289 coincident events are summarized in Table 7. Therefore, the alpha-conversion
 290 electron coincident spectrometer assembled in this work with conversion electron
 291 energies in the range of 19–59 keV will be sensitive to roughly 40 % of the
 292 coincident events of ^{241}Am .
 293

294 4. Conclusions

295 We have investigated three electrodeposited sources of ^{241}Am , ^{238}Pu and
 296 ^{239}Pu using low-resolution alpha spectrometry with a PIPS detector and also

Figure 7: Coincident gated energy spectrum of the PIPS (a) and SDD (b) detector in response to the ^{241}Am -a source. Each spectrum contains ≈ 6900 coincident events.

their low-energy x-ray and conversion electron spectrum using a windowless SDD. The low-energy measurements and the relative strengths of conversion electron contributions have been quantitatively evaluated by fitting the conversion electron peaks with an exponential (to describe the tailing) and a Gaussian. These measurements are vital in the understanding of the coincident technique being explored for the isotopic analysis of mixed samples with minimal sample preparation. The conversion electron yield has been compared to BrICC and experimental data when available. The largest discrepancy between the measured and BrICC fractional ICCs occur for ^{241}Am in all measured subshells. This has been previously recognized by the work of Gorozhankin and Bé [25]. It is understood that the largest disagreement between theoretical and measured ICC values occur in electric dipole (E1) transitions; which is in fact the transition type of the 59.5 keV level of ^{241}Am . This example justifies the need for experimental ICC measurements. In contrast, the 59.5-L shells measured in this work correlate well with previous experimental measurements reported in [22], which exemplifies the discrepancies present in BrICC.

As noted in previous work [8, 9, 26], the low-energy conversion electron spectra of the actinides of interest to the safeguards and forensics community have unique intrinsic structure. We have shown that ^{240}Pu was detected and quantified in the ^{239}Pu conversion electron measurement with clear peak separation as opposed to the highly convoluted response seen in the alpha spectrum. There-

Table 7: Contributions to the coincident events by integration of the SDD spectrum (b) in Figure 7.

Shell/s	Energy Range [keV]	Coincident events [%]
59.5-L1,L2+43.3-M1-3	35-39	14.5
33.2-M1-3,N1+43.4-L3	22-30	10.3
26.3-M1-3+43.4-L1,L2	19-22	8.1
59.5-M1-3	52-56	4.5
59.5-L3	40-43	3.1
59.5-Ntot	57-59	1.0

fore, an alpha-conversion electron coincident measurement implies a stronger separation capability while also allowing reduced detector energy resolution.

Furthermore, using the two silicon detectors evaluated and calibrated in this work an alpha-conversion electron coincident spectrometer was constructed and provides results of this measurement for ^{241}Am . We have demonstrated the feasibility of a coincident measurement and have described the signatures of interest for ^{241}Am . In comparison to alpha-gamma coincident techniques that require two different detectors, the alpha-conversion electron measurement can be made in the same medium under similar conditions. **This fact would be advantageous in a field system where cooling and power requirements are logistical issues.** We propose to demonstrate this ability with a segmented PIPS detector with single nuclides and mixed samples.

Acknowledgment

The authors are grateful for the drafting work of the alpha-conversion electron spectrometer provided by Kenna Middleton during an internship at PNNL. 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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