Room-temperature electron spectroscopy of ^{239}Pu and ^{240}Pu

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Abstract

Passivated, implanted, planar silicon (PIPS) detectors have been used for the measurement of electron spectra. The commercially available PIPS detectors, available in thicknesses of 100 μm , 300 μm , and 500 μm , have an energy resolution (FWHM) of ~ 2.2 keV, which is essentially the same as that of PIN diodes. Alpha and electron spectra of mass-separated ^{239}Pu and ^{240}Pu sources have been measured with a 300- μm thick PIPS detector and the electron to alpha ratios for the conversion lines of the 51.62- and 45.24-keV transitions have been determined. A procedure has been developed to determine the amount of ^{239}Pu and ^{240}Pu in a mixed source. The α -particle emission rate of the mixed source is measured, which is the sum of individual rates. From the electron spectrum of the mixed source, measured with the same set up as the alpha spectrum, the rates of ^{239}Pu electron lines are determined. Using the electron rate of the ^{239}Pu line and the electron to alpha ratio measured for the pure source, the α -particle emission rate of ^{239}Pu is determined. The difference from the total α -particle emission rate gives the α -particle emission rate of ^{240}Pu . In addition, electron intensities and conversion coefficients of the ^{239}Pu and ^{240}Pu transitions have been measured.

Keywords: measured; electron intensities; conversion coefficients; decay of ^{239}Pu and ^{240}Pu

1. Introduction

Electron spectroscopy is a powerful tool for the measurement of conversion coefficients and it can also be used, like x rays, to determine the elemental identity and the quantity of a substance. However, compared to γ -ray spectroscopy, it is somewhat inconvenient to use because 1) it requires a thin source and 2) the measurement must be carried out in vacuum. Lithium-drifted silicon detectors, cooled with liquid nitrogen, have been used for electron spectroscopy. Typically, 3-mm thick detectors with an area of $\sim 1 \text{ cm}^2$ have been used which have a resolution [full width at half maximum (FWHM)] comparable to that of germanium detectors ($\sim 1.0 \text{ keV}$ for a 100-keV electron line) [1]. With the availability of high-purity silicon, PIN diodes have lately been used at room temperature for electron measurements. These detectors, which have a typical thickness of $300 \mu\text{m}$, give a resolution (FWHM) of $\sim 2.4 \text{ keV}$ at 100-keV electron energy [2, 3]. By cooling the PIN diodes with liquid nitrogen, the resolution (FWHM) was improved to 1.1 keV [4]. These investigators also measured the energy loss in the detector window, which was very low, indicating a very thin entrance window on the detector. However, the PIN diodes have edges around the silicon chip which have different thickness from that of the bulk and, hence, collimators are used to shield the edges. In the present work, we have used commercially available passivated, implanted, planar silicon (PIPS) detectors, which are housed in a metal frame to cover the edges, to measure electron spectra and deduce multipolarities of γ -ray transitions.

In our experiments, we have used 25-mm^2 PIPS detectors of various thickness for electron spectroscopy. With these detectors we have obtained a resolution (FWHM) of 2.2 keV at 100-keV electron energy for $100 \mu\text{m}$, $300 \mu\text{m}$, and $500 \mu\text{m}$ thick detectors. Because of their smaller thickness, compared to that of cooled lithium-drifted silicon detectors, these detectors are less sensitive to γ rays and x rays, and hence provide a cleaner spectrum for low energy electrons. The spectrum of a mass-separated ^{244}Cm source, measured with a 3-mm thick cooled Si(Li) detector, is displayed in Fig. 1. This spectrum contains Pu L x

rays which interfere with the electron lines. The advantage of PIPS detectors is that an optimum thickness can be chosen for the electron spectrum to be measured. We have used 100 μ and 300 μ thick PIPS detectors to measure the electron spectra of even-even actinide nuclei, which decay by a transition of ~ 44 keV energy. For nuclei which decay by higher energy transitions, we use 500 μ thick detectors as was done for the measurement of conversion coefficients in the α decay of ^{249}Cf [5]. In this spectrum, L subshell, and M and N electron lines are well resolved.

One area where room-temperature electron spectroscopy can be effectively used is the determination of ^{239}Pu and ^{240}Pu isotopes in a sample. In general, ^{239}Pu samples have small amount of ^{240}Pu present and this content should be measured. For larger quantities of Pu samples, γ -ray spectroscopy and mass spectrometry are used for isotopic analysis. But for smaller samples, α -particle spectroscopy is normally used for the analysis of actinide nuclides. However, the energies of the ^{239}Pu (5156.59 keV) and ^{240}Pu (5168.13 keV) α groups are only 11.54 keV apart [6] and hence it is difficult to resolve ^{239}Pu and ^{240}Pu α groups. In the decay of these two nuclides, the excited states populated in the daughter nuclei deexcite by low-energy transitions which are highly converted. Thus, these isotopes can be identified by their conversion electron lines or by the L x rays which follow the conversion in L subshells. L x-ray yields have been measured with high precision [7] to be 0.0463(5) photons/ α for ^{239}Pu and 0.1032(15) photons/ α for ^{240}Pu ; but the energies and intensities of L x rays in the decay of both nuclides are identical and hence they can not be used. However, the energies of conversion-electron lines in both nuclei are different and hence they can be used for distinguishing the two isotopes.

Intensities and conversion coefficients of all 51.62- and 45.24-keV transitions in the decay of ^{239}Pu and ^{240}Pu have not been measured. Only subshell ratios were measured in the past in order to deduce transition multipolarities. In the present work we have measured the intensities and conversion coefficients of all electron lines of 51.62- and 45.24-keV transitions.

2. Experimental methods and results

Thin ($< 10 \mu\text{g}/\text{cm}^2$) isotopically pure ($>99.9\%$) sources of ^{239}Pu and ^{240}Pu , prepared in an electromagnetic isotope separator [8], were used in the present study. Alpha and electron spectra were measured with a $25\text{-mm}^2 \times 300\text{-}\mu\text{m}$ PIPS
65 detector. The detector and the electron source were placed in a chamber which was evacuated. A cooled FET preamplifier, AMPTEK model No. A250CF [9], was attached to the detector, and the output of the preamplifier was connected to an amplifier and a multichannel analyzer. Two amplifiers were used, one for α -particle counting and the other for electron spectra (which had 6 times the
70 gain of the amplifier for alpha counting). α -particle spectra of ^{239}Pu (1.30 kBq) and ^{240}Pu (6.10 kBq) sources, measured at a solid angle of 0.27% of 4π using a time constant of $1 \mu\text{s}$ for the amplifier, are displayed in Fig. 2. These spectra are plotted together in Fig. 3 to show the closeness of the two main peaks. Electron spectra of the two Pu isotopes are displayed in Fig. 4 and the peak
75 areas were determined with a peak-fitting program *gf3* [10]. The two spectra displayed together in Fig. 5 show that the ^{239}Pu and ^{240}Pu electron lines are well resolved.

3. Discussion

The electron spectrum of ^{240}Pu , shown in Fig. 4, has intense conversion
80 lines from only one transition, the 45.24-keV E2 transition. These are the $L_1 + L_2$, L_3 , M, and N+O lines. At lower energies the spectrum contains the KMN, and KNN Auger lines. Because the spectrum is simple, the areas of the conversion lines can be accurately determined. We have analyzed the spectrum and determined the peak areas. Since the alpha and electron spectra
85 are measured at exactly the same solid angle, the electron to alpha ratio has the uncertainty from the counts in the peak areas only. This ratio, determined from the alpha and electron spectra, is listed in Table I.

The ^{239}Pu spectrum contains main conversion-electron lines from the 51.62-keV E2 and the 38.7-keV M1/E2 transitions. The four conversion lines from

90 the 51.62-keV transition are well resolved and their areas are determined. The
 electron to alpha ratios for the electron lines from the 51.62-keV transition,
 obtained from the present spectra, are included in Table I. The L conversion
 lines of the 38.7-keV transition have large background and hence their electron
 intensities are not included in Table I. The 38.7 M line has almost the same
 95 energy as the 51.62 L_3 line and hence the electron to alpha ratio for the 51.62
 L_3 transition contains the contribution of the 38.7 M line. This contribution
 has been estimated as 0.61% from the known α_{52} intensity, the intensity of the
 51.62-keV transition, and the conversion coefficients of the 38.7-keV transition,
 and subtracted from the measured intensity of the 51.62 L_3 transition. Table I
 100 contains this corrected intensity and the conversion coefficient deduced from it.

From the alpha and electron spectra of a mass-separated ^{243}Cm source, we
 have determined that due to scattering, the electron detection efficiency relative
 to alpha detection is $82\pm 4\%$. Using this efficiency, we have determined the
 absolute intensities of ^{239}Pu and ^{240}Pu electron lines. The total intensity for
 105 the 45.24-keV transition in Table I is $26.7\pm 1.2\%$. This is in excellent agreement
 with the value of $27.2\pm 0.3\%$ for the $\alpha_{45} + \alpha_{149}$ groups [11] obtained from an
 alpha spectrum. From the intensities in Table I and the measured intensities of
 51.62- and 45.24-keV γ rays as 0.0272(2)% [12] and 0.0447(9)% [13], respectively,
 we have deduced the conversion coefficients for the ^{239}Pu and ^{240}Pu transitions.
 110 These values are in good agreement with the respective theoretical values [14].

The ^{239}Pu and ^{240}Pu electron spectra are plotted together in Fig. 5 showing
 the separation of electron lines in the decay of the two isotopes. A sample
 containing both isotopes can be analyzed by measuring its α and electron spectra
 as was done for the pure samples. The α -particle emission rate from the α
 115 spectrum of the mixed source is the sum of the alpha rates from each isotope.
 The mixed ^{239}Pu and ^{240}Pu source usually contains larger quantity of ^{239}Pu
 and, as Fig. 5 shows, its conversion-electron lines are well separated from the
 ^{240}Pu lines. Thus, we can determine the areas of ^{239}Pu electron lines and using
 the electron to alpha ratios, determined for the pure source, we can determine
 120 the ^{239}Pu α -particle emission rate. The difference between the total alpha rate

and the ^{239}Pu alpha rate gives the α -particle emission rate of ^{240}Pu .

It is worth pointing that for the determination of the isotopic composition, one needs the ratio between the electron intensity and alpha intensity, which has uncertainty of only $\sim 1\%$, as shown in Table I. Also, area of only one electron peak in the spectrum of the mixed source with the smallest uncertainty is sufficient for the determination of the isotopic composition. In the electron spectrum of a mixed source, the peak areas have larger uncertainties due to larger backgrounds under the peaks and hence one expects larger uncertainty than for the spectrum of the pure isotope. To estimate this increase in uncertainty, we have added the two electron spectra of Fig. 4 and determined the areas of the peaks. We find that the areas of the peaks in the summed spectrum are, on the average, $\sim 3\%$ different from that determined from the spectra of pure sources. This gives an estimate of the uncertainty expected in the measurement. In addition to the isotopic composition, this procedure can also be used to obtain the absolute α -decay rate of each isotope by determining the solid angle of the PIPS detector with a calibrated standard source. It should be pointed out that the uncertainties in the measurement depend on the quality of the source, such as thickness and area, and the relative abundances of the two isotopes.

4. Summary

Using alpha and electron spectra measured with a room-temperature PIPS detector, we have shown that one can determine the isotopic composition of a sample containing ^{239}Pu and ^{240}Pu isotopes. Uncertainties of $\sim 5\%$ can be achieved in these measurements depending on the source quality and the isotopic composition. We have also measured with high precision the intensities and conversion coefficients for the strong transitions in the decay of ^{239}Pu and ^{240}Pu . It has been demonstrated that with cooled PIN diodes, one can improve the electron resolution (FWHM) to ~ 1.0 keV [4], which will make the measurement even more sensitive. Currently, silicon drift detectors (SDD) are commercially available for x-ray measurements with a resolution (FWHM) of

150 less than 200 eV at 50 keV energy. The detector housing of these spectrometers can be modified for the measurement of electron spectra and could provide unprecedented resolution and sensitivity.

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References

- [1] I. Ahmad and F. Wagner, Nucl. Instr. and Meth. **116** (1974) 465.
- [2] I. Ahmad, R. R. Betts, T. Happ, D. J. Henderson, F. L. H. Wolfs, and A.
160 H. Wuosmaa, Nucl. Instr. and Meth. Phys. Res. **A299** (1990) 201.
- [3] W. R. Wampler and B. L. Doyle, Nucl. Instr. and Meth. Phys. Res. **A349** (1994) 473.
- [4] Ch. Weinheimer, M. Schrader, J. Bonn, Th. Loeken, and H. Backe, Nucl. Instr. and Meth. Phys. Res. **A311** (1992) 273.
- 165 [5] I. Ahmad, J. P. Greene, F. G. Kondev, and S. Zhu, to be published.
- [6] A. Rytz, Atom. Data Nucl. Data Tables **47** (1991) 227.
- [7] M. C. Lépy and K. Debertin, Nucl. Instr. and Meth. Phys. Res. **A339** (1994) 218.
- [8] J. Lerner, Nucl. Instr. and Meth. **102** (1972) 373.
- 170 [9] AMPTEK Inc, 14 DeAngelo Drive, Bedford, MA 01730.
- [10] D.C. Radford, Nucl. Instr. and Meth. Phys. Res. **A361** (1995) 297.
- [11] I. Ahmad, Nucl. Instr. and Meth. Phys. Res. (1984) 319.

- [12] E. Browne and J. K. Tuli, Nucl. Data Sheets **107** (2006) 2649.
- [13] E. Browne, Nucl. Data Sheets **98** (2003) 665.
- ¹⁷⁵ [14] I. M. Band, M. B. Trzhaskovskaya, C. W. Nestor, Jr., P. O. Tikkanen, and
S. Raman, Atom. Data Nucl. Data Tables **81** (2002) 1.

Table 1: Data on ^{239}Pu and ^{240}Pu electron lines from the present work.

Transition energy (keV)	Electron energy (keV)	Electron/alpha ratio	Electron intensity	Conversion coefficient	
				experiment	theory (E2)
51.62 L ₁ +L ₂	30.8(1)	0.0291(4)	3.55(15)%	130(6)	125
L ₃	34.6(1)	0.0271(4)	3.30(14)%	100(5)	102
M	47.0(1)	0.0153 (3)	1.87(8)%	69(4)	63
N+O	50.8(1)	0.0055 (2)	0.67(3)%	25(2)	22
45.24 L ₁ +L ₂	24.4(1)	0.0867(8)	10.6(5)%	237(11)	229
L ₃	28.2(1)	0.0753(7)	9.2(4)%	205(10)	193
M	40.6(1)	0.0466(5)	5.7(3)%	127(6)	116
N+O	44.4(1)	0.0161(2)	1.20(5)%	44(2)	40

Figure 1: An electron spectrum of a mass-separated ^{244}Cm source measured with a cooled $80\text{-mm}^2 \times 3\text{-mm}$ Si(Li) detector. The energy scale is 72.5 eV per channel.

Figure 2: α -particle spectra of mass-separated ^{239}Pu and ^{240}Pu sources measured with a $25\text{-mm}^2 \times 300\text{-}\mu\text{m}$ PIPS detector. The energy scale is 1.29 keV per channel and the solid angle was 0.27% of 4π . The energy resolution (FWHM) is 11.0 keV. Counting time was 3380 minutes for ^{239}Pu spectrum and 450 minutes for ^{240}Pu spectrum.

Figure 3: α -particle spectra of of Fig. 2 plotted together to show the closeness of the two α lines. The two α_0 peaks are only 11.54 keV apart.

Figure 4: Electron spectra of mass-separated ^{239}Pu and ^{240}Pu sources measured with the same set up that was used for the measurement of the α -particle spectra in Fig. 2. The energy scale is 0.214 keV per channel and the energy resolution (FWHM) is 2.2 keV. Counting time was 4250 minutes for ^{239}Pu spectrum and 267 minutes for ^{240}Pu spectrum.

Figure 5: Electron spectra of Fig. 4 plotted together to show the separation of ^{239}Pu and ^{240}Pu lines. Blue lines represent the ^{239}Pu electrons.