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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 setup 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.

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 ~1 cm^2 have been used which have a resolution [full width at half maximum (FWHM)] comparable to that of germanium detectors (~1.0 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 μm, give a resolution (FWHM) of ~2.4 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 to 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 μm, 300 μm, and 500 μm 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 μm and 300 μm 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 μm 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.

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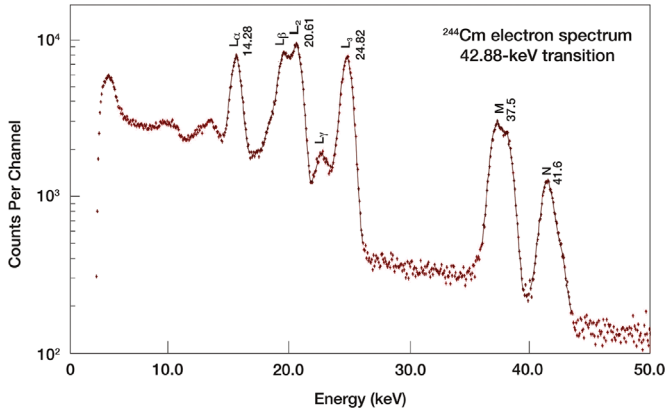

Fig. 1. An electron spectrum of a mass-separated ^{244}Cm source measured with a cooled 80-mm $^2\times$ 3-mm Si(Li) detector. The energy scale is 72.5 eV per channel.

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 deexcited 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 cannot 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 ($<10\,\mu g/cm^2$) 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-mm$^2\times$300-μm PIPS 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 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 μs 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 areas were determined with a peak-fitting program *gf*3 [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 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 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 1.

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 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 1. The L conversion lines of the 38.7-keV transition have large background and hence their electron intensities are not included in Table 1. The 38.7 M line has almost the same energy as the 51.62 L_3 line and hence the electron

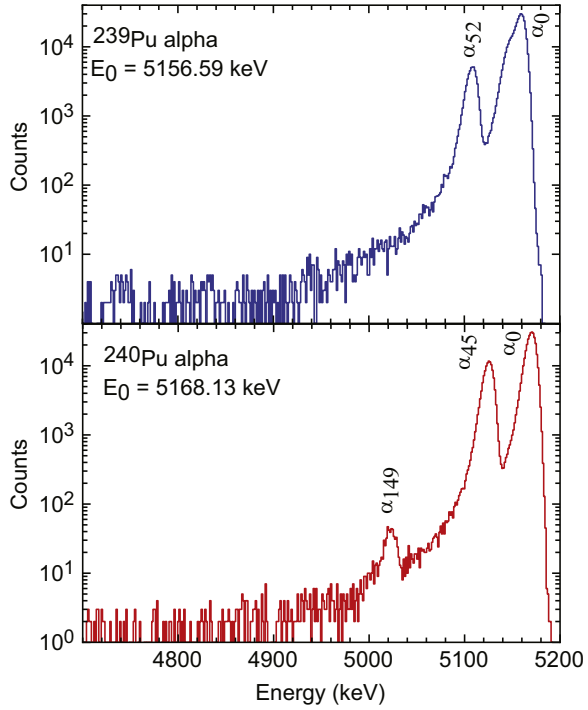

Fig. 2. The α-particle spectra of mass-separated ^{239}Pu and ^{240}Pu sources measured with a 25-mm$^2\times$300-μm PIPS detector. The energy scale is 1.29 keV per channel and the solid angle was 0.27% of 4π. The energy resolution (FWHM) is 11.0 keV. Counting time was 3380 min for ^{239}Pu spectrum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

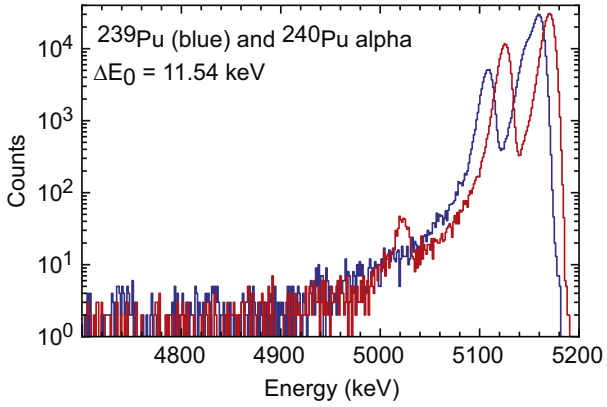

Fig. 3. The α-particle spectra of Fig. 2 plotted together to show the closeness of the two α lines. The two α_0 peaks are only 11.54 keV apart.