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Alpha and conversion electron spectroscopy of ^{238,239}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 mm^2 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.

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 used for alpha spectroscopy this ranges from 10–22 keV FWHM. These characteristics make alpha spectrometry a very popular radioanalytical method for isotopic identification of actinide samples. The method has downfalls including unresolvable alpha particle energies for nuclides of interest to the safeguards and forensics community, and chemistry procedures for sample preparation. As an example of the energy interference, the main alpha particles emitted from ^{239}Pu/^{240}Pu and ^{238}Pu/^{241}Am are separated by 11.6 keV and 13.4 keV [1], respectively. Hence, if a sample contains ^{239}Pu and ^{240}Pu, alpha spectrometry may be used in combination with mass spectrometry to determine the individual isotopics of the sample. Similarly, chemical processes can be performed to separate the plutonium from the americium and the individual samples can be analyzed with alpha spectrometry since mass spectrometry has difficulties with ^{238}Pu because of natural uranium background.

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

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spectrum of these isotopes is a crucial step in understanding the proposed coincident measurement.

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

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

2. Apparatus

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

2.1. Alpha measurements

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

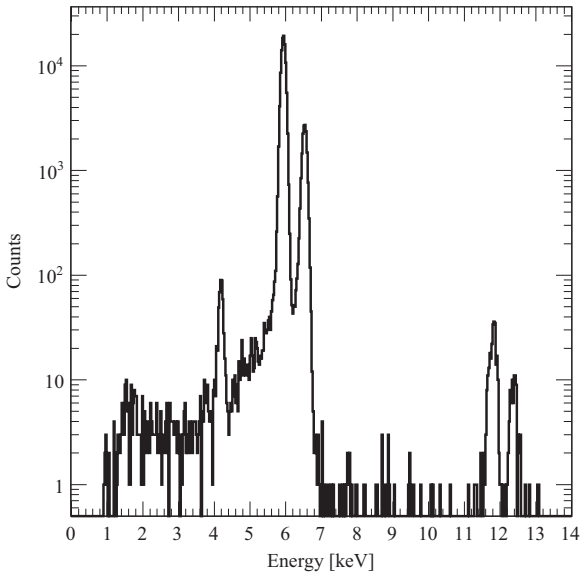

Fig. 1. Energy spectrum of ^{55}Fe taken at 234 K with the SDD equipped with the silicon nitride window. The FWHM energy resolution is 141 eV for the 5.9 keV peak.

2.2. Conversion electron measurements

The idea of a semiconductor drift detector was first proposed by Gatti and 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 Fig. 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 mm^2 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 in 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 Fig. 1. The FWHM energy resolution