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Conversion electron spectrometry of Pu isotopes with a silicon drift detector

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HIGHLIGHTS

- Built set-up for conversion electron spectrometry with silicon drift detector.
- Conversion electron spectra measured from ^{238,239,240}Pu and ^{241}Am.
- Compared with other SDD set-up and Si(Li) detector.

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ABSTRACT

An electron spectrometry set-up was built at IRMM consisting of a vacuum chamber with a moveable source holder and windowless Peltier-cooled silicon drift detector (SDD). The SDD is well suited for measuring low-energy x rays and electrons emitted from thin radioactive sources with low self-absorption. The attainable energy resolution is better than 0.5 keV for electrons of 30 keV. It has been used to measure the conversion electron spectra of three plutonium isotopes, i.e. ^{238}Pu, ^{239}Pu, ^{240}Pu, as well as ^{241}Am (being a decay product of ^{241}Pu). The obtained mixed x-ray and electron spectra are compared with spectra obtained with a close-geometry set-up using another SDD in STUK and spectra measured with a Si(Li) detector at IRMM. The potential of conversion electron spectrometry for isotopic analysis of mixed plutonium samples is investigated. With respect to the ^{240}Pu/^{239}Pu isotopic ratio, the conversion electron peaks of both isotopes are more clearly separated than their largely overlapping peaks in alpha spectra.

1. Introduction

Plutonium (Pu) is generally produced in a variety of isotopic mixtures in reactor fuel. The predominant isotope, ^{239}Pu, is produced by neutron capture of ^{238}U and the heavier isotopes by further neutron capture. Spent fuel may contain approximately 50–60% of ^{239}Pu, 25% of ^{240}Pu, 15% of ^{241}Pu, 5% of ^{242}Pu, and 2% of ^{238}Pu. The ^{238}Pu, formed through decay of ^{242}Cm, is responsible for much of the Pu decay heat because of its short half-life (87.74 a). The ^{239}Pu is fissile and can be used as fuel for a chain reaction in a reactor or a nuclear weapon. Weapon makers prefer a high enrichment in ^{239}Pu because of its low rate of heat generation and relatively low spontaneous emission of neutrons and gamma rays, whereas the presence of ^{240}Pu may limit a bomb's potential by causing predetonation due to its neutron flux from spontaneous fission. Modern weapon designs – using deuterium–tritium fusion to boost the fission yield, decrease weight and increase safety – can be made insensitive to the isotopic mix, but at a low level of sophistication a high concentration of ^{240}Pu is considered a barrier to nuclear proliferation (Feiveson et al., 2014).

The measurement of isotopic amount ratios is an important topic in safeguards and nuclear forensics applications, and needed to support non-destructive assay (NDA) methods – calorimetry and neutron coincidence counting – in providing the total plutonium content of a sample (Sampson, 1991). For large quantities of material, gamma spectrometry techniques are used for the analysis. The alpha decay of the ^{236,238,239,240,242,244}Pu isotopes and the beta decay of ^{241}Pu primarily populate the ground state and/or the lowest excited states of their daughter nuclides, and the gamma transitions depopulating those low-lying excited states have a high probability of being internally converted (DDEP, 2004–2015). As a result, the intensity of characteristic gamma-ray emission is very low and isotopic analysis by gamma spectrometry is a challenge

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for low activity levels of plutonium. Only the ingrowth of ^{241}Am (and a small fraction of ^{237}U) from the decay of ^{241}Pu leads to significant gamma emission. The most problematic isotope is ^{242}Pu, due to its low abundance, long half-life and its few gamma rays similar in energy and branching ratio to those from ^{240}Pu (Sampson, 1991).

Small amounts of Pu in environmental and biological samples are addressed with more sensitive techniques for isotopic analysis, such as mass spectrometry (Jakopič et al., 2010), alpha-particle spectrometry (Bortels and Collaers, 1987), neutron induced fission track analysis specific for ^{239}Pu (Moorthy et al., 1988) and liquid scintillation counting for ^{241}Pu (Ikäheimonen, 2000). The isotopic ratio measurements can provide important information for identifying a source of contamination, e.g. to distinguish particles from global nuclear weapon fallout and from a nuclear reactor accident (Kudo, 2001; Evrard et al., 2014). The age of plutonium material (i.e. formation of ^{241}Pu by irradiation or last chemical separation from ^{241}Am) can be derived from measurement of the ^{241}Am/^{241}Pu isotope ratio (Varga, 2007) or extrapolation of the ^{241}Am ingrowth over time (Sibbens et al., 2008). Plutonium clocks are used for age dating in nuclear forensics (Wallenius and Mayer, 2000; Sturm et al., 2014; Pommé et al., 2014).

Mass spectrometry techniques (TIMS, AMS, RIMS, ICP-MS) are the most sensitive and precise methods, except that the determination of ^{238}Pu is more difficult due to isobaric interference from ^{238}U (Jakopič et al., 2010). As an alternative, alpha spectrometry has advantages of simplicity, ability to use a radiotracer, and low cost (LaMont et al., 1998). As activity ratios differ from atom ratios by the decay constants, it relatively enhances the signal from the short-lived isotopes, in particular ^{238}Pu. Determining the ^{240}Pu/^{239}Pu ratio is possible (Bortels and Collaers, 1987; Raab and Parus, 1994; LaMont et al., 1998; Pöllänen et al., 2012) but problematic due to the limited energy resolution and the spectral interference of the main ^{239}Pu (5156.6 keV) and ^{240}Pu (5168.1 keV) alpha emission peaks. The main peaks of the other isotopes – ^{236}Pu (5767.5 keV), ^{238}Pu (5499.0 keV), ^{242}Pu (4902.3 keV), and ^{244}Pu (4589 keV) – are well separated, but ingrowth of ^{241}Am (5485.6 keV) interferes with ^{238}Pu (Sibbens et al., 2008). In old archived samples, the ^{241}Pu/$^{239+240}$Pu activity ratio can be evaluated by α spectrometry through the accrued ^{241}Am (Vukanac et al., 2006; Salminen-Paatero et al., 2014).

For a further analytical separation of ^{239}Pu and ^{240}Pu in environmental samples, the α-particle spectrometry can be complemented with additional measurements of the L x rays emitted from the same sources (Hisamatsu and Sakanoue, 1984; Komura et al., 1984; Arnold and Kolb, 1995; Pöllänen et al., 2009), taking into account the different emission probabilities of L x rays for ^{239}Pu and ^{240}Pu (Lépy and Debertin, 1994). The measurement can be done on the same source as for the α-particle spectrometry, possibly in an underground laboratory to reduce the background signals (Arnold, 2006).

Low-energy gamma transitions of actinides being highly converted, internal conversion electron (ICE) spectrometry offers an interesting complement – or even alternative – to alpha spectrometry. Shiokawa and Suzuki (1986) examined the principle on a ^{243}Cm/^{244}Cm mixture using a cooled silicon surface barrier detector. At approximately 150 K, they reached a resolution of 1.8 keV for electrons of 42 keV (60-L_3 line of ^{241}Am) and 2.25 keV at 624 keV (661-K line of ^{137}Cs). Shiokawa et al. (1989, 1990, 1991) developed a new ICE spectrometer using a windowless Si(Li) detector with improved energy resolution of 0.48 keV and 1.43 keV for 42 keV and 624 keV electrons respectively, and succeeded to determine isotopic ratios for plutonium and curium.

The Si(Li) detector is operated at liquid nitrogen temperature and sample changing requires specific measures to avoid condensation of humidity on the crystal surface. At STUK, Peräjärvi et al. (2014) investigated the performance of Peltier-cooled, windowless silicon drift detectors (SDD) for ICE spectrometry. These detectors are more user-friendly than Si(Li) detectors and they reach a resolution of 0.50 keV (FWHM) for electrons of 45 keV at operational temperatures between −20 °C and −10 °C. Good source quality is required, since the energy resolution shows considerable broadening for 1-μm-thick samples, but remains good (0.70 keV) for thin plutonium samples covered with a 0.5 μm Mylar absorber foil. This is significantly better than the 2.2 keV resolution of ion-implanted planar silicon detectors used for alpha spectrometry (Ahmad et al., 2015).

This work describes a new SDD set-up built at IRMM. Measured ^{238,239,240}Pu and ^{241}Am ICE spectra are shown and compared with spectra from other devices.

2. Set-up

Fig. 1 shows a schematic drawing of the set-up for the internal conversion electron spectrometer at IRMM. A picture of the detector and source chamber is shown in Fig. 2. The detector is a windowless Canberra X-PIPS® silicon drift detector with an active area of 100 mm^2 and thickness of 500 μm. A circular multilayer diaphragm composed of 200 μm Ta, 35 μm Cr, 20 μm Ti and 75 μm Al limits the effective area to approximately 80 mm^2. The detector and its field-effect transistor are thermo-electrically cooled down to −20 °C, in order to reduce noise and to ensure stable operation in changing environmental conditions. The detector element is mounted on a finger which is inserted into the vacuum chamber. The housing of the detector contains a reset-type preamplifier, a high-voltage bias supply and a temperature controller for the thermo-electric cooler.

The chamber is constructed from commercially available components, including a central cubic piece with matching flanges and a hinged door for easy access to the interior. The source holder is placed on the tip of a rotary actuated linear feedthrough, which enables moving the source up or down, thus reproducibly varying the source–detector distance from millimetres up to 13.5 cm at any time. By means of a turbo-molecular vacuum pump, the air pressure is kept well below 10^{-4} mbar. To avoid condensation of any residual humidity on the surface of the detector, a cold point has been created in the vacuum chamber by means of a thermo-electric cooler kept at a lower temperature (−40 °C) than the detector. The front panel of the set-up houses the controllers of the vacuum gauge, the turbo-molecular vacuum pump, and the power supply and temperature readout of the cold point.

In its standard configuration as an x-ray detector, the SDD has a 50 fF feedback capacitor with a gain of 0.9 mV/keV. This would be a convenient setting for the low-energy signals from the x rays and ICEs, but the impact of 5.5 MeV α particles on the detector would cause output signals of 5 V and regular resets of the pre-amplifier. Therefore, the feedback capacitor was increased to 0.5 pF to reduce the gain, potentially at the expense of some loss of energy resolution. The large signals from α particles cause afterpulses interfering with the low-energy spectrum and peak detection in the analog-to-digital convertor had to be disabled for 40 μs after detection of a signal.

The actinide sources measured in this work are thin layers of highly enriched material deposited on a glass, quartz or stainless steel substrate, mostly prepared by vacuum evaporation (Sibbens and Altzitzoglou, 2007) for the purpose of high-resolution alpha-particle spectrometry (Pommé and Sibbens, 2008; Sibbens et al., 2008, 2010). Whereas in alpha-particle spectrometry a magnet system (Paepen et al., 2014) can be used to eliminate the signal from conversion electrons, by reversing the poles of one of the magnets, the same system can be used as an electron lens to