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Deconvolution of ^{238,239,240}Pu conversion electron spectra measured with a silicon drift detector

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HIGHLIGHTS

- Performed conversion electron spectrometry with silicon drift detector.
- Deconvoluted conversion electron spectra from ^{238,239,240}Pu.
- Alpha deconvolution software applied successfully on ICE spectra.
- ICE emission probabilities in good agreement with BrIcc.

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ABSTRACT

Internal conversion electron (ICE) spectra of thin ^{238,239,240}Pu sources, measured with a windowless Peltier-cooled silicon drift detector (SDD), were deconvoluted and relative ICE intensities were derived from the fitted peak areas. Corrections were made for energy dependence of the full-energy-peak counting efficiency, based on Monte Carlo simulations. A good agreement was found with the theoretically expected internal conversion coefficient (ICC) values calculated from the BrIcc database.

1. Introduction

Plutonium has been investigated from many angles (Clark et al., 2018), among which analytical detection techniques for nuclear security and safeguards is an important one. With the exception of the β^--emitter ^{241}Pu, the long-lived plutonium isotopes are predominantly alpha emitters. For the determination of plutonium in environmental samples, nuclides of interest are generally ^{238}Pu, ^{239}Pu and ^{240}Pu. Since the intensities of their characteristic gamma-ray emissions are very low, isotopic analysis by gamma spectrometry is a challenge for low activity levels of plutonium. High-resolution alpha-particle spectrometry (Pommé and Sibbens, 2008) is a well-suited alternative technique for measuring isotopic activity ratios in a plutonium mixture, however it suffers from close interferences between the ^{239}Pu and ^{240}Pu peaks and between the ^{238}Pu and ^{241}Am peaks, respectively (see references in Pommé et al., 2016).

A promising alternative technique is spectrometry of the internal conversion electrons (ICE) from the highly converted low-energy gamma transitions following the decay of Pu isotopes. The principle of the method has been tested with cooled silicon surface barrier detectors (1.8 keV FWHM resolution at 42 keV) (Shiokawa and Suzuki, 1986), windowless Si(Li) detectors (0.48 keV FWHM at 42 keV) (Shiokawa et al., 1990), PIPS detectors cooled (DeVol et al., 2002) or at room temperature (2.2 keV FWHM) (Ahmad et al., 2015), and recently Peltier-cooled silicon drift detectors (SDD, 0.5 keV resolution) (Peräjärvi et al., 2014; Pommé et al., 2016; Dion et al., 2016). It was demonstrated that the ICE peaks from ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Am are well separated in energy. ICE spectrometry is better suited than alpha spectrometry in preserving its capability to determine the ^{240}Pu/^{239}Pu isotopic ratio as a function of sample thickness (Peräjärvi et al., 2014). However, as a complementary or standalone technique it requires further research on spectrum deconvolution and reference emission data.

In this work, the relative emission probabilities for the L and M+ ICE peaks for the 43.469 keV (^{238}Pu), 51.624 keV (^{239}Pu), and 45.244 keV (^{240}Pu) gamma transitions are investigated. To this end, independent deconvolution software tools have been developed at JRC and STUK to fit simultaneously the x-ray and conversion electron peaks obtained with a silicon drift detector. The ICE peak areas are

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determined and relative intensities are derived. A comparison is made with theoretical calculations and experimental data from literature.

2. Experiment

2.1. ICE spectrometry with an SDD

In previous work, the novel ICE spectrometry set-ups with SDD at STUK and JRC have been described in detail (Peräjärvi et al., 2014; Pommé et al., 2016). They are well suited for measuring low-energy x-rays and electrons. Internal conversion electron (ICE) spectra have been taken of thin plutonium sources, respectively enriched in the isotopes ^{238}Pu, ^{239}Pu, and ^{240}Pu. The spectra show major ICE peaks between 20 and 50 keV, with an energy resolution of 0.5 keV. The shape of the conversion electron energy peaks is comparable to alpha peaks, with typical peak shift and tailing due to energy loss in the absorbing materials. The x-ray energy peaks are comparably sharp and their position is invariable to the thickness of the source and other dead layers. As a result of the difference in peak shift, a separate energy calibration is applied for electron and x-ray peaks.

2.2. Spectral deconvolution

Just as in alpha spectrometry (Pommé, 2015), peak overlap is an issue in ICE spectrometry. It requires the use of spectral deconvolution software which can quantify the full-energy peaks as well as reproduce the continuum underneath the peaks. As suggested in earlier work (Pommé et al., 2016), the analytical functions used in alpha spectrometry are well suited to represent electron peaks with varying degrees of energy straggling. At the JRC, the spreadsheet application 'BEST' (Pommé and Caro Marroyo, 2015) and at STUK, the software 'ADAM' (Pöllänen et al., 2012) were used for spectral analysis. The analytical functions consist of a Gaussian distribution convoluted with exponential tailing functions, the number of which is chosen on the basis of the spectral shape. ADAM allows up to 3 left-handed tailing functions and can fit a continuum underneath the peaks by means of a polynomial function. BEST allows for a higher number of left-handed as well as right-handed exponentials, but fully relies on the peak shapes to cover also the continuum part of the spectrum. The x-ray peaks at 20.2 keV and 20.8 keV were represented by a Gaussian function each (whereas ideally a Voigt function is used and a low-energy continuum added to the peak function). Their intensity ratio was kept fixed at a value of 4.7/1 (Chu et al., 1999).

2.3. Simulation of counting efficiency

The total counting efficiency for the conversion electrons in a silicon detector deviates by about 14–18% (Martin et al., 2013) from the geometrical efficiency due to backscattering against the surface. The backscattering probability depends on the angle of incidence and the energy of the electron. Of the detected particles, a fraction of about 88–91% of the kinetic energy is deposited in the Si crystal and the peak-to-total ratio is about 81–85%. Monte Carlo simulations of particle transport (Salvat and Fernández-Varea, 2009) were performed with Penelope (Salvat et al., 2011) and EGSnrc (Kawrakow and Rogers, 2001) to estimate the energy dependency of the counting efficiency.

2.4. Theoretical calculations with BrIcc

Theoretical values of the ICC, using the frozen-orbital approximation, are available through the database 'BrIcc', which was accessed through a web interface (Kibédi et al., 2008). Input parameters are the daughter nuclides of the Pu isotopes as well as the energy and polarity of their gamma transitions. Some characteristics of the main transitions for ^{238,239,240}Pu are listed in Table 1. The 43.469 keV (^{238}Pu), 51.624 keV (^{239}Pu) and 45.244 keV (^{240}Pu) gamma transitions are highly converted and have E2 polarity. In the case of ^{239}Pu, a second gamma transition at 38.661 keV with mixed M1 + 22.2% E2 polarity (δ = 0.534 (24)) was taken into account (DDEP, 2017). The ICE intensities were calculated from the ICCs and the gamma transition probability, i.e. $I_{ce,x} = P_{\gamma+ce}\ \alpha_x/(1 + \alpha_T)$.

Table 1
ICCs calculated with BrIcc and subsequently derived ICE intensities for 4 gamma transitions in the decay of ^{238,239,240}Pu. The gamma transition probabilities are obtained from (DDEP, 2017).

Parent	^{238}Pu	^{239}Pu	^{239}Pu	^{240}Pu
Daughter	^{234}U	^{235}U	^{235}U	^{236}U
Transition (keV)	43.498	51.624	38.661	45.244
$P_{\gamma+ce}$ (%)	28.3 (8)	8.38 (18)	3.56 (21)	27.3 (8)
Polarity	E2	E2	M1 + 22.2% E2	E2
α_T	713 (10)	310 (5)	339 (20)	589 (9)
α_L	520 (8)	226 (4)	249 (14)	429 (6)
α_M	143.5 (20)	62.6 (9)	67 (4)	118.6 (17)
I_{ce} (%)	28.26	8.35	3.55	27.25
$I_{ce,L}$ (%)	20.61	6.09	2.61	19.85
$I_{ce,M}$ (%)	5.69	1.69	0.70	5.49
$I_{ce,N-Q}$ (%)	1.96	0.577	0.24	1.92
$I_{ce,M-Q}$ (%)	7.65	2.26	0.94	7.40

3. Results

3.1. Counting efficiency

For the simulation of the full-energy-peak (FEP) detection efficiency, $\varepsilon_P(E)$, the detector was represented as a silicon wafer with 100 mm^2 area, 500 µm thickness, and 50 nm dead layer. Electrons with energy between 10 and 120 keV were generated from a point or 1-cm-diameter disk source, at 1 cm, 2 cm or 5 cm distance on the symmetry axis, using random angles within the solid angle subtended by the 80 mm^2 aperture of the circular diaphragm in front of the detector. The simulated detection probabilities for electrons emitted within the aperture are shown in Fig. 1.

Differences in counting efficiency for the different configurations are due to the increasing probability for backscattering with increasing deviation from a perpendicular incidence angle. The angular dependence of the detection efficiency can be approximated by a polynomial function

$$\frac{\varepsilon_P(\theta)}{\varepsilon_P(0)} \approx 1 + a_1\theta + a_2\theta^2 + a_3\theta^3 \tag{1}$$

in which the angle θ (expressed in radian) takes the value 0 for a perpendicular hit. For a silicon disk without dead layer, the angular dependency at $E = 35$ keV simulated with EGSnrc is quasi parabolic between $\theta = 0$ and $\theta = 1$, with $a_1 = 0.0416$ and $a_2 = -0.293$.

The $\varepsilon_P(E)$ values obtained with EGSnrc and Penelope show a similar energy dependency, even though they differ in absolute value by about 1% (see Fig. 1). However, only the detection efficiency ratios among the ICE peaks is of relevance in this work. All data sets, when divided by their reference value at $E = 120$ keV, can be rather well represented by the same polynomial function (see bottom graph of Fig. 1)

$$\frac{\varepsilon_P(E)}{\varepsilon_P(120\ \text{keV})} \approx \exp[a_0 + a_1\ln(E) + a_2\ln(E)^2 + a_3\ln(E)^3] \tag{2}$$

The fit to the EGSnrc $\varepsilon_P(E)/\varepsilon_P(120\ \text{keV})$ data for a point source at 2 cm distance, with $a_0 = -0.594$, $a_1 = 0.39556$, $a_2 = -0.09007$, and $a_3 = 0.006969$, was taken as the reference efficiency curve in this work. The effective thickness of the dead layer is a major source of uncertainty at low energy.

3.2. Spectral deconvolution

The spectra were first subjected to a fit of all ICE peaks in one go,