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Characterization of strong ^{241}Am sources

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HIGHLIGHTS

- Age and impurities can be used as a signature for ^{241}Am sources.
- Nuclear reactions take place in sources with low Z impurities.
- Some sources contain ^{243}Am as an impurity.

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ABSTRACT

Gamma ray spectra of strong ^{241}Am sources may reveal information about the source composition as there may be other radioactive nuclides such as progeny and radioactive impurities present. In this work the possibility to use gamma spectrometry to identify inherent signatures in ^{241}Am sources in order to differentiate sources from each other, is investigated. The studied signatures are age, *i.e.* time passed since last chemical separation, and presence of impurities. The spectra of some sources show a number of Doppler broadened peaks in the spectrum which indicate the presence of nuclear reactions on light elements within the sources. The results show that the investigated sources can be differentiated between by age and/or presence of impurities. These spectral features would be useful information in a national nuclear forensics library (NNFL) in cases when the visual information on the source, *e.g.* the source number, is unavailable.

1. Introduction

Nuclear forensic science is an important tool for combating illicit trafficking of nuclear material as well as other criminal activities related to nuclear and other radioactive materials. Nuclear forensic investigations commonly include characterization of nuclear material and other radioactive materials such as isotope ratio measurements, age determination (*i.e.* time passed since last chemical separation), impurity measurements and physical characterization (Wallenius et al., 2006; Kristo, Tumey, 2013; Stanley et al., 2013). Traditionally, the nuclear material investigated is uranium or plutonium but there are also other nuclides, such as ^{241}Am, that are fissionable and hence regarded as an alternative nuclear material (IAEA, 2002). ^{241}Am is a radionuclide that can be used in other contexts as well, such as ionizing smoke detectors where ^{241}Am is used in small amounts (about 30 kBq in the small ones). Stronger ^{241}Am sources are used in industrial gauging applications and in combination with low Z elements such as beryllium or lithium, ^{241}Am can be used as a neutron source. However, the useful applications of ^{241}Am in society also entails that the sources may be susceptible to theft or other illegal activities. The Code of Conduct on the Safety and Security of Radioactive Sources (2004) states that every State should establish a national register of radioactive sources and further, that the State should ensure that radioactive sources are identifiable and traceable, or when this is not practicable, ensure that there are alternative processes for identifying and tracing sources. One way of keeping track of sources is by building national nuclear forensics libraries (NNFL) where information about radioactive sources and nuclear material in a State is kept in order to track the origin of a source when necessary. The information may, besides visual information

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and serial numbers, be information inherent in the source, *i.e.* the signatures described above.

Some work has been done to gather information that can be used as signatures for ^{241}Am. Gehrke and East (2000) determined the age of an ^{241}Am source by looking at the ratio between ^{241}Am and ^{233}Pa, the progeny of ^{241}Am. Another possibility of inherent signatures in strong alpha emitting sources is to measure *e.g.* emitted gamma photons from nuclear reactions taking place within the source. A number of papers and reports have been studying the alpha induced gamma rays from nuclear reactions on light elements (Lappalainen et al., 1983; Sastri and Schelhaas, 1985; Fazzari et al., 2003; Martin, 1975). These reactions occur when a strong alpha emitter is in close contact with light elements, *e.g.* fluorine, sodium, aluminum or magnesium. The low Z elements may be present as a part of the encapsulation of the source or as impurities within the source. The alpha particles will react with a low Z nucleus which in turn may emit a neutron or a proton. The resulting nucleus de-excites or decays resulting in emission of characteristic gamma rays. Moreover, other radionuclides may also be present as impurities, which could be measured and quantified relative to the main radioactive component of the source.

The aim of this work was to investigate the possibility of using gamma spectrometry to find inherent signatures in order to discriminate between different ^{241}Am sources in cases when visual signatures may not be accessible. A number of high activity ^{241}Am sources were measured using gamma spectrometry and the spectra were compared for possible signatures. During the initial gamma measurements, instead of identifying the material as ^{241}Am, the automatic identification of the instrument indicated that the measured material was plutonium due to the presence of neutrons together with ^{241}Am. The investigated signatures were age and impurities in the form of both radioactive nuclides and stable elements. Furthermore, Monte Carlo simulations have been used to clarify and explain the origin of the impurities seen in the gamma spectra.

2. Experimental

2.1. Source description and Monte Carlo model

Five sources were studied, of which two sources were apparently similar by visual inspection. The sources and measurement conditions were as follows:

- Source 1: Nominal activity 185 GBq ^{241}Am, sealed source contained in a lead shield during gamma spectrometric measurement.
- Source 2: Nominal activity 185 GBq ^{241}Am, sealed source contained in a lead shield during gamma spectrometric measurement.
- Source 3: Nominal activity 3.7 GBq ^{241}Am, sealed source measured with 1.10 mm Cd shielding.
- Source 4: Electroplated ^{241}Am source.
- Source 5: Smoke detector.

In order to evaluate the range of de-excitation time for nuclei excited due to nuclear reactions in an ^{241}Am source for nuclei with different initial energies, MCNPX+SRIM simulations were performed (Pelowitz, 2011; Ziegler and Biersack, 2014). The model used in the Monte Carlo simulations had the same geometrical parameters and material composition as Source 1 and Source 2. The dimensions of these sources can be found in Fig. 1. The total mass of each source was 68 g assuming a gross density of approximately 6.8 g/cm^3.

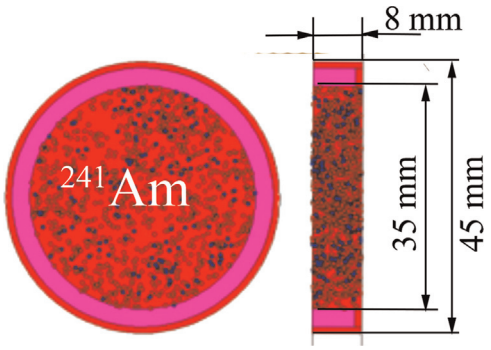

Fig. 1. Dimensions of Sources 1 and 2.

2.2. Gamma spectrometry and intrinsic efficiency calibration

Sources 1–4 were measured at a distance of about 30 cm using a p-type coaxial high purity germanium detector (Detective-EX, EG&G Ortec, Oak Ridge, TN, USA) which has a relative efficiency of about 15% and a resolution of 2.5 keV at 1332 keV. In addition, Source 5, the smoke detector, was measured for comparison. This spectrum was acquired with a p-type coaxial HPGe detector (EG&G Ortec, Oak Ridge, TN, USA) having a relative efficiency of 50% and a resolution of 2.0 keV at 1332 keV. The smoke detector was placed in a lead-shielded laboratory setup and measured for about one week. In order to characterize the response of these particular measurements, intrinsic response functions were established using a number of ^{241}Am gamma lines covering the energies from 59.5 to 801.9 keV and the response curves, *i.e.* the relative efficiency, were fitted to the following empirical polynomial previously published by Ramebäck et al. (2010):

$$\Psi = e^{c1+\frac{c2}{E^2}+c3(\ln(E))^2+c4(\ln(E))^3+\frac{c5}{E}} \quad (1)$$

Using the response curve, the activity of ^{233}Pa relative to ^{241}Am could be calculated and furthermore, the age of the source could be determined as well as the level of impurities (normalized to the activity of ^{241}Am). All spectra had the background subtracted prior to evaluation. The ^{241}Am gamma lines that were used for the intrinsic calibration and the following age determination can be found in Table 1.

2.3. Neutron measurements

As a part of evaluation, Source 1 was measured with two liquid

Table 1
Gamma energies with corresponding photon emission probabilities used for the intrinsic calibration and the age determination. Data are taken from Decay Data Evaluation Project (2013).

	E_γ [keV]	I_γ [%]
^{241}Am	59.5	35.92
^{241}Am	103.0	0.0195
^{241}Am	125.3	0.0041
^{241}Am	208.0	0.000786
^{233}Pa	300.1	6.6
^{233}Pa	311.9	38.3
^{241}Am	322.6	0.000151
^{233}Pa	340.5	4.47
^{241}Am	376.7	0.000137
^{241}Am	383.8	0.0000281
^{233}Pa	398.5	1.408
^{241}Am	619.0	0.000060
^{241}Am	662.4	0.000367
^{241}Am	722.0	0.000196
^{241}Am	801.9	0.0000012