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Surrogates of plutonium for detection equipment testing

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ABSTRACT

Fight against illicit trafficking of nuclear material relies on the possibility to detect nuclear material concealed in vehicles, people or cargo containers. This is done by equipping and training law enforcement and security staff in border stations or other points of access to strategic places and critical infrastructures with radiation detection equipment.

The design, development, testing and evaluation of these instruments ideally require the use of real nuclear material to assess, verify and certify their detection performance. Availability of special nuclear material may be an issue, especially for industry, since only few specialized laboratories are licensed for such material.

This paper tries to analyse and describe the possibility to use suitable surrogates that may replace the use of real nuclear material in testing the detection capabilities of instruments used in nuclear security.

1. Introduction

The public perception of the risk connected with the terrorist thread and the possibility of terrorist groups to produce a nuclear weapon or a radioactive device (dirty bomb) has grown since Sept. 11th. This raised the necessity from national authorities and international organisations to establish counteractions in order to prevent or reduce the possibility of illicit trafficking of nuclear material. Interdiction of trafficking requires an effective capability of radiation detection at transit points, such as borders, ports, airports or other strategic places.

A wide variety of detection equipment has been developed and is commercially available to detect radioactive or nuclear materials in people, vehicles or containerised cargos for different types of applications and uses: Radiation Portal Monitors (RPM), Personal Radiation Detectors (PRD), both also available with spectroscopic capability (SRPM and SPRD), Radio-Isotope Identifiers (RID), highly sensitive Gamma or Neutron Search Devices (GSD and NSD), Portable Radiation Scanners (PRS).

Testing this equipment is important in order to understand their capabilities and limitations, to derive indications for improvements and R&D, to select the most adequate instrument for each individual need and to optimise the inspection procedures. International standards (ANSI and IEC) have been developed to state the minimum requirements that these instruments need to fulfil [1]. The standards recommend also the test procedures that should be used to verify the compliance with the requirements.

Validation campaigns have been organised to assess the performances of border equipment; the most famous one is the ITRAP (Illicit Trafficking Radiation Assessment Program 1996–2000) carried out by the Austrian Research Centre Seibersdorf (ARCS) in collaboration with the International Atomic Energy Agency (IAEA) [11]. More recent testing programs aim to the verification of compliance with the international standards: ITRAP+10 recently established in both sides of the Atlantic and funded by the European Commission and by the US Department of Homeland Security, and the Graduated Radiological and/or Nuclear Detector Evaluation and Reporting (GRadER) programme, also launched by the US Department of Homeland Security.

All these testing procedures require the use of Special Nuclear Material (SNM) and in particular Highly-Enriched Uranium (HEU) and Weapon-Grade Plutonium (WG-Pu). These materials are not easily available and only few specialized laboratories have access and can use real nuclear material; this limits the possibility of testing equipment to few places.

There is therefore the interest to investigate on the possibility to develop suitable surrogates of special nuclear material that can replace the use or reduce the need of nuclear material for testing purposes.

Some papers have been published recently on the possible production of HEU spheres that can replicate the spectrum produced in a large HEU sample of 2.5 kg using only less than 50 g of HEU [2]. This requires still a laboratory licensed to detain SNM, but the lower mass reduces significantly the safeguards and security requirements for the laboratory and for the transport.

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In this paper we will analyse the possibility to produce a total surrogate of plutonium, which means a sample producing in the detector the same response as plutonium, but without any use of nuclear material. We will focus most of this study to the tests of the equipment producing alarms based on radiation levels only (RPM, PRD, GSD and NSD) and to a less extent we will consider spectrometric equipment. We do not see any possibility of using total surrogates for the testing of gamma spectrometers; in this case it will still be possible to design simulators of large samples containing minimum amount of plutonium mass.

2. Basic properties of radiation from plutonium

Plutonium isotopes are mostly alpha-emitters, with the exception of ^{241}Pu that is a beta-emitter. Both alpha and beta radiations are stopped either within the plutonium itself or within the walls of the sample container. The radiation escaping from the sample consists therefore only of photons and neutrons.

The emission of photons is always associated with the alpha and beta decay processes, the probability of gamma emission and its energy is isotope specific. This is why the gamma spectrum is a fingerprint for the identification of the source. In a photon spectrum of plutonium, X-rays are also present in addition to gamma peaks. Yields and energies of gamma lines of the plutonium isotopes can be found in literature [3].

Neutrons are generated in plutonium due to spontaneous fission, a secondary decay mode of most plutonium isotopes. Even isotopes (^{240}Pu, ^{242}Pu and ^{238}Pu) are the main contributors to the spontaneous fission neutron source. Spontaneous fission is the only neutron source in pure metal plutonium. When plutonium is chemically bound to a light element, as in oxides, the alpha particle from Pu decay may interact with the light element and cause an (alpha,n) reaction; this is a second possible neutron source in samples of plutonium compounds.

Due to the relatively short life of ^{241}Pu, which decays to ^{241}Am with a half-life of approximately 14 years, ^{241}Am is practically always present in all plutonium samples. Plutonium does not exist in nature and it is produced by irradiation of uranium in nuclear reactors. Therefore the plutonium may have very different isotopic compositions depending on the burnup cumulated during irradiation. For military applications a high content in the fissile isotope ^{239}Pu is desired, so typically weapon-grade plutonium contains more than 90% of ^{239}Pu. Commercial plutonium reprocessed by nuclear fuel irradiated in power reactors contains much larger amounts of the heavier isotopes. For the purpose of this paper we will define two specific isotopic compositions representative of typical weapon-grade (WG) and reactor-grade (RG) plutonium and their isotopic composition is reported in Table 1. The most common form of plutonium is the dioxide (PuO_2), but pure metal may also be used. Table 2 summarises the main characteristics of the specific radiation emission.

3. Surrogates of neutron emission

Neutron detectors generally detect the presence of a neutron through the ionisation induced by a particle generated by a neutron reaction with the detector material. It is therefore an indirect detection. Due to this conversion process, the information on the energy of the original neutron is lost, so only neutron counting is generally performed and not neutron spectroscopy. This simplifies a lot the requirements for the identification of a surrogate for neutron emission, because the surrogate must be a neutron emitter with less stringent conditions on the energy spectrum.

Table 1
Typical isotopic composition of weapon-grade and reactor-grade plutonium.

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am/Pu
WG-Pu	~0	93.8%	6.0%	0.2%	~0	0.001
RG-Pu	1%	60%	24%	10%	5%	0.05

Table 2
Activity and neutron source per gram of plutonium.

	Activity (Bq/g)	SF neutron emission (n/s)	(alpha,n) neutron emission in oxide (n/s)
WG-Pu	1.04E+10	61.8	44.3
RG-Pu	4.0E+11	359	325

Nevertheless the response of neutron detectors (efficiency or detection probability) is generally a function of the neutron energy, so if the surrogate must reproduce the response of the detector, it should have an emission spectrum reasonably close to the spectrum of plutonium. The quality of the spectrum reproduction will be evaluated by computing the detection probability in a classic neutron detector. As a reference we will use ^{3}He proportional counters embedded in a polyethylene slab, because this is the most commonly used neutron detector in RPM.

In conclusion the requirement for a surrogate will be:

- to have a total neutron emission equivalent to that of plutonium;
- to produce the same counting rate (average detection efficiency) in a ^{3}He detector.

3.1. Metal plutonium

When plutonium is in metal form, the neutron emission comes exclusively from spontaneous fission (SF). The shape of the neutrons emitted per spontaneous fission is quite similar for most of isotopes, in particular for the three even numbered isotopes of plutonium that are the major contributors to the neutron emission by SF. RG-Pu is generally in oxide form, whereas the metal form is more frequently used in military applications for WG-Pu where SF derives only from ^{240}Pu. From the two considerations above the reference spectrum of neutrons from SF in metal plutonium can be considered the spectrum of ^{240}Pu SF neutrons. Fig. 1 shows the comparison of different SF spectra for various plutonium isotopes and compositions; it can be easily seen that the different curves are nearly undistinguishable from each other.

The SF spectrum is a continuous spectrum and is generally represented by an analytical expression based on Watt distribution:

$$f(E) = Ce^{-E/a}\sinh\sqrt{bE}$$

where E is the neutron energy, C a normalisation constant such that the integral of the distribution function from zero to infinite gives 1, a and b are two isotope-specific parameters. Table 3 gives the Watt spectrum parameters for some isotopes of interest [4].

The most commonly diffused radioactive neutron source based on spontaneous fission is ^{252}Cf. This source is frequently used in laboratory applications. The spectrum of ^{252}Cf neutrons differs only marginally from that of ^{240}Pu, as can be seen from Fig. 2. Therefore it is easy to predict that a ^{252}Cf source can be an ideal candidate as surrogate of metal WG-Pu for SF neutron emission.