Applied Radiation and Isotopes 90 (2014) 187–191

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Detection of fast neutrons from shielded nuclear materials using a semiconductor alpha detector

R. Pöllänen*, T. Siiskonen

STUK – Radiation and Nuclear Safety Authority, P.O. Box 14, FI-00881 Helsinki, Finland

HIGHLIGHTS

- We investigated the response of a semiconductor alpha detector to fast neutrons emitted by nuclear materials.
- Low background and insensitivity to gamma rays are advantages of the detector operating at ambient air pressure.
- A polyethylene converter placed in front of the detector increased the detection efficiency by a factor of four.
- Intrinsic detection efficiency for fast neutrons from unshielded ^{252}Cf and ^{241}AmBe sources was 2.5×10^{-4} and 7.6×10^{-4}, respectively.

ARTICLE INFO

Article history:
Received 10 September 2013
Received in revised form
26 February 2014
Accepted 24 March 2014
Available online 13 April 2014

Keywords:
Neutron
Alpha spectrometry
Silicon detector
Monte Carlo simulation

ABSTRACT

The response of a semiconductor alpha detector to fast (> 1 MeV) neutrons was investigated by using measurements and simulations. A polyethylene converter was placed in front of the detector to register recoil protons generated by elastic collisions between neutrons and hydrogen nuclei of the converter. The developed prototype equipment was tested with shielded radiation sources. The low background of the detector and insensitivity to high-energy gamma rays above 1 MeV are advantages when the detection of neutron-emitting nuclear materials is of importance. In the case of a ^{252}Cf neutron spectrum, the intrinsic efficiency of fast neutron detection was determined to be 2.5×10^{-4}, whereas three-fold greater efficiency was obtained for a ^{241}AmBe neutron spectrum.

1. Introduction

Prototype equipment was recently developed to detect and identify alpha-particle-emitting radionuclides from flat and smooth sources at ambient air pressure (Pöllänen et al., 2012). Good energy resolution was obtained using a solid-state alpha detector with alpha-particle collimation. Although the equipment was primarily designed for in situ alpha spectrometry, it can be applied to neutron detection if a suitable converter is placed in front of the detector.

The response of an alpha detector to photons and electrons/beta particles has previously been investigated (Pöllänen and Siiskonen, 2013). The photon/electron counts were observed in the low-energy part of the energy spectrum, i.e. below approximately 1 MeV, and the counts generated by these quanta are therefore usually well separated from those caused by alpha particles.

From the perspective of in-field applications, such as the detection of special nuclear materials out of regulatory control (Runkle et al., 2010; Peerani et al., 2012), it is also important to know the detector response to neutrons. Possible neutron reactions in the detector and in the surrounding materials may cause extra counts in the alpha-particle energy spectrum and may impair spectrum unfolding. However, if alpha particles are blocked using a thin hydrogen-rich polyethylene cover in front of the detector, it is possible to obtain indications of fast neutrons using elastic neutron scattering from protons. Although a low detection efficiency is anticipated, the detection of fast neutrons may benefit measurements in the field, especially when no other neutron detectors are available and when shielded neutron-emitting nuclear materials are suspected.

In the present paper, we investigate the response of a semiconductor alpha detector to neutrons using Monte Carlo simulations and measurements from calibrated ^{252}Cf and ^{241}AmBe sources. For testing we also used high-activity shielded neutron sources to verify the measurement concept in the field. Nuclear materials are here of importance because they may emit neutrons through spontaneous fission, although alpha-induced neutron emissions are also possible. Even thick layers of attenuating

* Corresponding author. Tel.: +358 9 75988425; fax: +358 9 75988433.
E-mail address: roy.pollanen@stuk.fi (R. Pöllänen).

http://dx.doi.org/10.1016/j.apradiso.2014.03.031
0969-8043/

materials, which can absorb high-energy gamma rays to a large extent, cannot necessarily block all neutrons. Thus, neutron signals, e.g. from an orphan source, may indicate the presence of special nuclear material. Here, we mainly concentrate on fast (>1 MeV) neutrons.

2. Principles of fast neutron detection using a silicon semiconductor alpha detector

In a medium, the main energy-loss mechanism for neutrons generated by fission is elastic scattering. In laboratory coordinates, the energy transfer in an elastic collision between a neutron of mass m_n and nucleus of mass M_r is as follows:

$$E_r = E_n \frac{4M_r m_n}{(M_r + m_n)^2} \cos^2\phi, \qquad (1)$$

where E_r and E_n are energies of the recoil nucleus and incoming neutron, respectively, and ϕ is the angle between the incoming neutron path and that of the recoil nucleus. In an ideal case, when the angle $\phi = 0$ and when the target nucleus is hydrogen ($E_r = E_n$), the kinetic energy of the incoming neutron is transferred to the kinetic energy of a proton. If the recoil protons are detected at a well-defined scattering angle, the energies of the incoming neutrons may be deduced.

A number of scientific papers have recently been published in which silicon-based detectors with recoil radiators have been applied in fast neutron measurement (see, for example, Agosteo et al. (2003), Zaki Dizaji et al. (2014), Flammang et al. (2007) and references therein). In the present study, a thin (1.5 mm) layer of polyethylene, $n(CH_2)$, was used in front of the active layer of the detector to convert neutrons to recoil protons (Fig. 1). The converter was kept in contact with the supporting structure of the detector, and protons with practically all scattering angles between $-90°$ and $90°$ were detected. Therefore, one-to-one correspondence between the detected proton energy and the energy of the incoming neutron was lost.

In addition to hydrogen and carbon in polyethylene, neutron interactions with silicon and other materials surrounding the detector have an influence on the detector response. Natural silicon is composed of three stable isotopes with the following atom percentages: ^{28}Si (92.2%), ^{29}Si (4.7%) and ^{30}Si (3.1%). In a head-on collision between a neutron and a ^{28}Si nucleus, the maximum fraction of kinetic energy that the neutron can lose is 13.4%, which is also the maximum kinetic energy of the recoil nucleus. For example, the recoil nucleus of ^{28}Si may receive kinetic energy of 100 keV if the incoming neutron energy is 0.8 MeV. This is the most probable value in the fission spectrum of ^{252}Cf. When the incoming neutron energies are 2.3 MeV (average value) and 6 MeV, the recoil nuclide maximum kinetic energies are of the order of 300 keV and 800 keV, respectively. All these are in the energy area in which photons, beta particles and electrons produce counts that may mask the counts generated by recoil nuclei (Pöllänen and Siiskonen, 2013). The main inelastic neutron interactions in natural silicon within the energy range considered in this work are (n,p) and (n,α) reactions (Table 1).

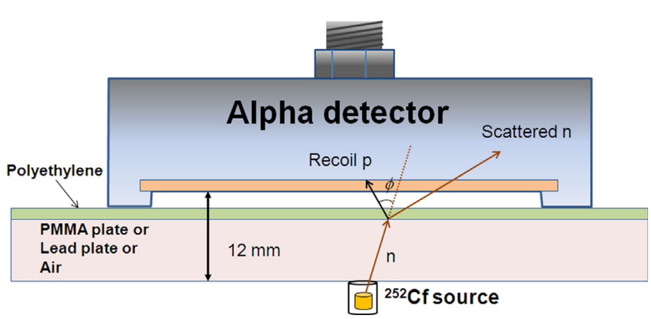

Fig. 1. Measurement geometry for detecting recoil protons from a ^{252}Cf source. In all measurements, a polyethylene converter of thickness 1.5 mm was placed in front of the detector. The distance from the surface of the converter to the active volume of the detector was 1 mm. The thickness of the PMMA/Pb plate/air layer was 1 cm, and the total source–detector distance was approximately 12 mm.

Table 1
The main inelastic reactions of fast neutrons in silicon (Schnöller, 1984). E_{th} refers to the threshold energy and σ_{ave} is the average cross section for the reaction in a fission neutron spectrum. The daughter nuclide half-life is $t_{1/2}$, and $E_{\beta,max}$ and E_γ refer to the most probable maximum energy of beta particles and gamma rays, respectively. Isotopes ^{25}Mg and ^{26}Mg are stable.

Interaction	E_{th} (MeV)	σ_{ave} (mbarn)	$t_{1/2}$ (min)	$E_{\beta,max}$ (keV)	E_γ (keV)
^{28}Si(n,p)^{28}Al	4.0	2	2.25	2863 (100%)	1779 (100%)
^{29}Si(n,p)^{29}Al	3.0	0.56	6.6	2406 (90%)	1273 (91%)
^{30}Si(n,p)^{30}Al	8.0	0.05	0.06	5062 (67%)	2235 (65%)
^{28}Si(n,α)^{25}Mg	2.75	0.56	–	–	–
^{29}Si(n,α)^{26}Mg	0.03	7.9	–	–	–
^{30}Si(n,α)^{27}Mg	4.3	0.07	9.5	1767 (71%)	844 (72%)

3. Determination of the detector response

An alpha detector (Canberra CAM450AM) with a nominal active area of 450 mm^2 was applied to detect charged particles generated in the converter or in the detector. CAM series detectors are commonly used in continuous air monitoring to identify the presence of alpha-particle-emitting radionuclides in air filters. The thickness of the detector active area was 300 μm, which is sufficient to stop all recoil protons with energy <6 MeV, and that of the dead layer 1.5 μm equivalent silicon. The junction area, i.e. the active area of the detector from which the radiation information from neutrons can be obtained, was 531 mm^2. The number of channels used in the measurements was 1024 (Amptek 8000A Pocket MCA) and the bias voltage was $+70$ V. The manufacturer of the detector recommends this voltage for alpha and beta detection.

Two different ^{252}Cf sources (activities 14650 Bq and 15.7 MBq) were used to investigate the detector response. The former source was used to illustrate the detector response using the same measurement geometry as was applied when investigating the detector response to different gamma-ray/beta particle emitters (Pöllänen and Siiskonen, 2013). The latter source was used to determine the detector efficiency for fast neutrons. All measurements were performed at ambient air pressure in a laboratory where the radon concentration in air is of the order of 1 Bq m^{-3}. However, this concentration may vary, mainly depending on the ventilation rate. The detector background is usually very low, i.e. less than 1 count per hour at energies higher than 1 MeV.

In addition to air, polymethyl methacrylate (PMMA) and lead plates with a thickness of 1 cm were placed between the ^{252}Cf source of lower activity and the detector to investigate the effect of an absorbing material (see Fig. 1). Although with these materials there are differences in the detector response to photons and electrons, their thicknesses do not have a marked influence on the recoil proton energy distribution (Fig. 2). To compare the neutron response to that caused by gamma rays/electrons, a set of point sources (^{22}Na, ^{60}Co, ^{137}Cs and ^{152}Eu; these nuclides gave a notable response in Pöllänen and Siiskonen (2013)) was measured together. In Fig. 2, practically all counts locating at channels higher than approximately 40 (referring to the energy of approximately 1 MeV) are either from recoil protons originating from the polyethylene plate or generated by neutron-induced interactions in the active volume of the detector or materials surrounding it.