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Neutron producing reactions in PuBe neutron sources

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ABSTRACT

There are a plenty of out-of-use plutonium–beryllium neutron sources in Eastern Europe presenting both nuclear safeguards and security issues. Typically, their actual Pu content is not known. In the last couple of years different non-destructive methods were developed for their characterization. For such methods detailed knowledge of the nuclear reactions taking place within the source is necessary. In this paper we investigate the role of the neutron producing reactions, their contribution to the neutron yield and their dependence on the properties of the source.

1. Introduction

PuBe neutron sources have been produced over decades. Despite their many advantages, their production stopped in the eighties because of their nuclear material content that represents nuclear safeguards and security issues. However, even today there are hundreds, if not thousands, of such sources in Eastern Europe, mostly out of use.

PuBe (α,n) isotopic neutron sources contain Pu and Be those compose an intermetallic compound. The active core is encapsulated in an inner welded, hermetically sealed capsule. The inner capsule is placed in an outer hermetically welded capsule. Capsule material is stainless steel [1]. For manufacturing details see Fig. 1.

The plutonium isotopes and ^{241}Am emit alpha radiation of various energies and the alpha particles interact with Be producing neutrons through (α,n) reactions. This is the primary source of neutrons in such neutron sources. However other reactions, such as $(n,2n)$ on Be and neutron-induced fission of Pu isotopes can take place in the sample, contributing to the total neutron yield. The spontaneous fission of Pu plays a negligible role in neutron production [2].

For determining the Pu content of PuBe sources different non-destructive (NDA) methods were developed. Details of these procedures are discussed in Refs. [2–5].

The total Pu content and the amount of the individual Pu isotopes could be calculated from the combination of the alpha activities of the Pu isotopes and the isotopic composition. The latter can be determined by high resolution gamma spectrometry. However, neither the alpha activities of the individual isotopes nor their sum can be measured directly, but can be calculated – for instance – from the neutron output. Then the Pu content can be calculated from the neutron output and the isotopic composition, relying on specific neutron yields (g_i), which are the products of the specific alpha activity (A_i) and the probability of (α,n) reaction (y_i) for the individual Pu isotopes and ^{241}Am, as:

$$m_{Pu} = \frac{N}{M\sum_i f_i g_i} \tag{1}$$

where N is the neutron output, M is the multiplication in the source due to secondary (neutron-induced) reactions, f_i is the abundance of the i-th (Pu and Am) isotope, g_i is the specific neutron yield of the i-th isotope.

This method for determination of Pu content is called combined method [2]. The neutron output can be measured, for instance, by a calibrated neutron coincidence counter, that can provide not only the total number of detected neutrons but also the rate of time correlated neutrons (doubles and triplets) that carries information about the rate of secondary reactions. The isotopic composition is determined from the gamma spectrum using the evaluation code MGA++ [6]. The specific alpha activities are well known, but the probability of (α,n) reaction, or, by other words, the alpha-to-neutron conversion factors (y_i) should be determined.

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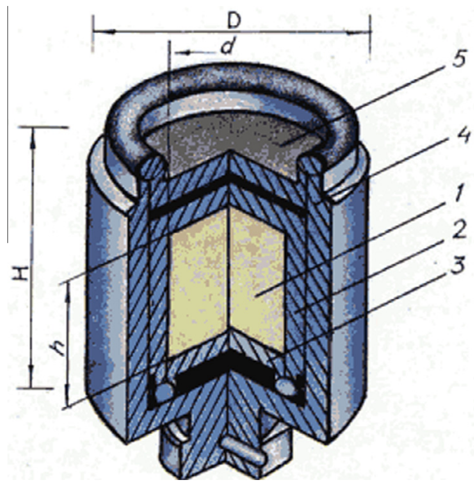

Fig. 1. Manufacturing details: active core (1), inner welded, hermetically sealed capsule body: (2), cover: (3), outer hermetically welded capsule body: (4), cover: (5).

The plutonium isotopes emit α-particles with different energy and intensity (see Table 3), which are slowing down in the material losing their energy continuously and producing neutrons in parallel. The cross section of (α,n) reaction is a sensitive function of alpha energy and the number of neutrons produced by alpha particles depends both on the initial alpha-energy and the stopping power of the material. Therefore the individual y_i conversion factors for all Pu isotopes and ^{241}Am should be known for the determination of the Pu content. These conversion factors can be calculated either theoretically or determined empirically. Both of them were carried out and results are compared in this paper.

Former neutron coincidence measurements already proved the presence of secondary reactions in PuBe sources [7]. Their role and contribution to the neutron output was also investigated by MCNP simulation and the results are summarized in this paper.

2. Theoretical calculation

2.1. (α,n) reaction

The slowing down of the alpha particles can be described by the material's stopping power (SP):

$$\mathrm{SP}(E) = -\frac{dE}{dx}. \tag{2}$$

and the stopping cross section (ε)

$$\varepsilon = -\frac{1}{N}\frac{dE}{dx}. \tag{3}$$

where N is the total atom density of the material. In the case of mixture composed of different elements the stopping cross section can be calculated by the Bragg–Kleeman relationship [8]:

$$\varepsilon(E)_{mix} = \frac{1}{N}\sum_i \varepsilon(E)_i n_i \tag{4}$$

where n_i is the atom density of the i-th constituent.

As first approximation, we considered the source material as $PuBe_{13}$ intermetallic compound. In this case the fraction of the constituents is well defined, and its stopping power can be calculated from those of Be and Pu found in [9,10]. The stopping cross sections of Be and Pu and their calculated value for $PuBe_{13}$ are indicated as a function of energy in Fig. 2.

While the alpha particles are stopping down, neutrons may be produced by the following reactions:

$$^{9}\mathrm{Be} + \alpha \rightarrow {}^{13}\mathrm{C}^{*} \rightarrow {}^{12}\mathrm{C} + n + 5.71\ \mathrm{MeV} \tag{5}$$

$$^{9}\mathrm{Be} + \alpha \rightarrow {}^{8}\mathrm{Be} + \alpha + n - 1.67\ \mathrm{MeV} \tag{6}$$

The cross sections of these reactions as a function of energy are indicated in Fig. 3.

The probability of neutron production in PuBe material can be given by the following equation [11]:

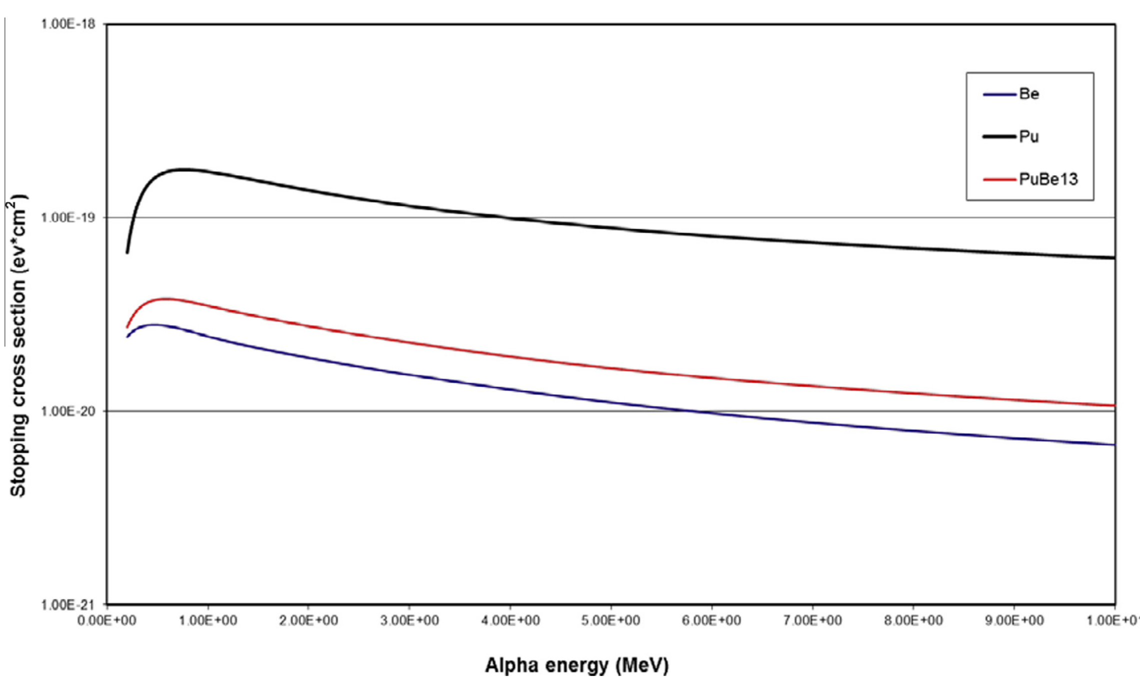

Fig. 2. The stopping cross section of Be, Pu, and $PuBe_{13}$ as a function of alpha-energy.