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# The *ideal* neutron energy spectrum of $^{241}AmLi(\alpha,n)^{10}B$ sources

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### HIGHLIGHTS

► A new Monte Carlo based method is applied to obtain the unperturbed (at birth)<sup>241</sup>AmLi spectrum.

- ► Various parameters that affect photon and neutron yields and energy distributions are investigated.
- ► The µ-particle size of neutron sources is investigated and deduced for an actual source.

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# ABSTRACT

The pure and unperturbed (*ideal*, at birth) spectra of <sup>241</sup>AmLi( $\alpha$ ,n)<sup>10</sup>B sources were determined using Monte Carlo simulations, along with the neutron spectrum measured outside a source's encapsulation. Various parameters that affect photon and neutron yields and energy distributions were investigated. The source's microparticle size was deduced. Subsequently, the calculated theoretical neutron energy spectra of the <sup>241</sup>AmLi( $\alpha$ ,n) reaction were compared to both the *ideal* spectra and the calculation of Geiger and van der Zwan predominantly used since early 1970s.

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#### 1. Introduction and Scope

Owing to its low mean energy which is about 450 keV well below the <sup>238</sup>U fission threshold, the <sup>241</sup>AmLi( $\alpha$ ,n)<sup>10</sup>B reaction is widely used in active non-destructive assay (NDA) techniques for nuclear and non-proliferation safeguards (e.g. active well neutron coincidence counters) as well as in fluence calibration of neutron monitors, neutron dosimetry and spectrometry amongst others. In all such applications, the instruments and their response functions are often modelled using Monte Carlo simulations, which when validated by measurements, provide sound and reliable calibration methods, as shown for instance in Tagziria and Peerani (2009), Peerani and Looman (2002), Peerani et al. (2008) and Croft et al. (2011) and references within. The main source of uncertainty often depends on the accurate knowledge of the energy spectrum outside the source encapsulation, which is strongly affected by the encapsulation materials and by the

different materials that make up the source, such as hydrogen, lithium, oxygen and possibly beryllium contamination (signalled by the presence of 4.3 MeV  $\gamma$ -rays). Furthermore, the microparticle size of the oxygen (<sup>17</sup>O and <sup>18</sup>O) crystals are shown (Vlaskin and Chvankin, 1993) to influence the photon and neutron yields as well as the neutron energy distribution spectra of <sup>241</sup>AmLi sources. The <sup>17,18</sup>O( $\alpha$ ,n) neutron distribution extends beyond 1.5 MeV where  ${}^{241}$ AmLi( $\alpha$ ,n) neutrons die out well above the <sup>238</sup>U fission threshold. Thus the most often used Geiger and van der Zwan (1971) calculated spectrum, although adequate in some applications, provided one knows the encapsulation details, has its limitations, especially at low ( < 100 keV) energies. The latter spectrum calculations are based on measurements of the <sup>7</sup>Li( $\alpha$ ,n)<sup>10</sup>B differential cross-section, by the same authors (van der Zwan and Geiger, 1972) and on published stopping power data then available for  $\alpha$ -particles in the source material.

Early direct measurements of the source spectrum have been carried out by Werle (1970), Bennett (1965), Weaver et al. (1982) and Trykov et al. using different methods. However these often lacked details and failed especially to address the energy region below about 100 keV in general due to experimental limitations

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such as inadequate gamma-neutron discriminations and noise interference at these energies. In active well coincidence counting (AWCC or UNCL) and other applications the little known lower energy part of the spectrum represents a real source of uncertainty.

More recently, the neutron and photon energy spectra of two different <sup>241</sup>AmLi( $\alpha$ ,n)<sup>10</sup>B radionuclide sources were extensively investigated and measured by Tagziria et al. (2003, 2004), which resulted in reliable good quality neutron energy spectra. These spectra are as measured outside the source's encapsulation and thus as such are source specific. There are many different <sup>241</sup>AmLi sources used in different laboratories, each with its characteristics, encapsulation, geometry, <sup>241</sup>AmO<sub>2</sub>, Li–H content etc. Having investigated in this work and in references by Tagziria et al. (2003, 2004) the various parameters (material compositions, micro-particle size, etc.) that affect the neutron and photon spectra of the sources, we have shown that their energy distributions outside the encapsulation (as actually used) and their fluence rates can be very different from one source to the other. The effect of microparticle sizes was also first studied by Trykov et al.. These effects make it more difficult than is for instance the case for <sup>252</sup>Cf and <sup>241</sup>Am-Be sources (International Organization for Standardization, 1989) to recommend a unique energy spectrum outside all capsules for <sup>241</sup>AmLi. For, neither the Cf nor the AmBe source has for instance any other elements or contaminants which can severely alter the characteristics of their neutron spectra especially regarding the absence of hydrogenous isotope within them.

In applications where the source spectra are used in Monte Carlo simulation of counting systems, the source spectrum model, hereafter called the *ideal* spectrum, should preferably be that of the pure <sup>241</sup>AmLi( $\alpha$ ,n) reaction neutrons, as they originate (at birth) within the source and unperturbed by the encapsulation materials and all the other source materials. The engineering design of the capsule and the details of the source material must be well known in order to be correctly modelled. A technique based on Monte Carlo simulation and measured spectra, combined with a response matrix inversion procedure, is developed in this work and used to extract the *ideal* spectrum of the <sup>241</sup>AmLi. The technique is described in Section 3 and applied to the NPL and

JRC sources thereafter. Details of the measurements and their analysis are reported in references by Tagziria et al. (2003, 2004), where other aspects such as the anisotropy effects on the neutron fluence of the source and the gamma spectrometry are also studied.

## 2. <sup>241</sup>AmLi measured spectra

The neutron and photon energy spectra of two rather different (in size, geometry and composition)  $^{241}$ AmLi( $\alpha$ ,n) $^{10}$ B radionuclide sources were measured at the National Physical Laboratory (NPL) in the UK (Tagziria et al., 2003) and independently under contract to JRC at the PTB in Germany (Tagziria et al., 2004) using a variety of spectrometry systems (Alevra and Thomas, 2003; Tagziria and Hansen, 2003; Shalev and Cutler, 1973; Klein and Neumann, 2002), and unfolding techniques (MAXED, MIEKE etc). These two extensive measurement campaigns resulted in the neutron energy spectra, shown in Figs. 1–3. The inset in Fig. 3 is the spectrum of the JRC source in linear energy scale. The theoretical



**Fig. 2.** Spectrum of the JRC <sup>241</sup>Am–Li neutron source measured at the PTB (Tagziria et al., 2004)—plotted as fluence per unit lethargy.



Fig. 1. Spectrum of the <sup>241</sup>Am–Li source measured by Tagziria et al. (2003) at the NPL compared to the calculations of Geiger and van der Zwan—plotted as fluence per unit lethargy.

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