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Radiogenic neutron yield calculations for low-background experiments

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ABSTRACT

Nuclear recoil backgrounds are one of the most dangerous backgrounds for many dark matter experiments. A primary source of nuclear recoils is radiogenic neutrons produced in the detector material itself. These neutrons result from fission and (α, n) reactions originating from uranium and thorium contamination. In this paper, we discuss neutron yields from these sources. We compile a list of (α, n) yields for many materials common in low-background detectors, calculated using NeuCBOT (Neutron Calculator Based On TALYS), a new tool introduced in this paper, available at https://github.com/shawest/neucbot. These calculations are compared to computations made using data compilations and SOURCES-4C.

1. Introduction

Neutron backgrounds are often considered among the dominant backgrounds in many low-background experiments, such as neutrino-less double β-decay searches and direct dark matter detectors.

Neutrino-less double β-decay experiments generally rely on having a high energy resolution and very little background near the endpoint of the double β-decay spectrum. While experiments generally take great pains to avoid having radioactive contaminants that produce a signal in the relevant energy range, some is inevitably always present. When these contaminants undergo nuclear reactions, they may create fast neutrons. These neutrons or the γ-rays they produce when they capture or inelastically scatter may create a signal in the energy region of interest. For example, EXO includes signals produced by neutron captures on ^{136}Xe, ^{1}H, ^{65}Cu, and ^{63}Cu among their dominant backgrounds [1]. Neutron-induced backgrounds such as the ^{76}Ge$(n,\gamma)^{77}$Ge reaction are considered important potential backgrounds in GERDA [2], and neutron captures on ^{1}H are expected to be among the primary backgrounds in SNO+ [3].

In dark matter experiments looking for Weakly Interacting Massive Particles (WIMPs), electromagnetic backgrounds such as γ-rays and β-decays produce electron recoils, while neutrons and nuclei may result in nuclear recoils. Since WIMPs are also expected to produce nuclear recoils, techniques that discriminate between electron and nuclear recoils are very effective at removing backgrounds. However, neutron-induced nuclear recoils remain an important background for these experiments, since they can produce a signal similar to what is expected from WIMPs [4–8].

Nuclear recoil backgrounds result from α-decays on the inner surface of the detector ejecting a nucleus into the active volume of the detector or from neutrons scattering in the active volume. Neutrons may be cosmogenic in origin if they are produced by cosmogenic muons interacting with the environment through processes such as spallation, or they may be radiogenic, primarily resulting from spontaneous fission of ^{232}Th, ^{238}U, and ^{235}U, or from the (α, n) reaction occurring when α-particles produced in these decay chains interact with nuclei in the material they are traveling in.

Cosmogenic neutrons are typically mitigated by moving a detector deep underground, where the muon flux is greatly reduced. External muon vetoes, as described in [9], can be used to tag muons coincident with nuclear recoils produced by neutrons. A detailed discussion of these backgrounds is provided in [10]. The focus of this document will be radiogenic neutrons.

Radiogenic neutrons result from nuclear interactions within a given material. While they can be reduced with careful material selection, some amount is always present. These neutrons may scatter once in the sensitive volume of a detector and then leave, producing a signal identical to that expected from a WIMP. While external veto systems, such as the design discussed in [9,11], may be able to tag these events, a quantitative description of them is necessary for any low-background experiment to understand and minimize its backgrounds.

2. Decay chains and secular equilibrium

In this discussion, we will focus on three decay chains: ^{232}Th, ^{238}U, and ^{235}U. Isotopes in these chains are expected to produce

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most of the radiogenic neutron backgrounds present in most low-background experiments. Typically, these experiments strive to reduce their contamination of these isotopes through screening campaigns, such as γ-ray spectroscopy, as discussed in [12], glow-discharge mass spectrometry, as discussed in [13], and other techniques.

We will assume secular equilibrium in these chains, with a few exceptions. Secular equilibrium may be broken where a long-lived gaseous isotope can emanate from a material, where manufacturing or purification processes may selectively remove some isotopes, where the material may have been exposed to elements partway down a decay chain, such as may be the case for radon, or where other chemical processes may differently affect elements with different chemical properties, such as solubility in water. If these isotopes or their precursors have half-lives longer than the scale of the experiment, it may not be appropriate to assume secular equilibrium for the entire decay chain. In these cases, we will divide the chains into sub-chains in which we expect secular equilibrium to be preserved with respect to the top of the sub-chain.

In particular, ^{238}U has a half-life of 4.5×10^{9} years and decays to ^{226}Ra, with a half-life of 1600 years, through four intermediate isotopes. Since ^{234m}Pa, one of the precursors of ^{226}Ra, emits a 1 MeV γ-ray that can be measured experimentally and ^{226}Ra has a 186 keV γ-ray, we split the ^{238}U decay chain into an upper and lower chain, defining all isotopes before ^{226}Ra to be in the upper chain, and ^{226}Ra and its progeny to be in the lower chain. While the decision to break the chain here is partly practical, one possible explanation is that radium is an alkaline earth metal, while its precursors are all actinides. Since these two groups have different chemical properties, it is possible that some chemical processes can affect these elements differently. Since ^{226}Ra has a very long half life, secular equilibrium will remain broken if ^{226}Ra is taken out of equilibrium with its precursors. Furthermore, since ^{210}Pb has a half-life of 22.2 years, far longer than its precursors, excess ^{210}Pb may accumulate in materials due to ^{222}Rn exposure in the air, causing secular equilibrium to be broken once again. Therefore, we account for the ^{210}Pb decay chain, consisting of ^{210}Pb and its progeny.

Throughout this document, we will discuss neutron yields in terms of neutrons produced per decay of the top of the decay chain. For a decay chain in secular equilibrium, this includes neutrons produced by all of the isotopes in this chain, weighted by the relevant branching ratios.

3. Direct neutron emission and spontaneous fission

While the primary focus of this document will be on neutrons produced by the (α,n) reaction, it is worth drawing attention to two other processes that produce radiogenic neutrons: direct neutron emission and spontaneous fission. The rates of both of these reactions depend only on the amount of uranium and thorium present in each detector component, and not on the material in which the contamination is present. In materials with particularly low (α,n) cross sections, these other processes may contribute significantly to the neutron background. We therefore discuss these radiogenic neutron sources for comparison.

^{214}Bi α-decays to ^{210}Tl with a branching ratio of 0.021%, which then β-decays to ^{210}Pb. There is a 0.007% chance that this β-decay will go to an excited state of ^{210}Pb, which decays by emitting a 200–260 keV neutron to ^{209}Pb [14]. Due to these branching ratios, we expect to see these neutrons in $\sim1.5\times10^{-8}$ of all decays of the lower ^{238}U chain.

Heavy nuclei that ordinarily α-decay may instead fission into smaller nuclei. When this happens, many particles may be ejected as well, including several MeV-scale γ-rays and some number of neutrons. The distribution of the number of γ-rays produced is discussed in [15], and the distribution of the number of neutrons emitted is discussed in [16]. These studies showed that the number of neutrons emitted in the spontaneous fission of ^{238}U can be modeled by a Gaussian distribution with a cutoff at 0, a mean of 2.05±0.04, and a standard deviation of 1.04±0.03.

These fission reactions are described in Table 1, which summarizes the spontaneous fission branching ratio BR_{SF}, the mean neutron kinetic energy $\langle E\rangle$, the mean neutron multiplicity $\langle\nu\rangle$, and the total number of neutrons produced per second per Becquerel of each decay chain for each isotope in these chains that may undergo spontaneous fission, as calculated using the SOURCES-4C code [17].

Table 1
Spontaneous fission branching ratios, mean neutron energies in MeV, mean neutron multiplicities, and neutron yields in n/s/Bq, calculated using SOURCES-4C [17].

Chain	Iso.	BR_{SF}	$\langle E\rangle$	$\langle\nu\rangle$	Yield
^{232}Th	^{232}Th	1.80×10^{-11}	1.60	2.14	3.85×10^{-11}
^{238}U	^{238}U	5.45×10^{-7}	1.69	2.01	1.10×10^{-6}
	^{234}U	1.64×10^{-11}	1.89	1.81	2.97×10^{-11}
	^{230}Th	3.8×10^{-14}	1.71	2.14	8.13×10^{-14}
^{235}U	^{235}U	7.00×10^{-11}	1.89	1.86	1.30×10^{-10}
	^{231}Pa	1.60×10^{-13}	1.93	1.93	3.09×10^{-13}

4. (α,n) neutrons

The (α,n) reaction occurs predominantly in low-to-mid-Z materials with contamination from α-emitting isotopes. When these isotopes decay, the emitted α particle may capture on another nucleus in the material to form a compound nucleus, which may decay by neutron emission. For the calculations discussed here, we consider a thick target in which the α particle captures in the same material in which it was produced. Calculations of the neutron yield from the (α,n) reaction (*i.e.*, the (α,n) yield) therefore depend on the energy spectra of α-decays and the elemental and isotopic composition of the material. These calculations also depend on the stopping power of α particles of a given energy in the material as well as the (α,n) cross sections and the structure of involved nuclei.

While these neutrons are sometimes accompanied by a γ-ray, either correlated with the decay of the α-emitter or from the relaxation of the final nucleus, neutrons are often produced alone. This possibility makes (α,n) neutrons particularly troublesome backgrounds, as there may be no accompanying signal to help tag them. The rest of this document will therefore be focused on calculating (α,n) yields, including the introduction of NeuCBOT as a tool for calculating these yields (see Section 4.2). In order to benchmark NeuCBOT against other standards, we will calculate (α,n) yields for several materials using NeuCBOT and compare these values to calculations performed using measured yields on individual isotopes (see Section 4.3) as well as yields and neutron energy spectra predicted by SOURCES-4C (see Section 4.4).

4.1. Materials considered

The (α,n) reaction rate and neutron energy spectrum depend on both the energy of the emitted α particle and the various nuclei with which it interacts, either through their contribution to the stopping power or through the (α,n) reaction itself. It is therefore important to define the chemical compositions of the materials for which we are calculating (α,n) yields.

We summarize the chemical compositions used for these calculations in Table 2. The same chemical and isotopic compositions were used for calculations performed using NeuCBOT, SOURCES-4C, and measured yields. Notably, SOURCES-4C requires that elemental and isotopic compositions be specified by the fraction of atoms of individual elements and isotopes in a material. The mass fractions given in Table 2 were therefore converted to isotopic fractions for SOURCES-4C calculations. For each element, we assume natural isotopic abundances as reported in [18].

Since the same material compositions were assumed for calculations performed using NeuCBOT, measured yields, and SOURCES-4C, uncertainties in material compositions do not affect the comparison between the three different methods. Nevertheless, in order to understand the uncertainties in the yields reported in this document, we discuss the