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Cosmogenic activation in germanium and copper for rare event searches

S. Cebrián *, H. Gómez, G. Luzón, J. Morales [1], A. Tomás, J.A. Villar

Universidad de Zaragoza, Pedro Cerbuna 12, 50009 Zaragoza, Spain

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ABSTRACT

Experiments looking for rare events like the direct detection of dark matter particles or the nuclear Double Beta Decay are operated in deep underground locations, to suppress or very effectively reduce the effect of cosmic rays; but cosmogenic activation produced at sea level in detectors and other materials can become a serious hazard for them. Copper and germanium are very frequently used in this kind of experiments requiring an ultra-low radioactive background and therefore have been chosen as activation targets in this work. First, the excitation functions for relevant induced long-lived radioactive isotopes have been estimated; special care has been taken in using those calculations giving the best agreement with measured production cross-sections and in distinguishing production by neutrons or by protons when relevant and possible. Then, the corresponding rates of production of the nuclides in natural and enriched (86% ^{76}Ge and 14% ^{74}Ge) germanium and copper have been evaluated considering two different cosmic ray spectra. Comparison of the obtained activation yields at surface with all the known previous results (based either on calculations or experiments) has allowed to draw conclusions on the general methodology for evaluating cosmogenic activation.

1. Introduction

Experiments searching for rare phenomena like the nuclear Double Beta Decay (DBD) or the interaction of Weakly Interacting Massive Particles (the so-called WIMPs, which could be filling the galactic dark halo) require detectors working in ultra-low background conditions. Operating in deep underground locations, using active and passive shields and selecting carefully radiopure materials reduce very efficiently the background for this kind of experiments [1,2]. In this context, long-lived radioactive impurities in the materials of the set-up induced by the exposure to cosmic rays at sea level (during fabrication, transport and storage) may be even more important than residual contamination from primordial nuclides and become very problematic.

One of the most relevant processes in the cosmogenic production of isotopes is the spallation of nuclei by high energy nucleons. At sea level, the flux of neutrons and protons is virtually the same at energies of a few GeV; however, at lower energies the proton to neutron ratio decreases significantly because of the absorption of charged particles in the atmosphere. For example, at 100 MeV this ratio is about 3% [3]. Consequently, nuclide production is mainly dominated by neutrons at the Earth's surface, but if materials are flown at high altitudes, in addition to the fact that the cosmic flux is much greater, protons will produce activation as well.

The production rate R of an isotope with decay constant λ by the exposure to a flux ϕ of cosmic rays can be evaluated as:

$$R \propto \int \sigma(E)\phi(E)dE \quad (1)$$

σ being the production cross-section and E the particle energy. For an exposure time t_{exp} and a cooling time t_{cool}, the induced activity A is:

$$A = R\left[1 - \exp(-\lambda t_{exp})\right] \exp(-\lambda t_{cool}) \quad (2)$$

Consequently, to estimate the cosmogenic induced activity in a material knowing its history (time and places of exposure to cosmic rays) there are two basic ingredients: the flux of cosmic rays and the cross-section of isotope production.

Materials must be kept shielded against the hadronic component of the cosmic rays to prevent cosmogenic activation and are frequently stored underground. Flying must be avoided and the exposure on the surface of the Earth should be reduced as much as possible. Since these requirements usually complicate the preparation of experiments (for example, crystal growth and mounting of detectors) it would be desirable to have reliable tools to quantify the real danger of exposing the materials to cosmic rays.

Different experiments and projects looking for rare events use germanium crystals either as conventional diodes (like IGEX [4], Heidelberg–Moscow [5], Majorana [6], GERDA [7] or TEXONO [8]) or as cryogenic detectors (like CDMS [9], EDELWEISS [10] or even

* Correspondence to: S. Cebrián, Facultad de Ciencias, Universidad de Zaragoza, Pedro Cerbuna 12, 50009 Zaragoza, Spain. Tel.: +34 976761243; fax: +34 976761247.
E-mail address: scebrian@unizar.es (S. Cebrián).

[1] Deceased.

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EURECA [11]). Copper is a material widely used in experiments, either as shield or part of the detector set-up; in addition, for copper there is a large amount of experimental data for production cross-sections, allowing a better validation of calculations. For these reasons, germanium and copper have been chosen as activation targets to be studied.

Therefore, the aim of this work has been to quantify the activation yields for the long-lived radioisotopes cosmogenically induced at surface in germanium and copper which are relevant in rare event searches. This was also the goal in some previous recent articles [12,13]; but in these cases evaluation of production cross-sections was mainly based on an unique approach, the SHIELD code in [12] or the TALYS code in [13]. For each one of the analyzed products in the selected targets the methodology followed in the activation studies presented here has been the following:

1. To collect all the available information on the isotope production cross-sections. Data, either from measurements or from computational calculations, have been searched for in different libraries of nuclear data and from individual references. Some specific new calculations of production cross-sections have been made too, i.e., using the YIELDX code described later.
2. To choose the best description of the excitation function of the product, considering all the information collected. An attempt to quantify the deviations between the available experimental cross-sections and the corresponding different calculations has been made, in order to try to find the most reliable estimates. Deviation factors F have been calculated following a definition typically used in the literature (see for instance [14]):

$$F = 10^{\sqrt{d}}, \quad d = \frac{1}{N}\sum_i (\log \sigma_{exp,i} - \log \sigma_{cal,i})^2 \qquad (3)$$

being N the number of pairs of experimental and calculated cross-sections $\sigma_{exp,i}$ and $\sigma_{cal,i}$ at the same energies. Parameter d is the mean square logarithmic deviation.
3. To estimate the production rate of the induced nuclei at sea level using the selected excitation functions $\sigma(E)$ and a particular cosmic ray spectrum $\phi(E)$, and to compare it with available previous results based either on measurements or calculations.

The structure of the paper is as follows. In Section 2 the selected excitation functions for the considered products are discussed, describing the sources of data taken into account including both measurements of production cross-sections and calculations using computational codes. The corresponding production rates are derived according to different assumptions on the cosmic ray spectrum and compared with previous estimates in Section 3. Finally, conclusions are summarized in Section 4.

2. Excitation functions

The excitation function for the production of a certain isotope by nucleons in a target over a wide range of energies (from some MeV up to several GeV) can be hardly obtained experimentally, since the measurements of production cross-sections with beams are long, expensive and there are not many available facilities to carry them out. The use of computational codes to complete information on the excitation functions is therefore mandatory. In addition, most measurements are performed on targets with the natural composition of isotopes for a given element, often determining only cumulative yields of residual nuclei. Reliable calculations are required to provide independent yields for isotopically separated targets in many cases. But in any case, experimental data are essential to check the reliability of calculations.

Experimental data can be found at several nuclear data libraries like EXFOR [15] and NUCLEX [16]. Concerning computational calculations there are two approaches: some codes implement semiempirical formulas for the isotope production cross-sections, while other ones are based on the complete Monte Carlo (MC) simulation of the interaction between nucleons and nuclei to derive the induced nuclides. The main advantage of the first approach is that the calculation time is much shorter while the advantage of MC codes is that they are applicable not only to proton-induced but also to neutron-induced nuclear reactions.

Most semiempirical codes are based on the formulas from Silberberg and Tsao [17–19]. The successive improvements in these equations have been incorporated in different codes like COSMO [20], Σ [21], YIELDX [19] and more recently ACTIVIA [22]. The formulas offer a wide coverage of targets and products but have been deduced from proton-induced reactions and are applicable only above $\sim$100 MeV, which can be an important limitation for some activation yields with a low energy threshold; they can be used for estimating neutron activation assuming that cross-sections for protons and neutrons are equal. YIELDX, including the latest updates of the semiempirical formulas [19], has been used in this work to perform semiempirical calculations.

There are many families of MC codes which can be used for activation studies (LAHET [23], CEM [24], HMS-ALICE [25], SHIELD [26], MARS [27], LAQGSM [28], INUCL [29], ABLA [30], GEM [31],... are just some examples). In addition, general-purpose simulation packages like GEANT4 [32], FLUKA [33] or MCNPX [34] having integrated some of these models are also useful. In this work libraries based on different MC calculations have been taken into account:

- Library from Ref. [35] gives excitation functions including available experimental data and calculated results using CEM95, LAHET, and HMS-ALICE codes for selected targets and products; energies up to 1.7 GeV are considered for both neutrons and protons.
- MENDL libraries [36] are based on several versions of the ALICE codes, containing excitation functions which cover a very wide range of target and product nuclides, either for neutrons or protons, for energies up to 100 MeV for neutrons (MENDL-2, using ALICE92) and 200 MeV for protons (MENDL-2P, using ALICE-IPPE).
- LA150 library [37] contains results up to 150 MeV, independently for neutrons and protons as projectiles, using for calculations HMS-ALICE.

Among all the isotopes which can be cosmogenically induced, only those long-lived nuclides with emissions in the regions of interest for rare event searches have been considered here. Germanium is used for investigating the neutrinoless DBD of ^{76}Ge, which should give a peak at the transition energy Q = 2039 keV [38], but other DBD channels of this nucleus produce a continuum spectrum up to Q and the signal from the direct detection of WIMPs is expected to concentrate in the low energy region up to some tens of keV.

2.1. Germanium

For neutrinoless DBD searches, only ^{60}Co ($T_{1/2}$ = 5.2 y) and ^{68}Ge ($T_{1/2}$ = 270.8 d) are relevant, but also ^{65}Zn ($T_{1/2}$ = 244.3 d), ^{63}Ni ($T_{1/2}$ = 100.1 y), ^{58}Co ($T_{1/2}$ = 70.9 d), ^{57}Co ($T_{1/2}$ = 271.8 d), ^{56}Co ($T_{1/2}$ = 77.3 d), ^{55}Fe ($T_{1/2}$ = 2.73 y) and ^{54}Mn ($T_{1/2}$ = 312.3 d) have been studied. Production of ^{68}Ge is also interesting in different contexts, like positron emission tomography used in medical diagnosis.