Study of cosmogenic activation in copper for rare event search experiments

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Abstract Rare event search experiments using germanium detectors are performed in underground laboratories to minimize the background induced by cosmic rays. However, the cosmogenic activation of cupreous detector components on the ground generates long half-life radioisotopes and contributes to the background level. We measured cosmogenic activation with 142.50 kg of copper bricks after 504 days of exposure at an altitude of 2469.4 m outside the China Jinping Underground Laboratory (CJPL). The specific activities of the cosmogenic nuclides produced in the copper bricks were measured using a low-background germanium gamma-ray spectrometer at CJPL. The production rates at sea level, in units of nuclei/kg/day, were 18.6 ± 2.0 for ^{54}Mn , 9.9 ± 1.3 for ^{56}Co , 48.3 ± 5.5 for ^{57}Co , 51.8 ± 2.5 for ^{58}Co , and 39.7 ± 5.7 for ^{60}Co . The measurement will help to constrain cosmogenic background estimation for rare event searches using copper as a detector structure and shielding material. Based on the measured production rates, the impact of the cosmogenic background in cupreous components of germanium detectors on the next generation CDEX-100 experiment was assessed with the expected exposure history above ground.

1 Introduction

Rare event search experiments, such as dark matter direct detection and neutrinoless double beta decay experiments, are conducted in deep underground laboratories with passive and active shields, and built with materials that are especially radiopure, to effectively reduce their intrinsic background [1–6]. Under ultra-low background conditions, the background induced in the detector's materials on the sur-

face could become even more prominent than that from the residual contamination of the primordial nuclides.

With high radiopurity and attractive mechanical properties, copper is widely used in the innermost shields or parts of detectors in rare event search experiments [3,4,7–10]. However, radioisotopes produced by cosmogenic activation during manufacturing, transport, and storage can increase the background level of copper. Among these cosmogenic nuclides, short-lived nuclides like ^{59}Fe and ^{48}V decay rapidly when stored in underground laboratories, while long-lived nuclides like ^{60}Co and ^{54}Mn remain and continue to contribute to the detector's background level [11,12].

The China dark matter experiment (CDEX), aiming at direct dark matter detection and the study of neutrinoless double beta decay of ^{76}Ge , operates p-type point-contact germanium detectors (PPCGe) at the China Jinping Underground Laboratory (CJPL) [7,13–16]. The copper used in the PPCGe detectors of the CDEX experiment leads to a crucial background contribution, which requires assessment due to cosmogenic activation in copper to establish the background model for the CDEX experiment. Although the production rates of cosmogenic activation in copper have been investigated via simulations and measurements, discrepancies exist among the results [17–21].

In this study, we present the measurements of the cosmogenic activation in copper samples after exposure to cosmic rays. Corresponding radionuclide production rates are also calculated by Monte Carlo simulations and compared with the measured results. Finally, we simulate the background spectra from cosmogenic radionuclides in the copper components of the PPCGe detector array used in the future CDEX experiment under the expected exposure history of detectors above the ground.

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Fig. 1 Schematic diagram of placement of OFHC copper bricks in spectrometer: right view (left) and top view (right)

2 Experiment and methods

Sixteen oxygen-free high thermal conductivity (OFHC) copper bricks (their chemical purities are greater than 99.995% with natural isotope abundance) with a total mass of 142.50 kg, $20 \times 10 \times 5$ cm in size, were exposed to cosmic rays and measured in this study. The cosmic-ray muon flux in the CJPL is suppressed by eight orders of magnitude compared to surface laboratories due to 2400 m rock overburden [22, 23]. These OFHC copper bricks have been housed in the CJPL for more than four years. The OFHC copper samples were measured to evaluate the residual cosmogenic radioactivity before activation on the surface with a low-background germanium gamma-ray spectrometer GeTHU at CJPL, with a background rate between 100 and 2700 keV of 0.510 ± 0.006 cts/kg/min [24]. To enhance the detection efficiency, the germanium detector of GeTHU was surrounded by these copper sample bricks, as shown in Fig. 1. The background spectrum was measured from 12 Aug 2019, to 28 Aug 2019, as depicted in Fig. 2; the characteristic gamma-ray peaks from cosmogenic radionuclides were not statistically significant and were lower than the minimum detectable activities (MDA) of GeTHU.

These copper bricks were exposed to cosmic rays for 504 days, from Aug. 31, 2019, to Jan. 16, 2021, at an altitude of 2469.4 m in the vicinity of CJPL. The specific activities of cosmogenic radionuclides inside these copper bricks were calculated by spectral analysis, where the net area of a peak was determined under the assumption of a linear background [25]. The detection efficiencies for concerned radionuclides were simulated by Geant4.10.06 [26–28]. The specific activities of cosmogenic radionuclides can be deduced by combining the production and decay rates,

$$A = R(1 - e^{-\lambda t}) + A_0 e^{-\lambda t}, \quad (1)$$

where A is the specific activity, and R denotes the production rate of a particular cosmogenic radionuclide; λ is the decay constant and t is the exposure time; A_0 is the specific activity before exposure. The specific activities of the cosmogenic radionuclides were unsaturated after 504 days of exposure

for those with long half-lives, such as ^{60}Co . We applied the exponential hypothesis shown in Eq. (1) with zero A_0 to calculate the production rates, as cosmogenic isotopes were not quantified in the pre-exposure measurement of the copper samples. The decay of cosmogenic radionuclides during the measurements was also considered, especially for those with short half-lives, such as ^{58}Co .

The cosmogenic production rate R can be calculated and simplified as

$$R = \int \frac{d\Phi(E)}{dE} \sigma(E) dE \approx \Phi_{tot} \int \frac{d\phi(E)}{dE} \sigma(E) dE \propto \Phi_{tot}, \quad (2)$$

where $\frac{d\Phi(E)}{dE}$ is the energy spectrum of cosmic rays, $\frac{d\phi(E)}{dE}$ is the energy spectrum normalized to an incident particle; $\sigma(E)$ is the cross section of cosmogenic activation for cosmic rays, and Φ_{tot} is the flux of cosmic rays. As the energy spectra of a neutron differs slightly by altitude within 20 km [29], its normalized energy spectra at different altitudes can be considered as a constant. Therefore, the cosmogenic production rates are directly proportional to the intensity of the cosmic-ray flux, which, at a given altitude, can be parametrized by an exponential [30],

$$\Phi_{tot,H} = \Phi_{tot,0} e^{\frac{p(H)-p(0)}{L}}, \quad (3)$$

where $\Phi_{tot,H}$ and $\Phi_{tot,0}$ indicate the flux of certain particles in the cosmic ray at altitudes H and sea level, respectively; $p(H)$ is the atmospheric pressure at altitude H [30, 31], and L is the typical absorption length for a certain cosmic-ray component. It is generally considered that the production rates of cosmogenic activations are dominated by neutrons, so the absorption length of the neutron is a suitable choice [21, 32]. Cosmogenic production rates are proportional to the neutron flux, and the same exponential correction can be applied to obtain the corresponding production rates at sea level,

$$R(H) = R(0) e^{\frac{p(H)-p(0)}{L}}. \quad (4)$$

For comparison, the cosmogenic production rates were also simulated by Geant4 with the physical list QGSP_INCLXX [27, 28, 33]. CRY-1.7 [34] provides the energy spectra of different cosmic-ray components (neutron, proton, muon, gamma, electron, and pion) at sea level for the Geant4 simulation. In this simulation, the cosmic particles, including the neutron, proton, muon, and gamma from CRY-1.7 were used by the event generator. We could count each product after interactions with the copper sample to calculate the production rate.

Fig. 2 Measured spectra of OFHC copper samples pre- (blue) and post-exposure (black) with a bin-size of 0.36 keV. Gamma-ray peaks induced by cosmogenic activations and primordial radionuclides are also indicated

3 Cosmogenic activation in copper

The copper bricks were measured during Jan. 16–30, 2021. The spectrum after exposure is also shown in Fig. 2, where the selected gamma-ray peaks of cosmogenic radionuclides are marked. In addition to the characteristic gamma-ray peaks from cosmogenic radionuclides, there also exist several gamma-ray peaks related to the primordial radionuclides and the 511 keV annihilation peak. Compared with the pre-exposure spectrum, the background level worsens due to the breakdown of the low-radon air system of the laboratory when measuring these bricks after exposure. GeTHU has a radon mitigation system flushing its sample space with the low-radon air (with boil-off nitrogen gas previously) [24]. Without flushing, the concentrations of radon and its progenies in the sample space increase, and the background rate in the energy region from 238 to 609 keV is higher, as shown in Fig. 2. However, these cosmogenic gamma-ray peaks are still significant enough to warrant calculation of their specific activities. Following gamma-ray spectral analysis, the specific activities of these cosmogenic radionuclides were calculated, and are listed in Table 1.

Table 1 Measured specific activities of cosmogenic radionuclides (with 1 σ uncertainty) in copper samples corresponding to the values on January 16, 2021. Gamma-ray peaks with better statistics were

Radionuclide	Half-life (day)	Peak energy (keV)	Specific activity (mBq/kg)
^{54}Mn	312.20	834.8	0.87 ± 0.10
^{56}Co	77.24	846.8	0.65 ± 0.08
^{57}Co	271.74	122.1	2.42 ± 0.28
^{58}Co	70.86	810.8	3.39 ± 0.17
^{60}Co	1925.28	1173.2	0.46 ± 0.07

Table 2 compares the cosmogenic production rates (obtained from Eq. (1) with $A_0=0$ and Eq. (4)), after altitude correction, with results from previous studies. The results of our measurements are generally consistent with those of our simulation with Geant4 (as shown in Table 2) except for ^{56}Co . The measured production rate of ^{57}Co was about 3% less than simulated, while that of ^{56}Co was 49% of the simulated result. Our measurements are relatively close to those of Ref. [19]. The discrepancies of production rates with the literature could be due to different cross-section databases, uncertainties of altitude correction, and the fluxes and energy spectra of cosmic rays at different experimental latitudes. Due to its short half-life, 77.2 days, ^{56}Co will decay rapidly when stored underground, resulting in a negligible background contribution. Thus, the difference between simulation and measurement is not a prominent obstacle to the assessment of the cosmogenic background of the underground experiments. Previous Geant4 simulation results [20] differ from this work mainly due to the differences of the physics lists “shielding” and “QGSP_INCLXX” in these two simulations.

4 Background assessment in PPCGe detector

The next phase of the CDEX experiment, CDEX-100, will operate about 100 kg of germanium detectors immersed in liquid nitrogen in a 1725-m³ cryostat at Hall C of the second phase of CJPL. The specific activities of cosmogenic radionuclides are calculated following their production rates and expected exposure history. The total exposure period is a combination of manufacturing and transportation above ground and storage underground. Their specific activities can be calculated as

$$A = R(0)e^{(p(H_1)-p(0))/L}(1 - e^{-\lambda t_{\text{manu}}})e^{-\lambda(t_{\text{trans}}+t_{\text{cool}})} + F \times R(0)e^{(p(H_2)-p(0))/L}(1 - e^{-\lambda t_{\text{trans}}})e^{-\lambda t_{\text{cool}}}, \quad (5)$$

where $R(0)$ is the measured production rate at sea level (Table 2); H_1 and H_2 are the altitudes of the factory and transportation above ground, respectively; t_{manu} , t_{trans} and

selected to determine specific activities when radionuclides had two or more gamma-ray peaks. Only statistical uncertainties are considered

Table 2 Production rates of cosmogenic radionuclides in copper at sea level (unit: nuclei $\text{kg}^{-1} \text{day}^{-1}$). Only statistical uncertainties are considered

	Method	Nuclides				
		^{54}Mn	^{56}Co	^{57}Co	^{58}Co	^{60}Co
This work	Measurement	18.6 ± 2.0	9.9 ± 1.3	48.3 ± 5.5	51.8 ± 2.5	39.7 ± 5.7
	Geant4	21.1	20.4	49.6	70.9	44.1
Breier et al. [21]	CONUS [35]	14	10	50	76	92
Zhang et al. [20]	Geant4	12.31	10.32	67.15	57.26	64.63
	ACTIVIA	30.00	20.13	77.45	138.06	66.12
	ACTIVIA ¹	14.32	8.74	32.44	56.61	26.28
Cebrian et al. [18]	MENDL+YIELDX ²	32.5	22.9	88.3	159.6	97.4
	MENDL+YIELDX ³	27.7	20.0	74.1	123.0	55.4
Baudis et al. [19]	Measurement	$13.3^{+3.0}_{-2.9}$	$9.3^{+1.2}_{-1.4}$	$44.8^{+8.6}_{-8.2}$	$68.9^{+5.4}_{-5.0}$	$29.4^{+7.1}_{-5.9}$
	COSMO	13.5	7.0	30.2	54.6	25.7
Laubenstein et al. [17]	Measurement	8.85 ± 0.86	9.5 ± 1.2	73.8 ± 16.7	67.9 ± 3.7	86.2 ± 7.6

¹ Calculation relies on cosmic neutron spectra from Gordon [36]

² Calculation relies on cosmic neutron spectra from Ziegler [37]

³ Calculation relies on cosmic neutron spectra from Gordon [36]

t_{cool} are the time of manufacture, transportation above ground and storage underground, respectively; and F describes the shielding effect for transportation with a steel or lead cover.

Each germanium detector unit has 0.96 kg of copper and a total of 91.4 kg of copper will be used for the germanium detector array, as shown in Fig. 3. For simplicity, we assume that the copper components followed the same exposure history as the detectors. The manufacturing time, transportation time and underground storage time are supposed to be three months, a half-month and one year, respectively. After three months of fabrication, the detectors will be transported with shielding, where cosmogenic activations could be reduced by a factor of 10 [9, 12], to the underground laboratory in a half-month. The manufacturing factory is selected to be at sea level, while the altitude of the whole transportation routine is selected to be 1500 m, which is the average altitude of Liangshan Prefecture, where CJPL is located. The cupreous components will be stored in CJPL for preparation work, with the decay of cosmogenic nuclides, the so-called cooling time, lasting about one year.

Importing the measured production rates of the cosmogenic activations, the considered activities in the simulation were obtained using Eq. (5) and the background contributions from cupreous components were estimated with a Geant4-based Monte Carlo framework called Simulation and Analysis of Germanium Experiment (SAGE) [38]. In the simulation, these cosmogenic radionuclides were distributed uniformly across all cupreous components, as shown in Fig. 3. The energy spectra of the different cosmogenic nuclides and the total background contributions were simulated, as shown in Fig. 4.

Fig. 3 Sketch of PPCGe detector array (left) and PPCGe detector unit (right) implemented in SAGE, together with its cupreous components, shown in orange. The PPCGe detector array is immersed in liquid nitrogen in the simulation

Under the above assumptions, the simulated background rates in the low-energy range (2–4 keV for dark matter search) and around 2039 keV (energy of neutrinoless double beta decay of ^{76}Ge) are 7.17×10^{-2} counts per keV per tonne per year (cpkty) and 1.82×10^{-3} cpkty, respectively. The CDEX-100 experiment aims at constructing a germanium detector array with ultra-low background rates, which should be less than 3650 and 0.1 cpkty for the 2–4 keV and around 2039 keV energy regions, respectively. The background contribution from copper can be ignored for dark matter search, while it is 2% of the total background budget for neutrinoless double beta decay research. The contribution of ^{57}Co dom-

Fig. 4 Simulated cosmogenic background spectra in 0–2.8 MeV region from copper components of germanium detectors for CDEX-100 experiment. Energy resolution is determined by the germanium detector used in CDEX-1B [14]

inates the cosmogenic background contributions less than 122 keV, and ^{54}Mn is dominant below 830 keV, as shown in Fig. 4. ^{60}Co prevails in the higher-energy region among these cosmogenic radionuclides, while only the cosmogenic background exists from ^{56}Co over 2.5 MeV. Among these cosmogenic nuclides, only ^{60}Co has a half-life over one year, which is difficult to significantly suppress through underground cooling. A feasible method to mitigate the contribution from ^{60}Co is to reduce the exposure time above ground, or to electroform the copper underground.

5 Summary

We investigated the cosmogenic activation in OFHC copper bricks. The specific activities of cosmogenic radionuclides in the exposed copper bricks at the altitude of 2469.4 m were measured with a low-background germanium gamma-ray spectrometer. The production rates of several long-lived cosmogenic radionuclides were calculated and compared with those of previous studies. The production rates at sea level, in units of nuclei/kg/day are 18.6 ± 2.0 for ^{54}Mn , 9.9 ± 1.3 for ^{56}Co , 48.3 ± 5.5 for ^{57}Co , 51.8 ± 2.5 for ^{58}Co , and 39.7 ± 5.7 for ^{60}Co , respectively. For comparison, we simulated their cosmogenic activation with Geant4, showing that the agreement between measurement and simulation was generally satisfactory. A general comparison with other published results is presented, showing discrepancies but also good agreement in some cases.

Based on the results of the production rates of cosmogenic radionuclides in copper, the cosmogenic background from copper components of detectors was simulated for the CDEX-100 experiment. The total background rates from cosmogenic nuclides are 7.2×10^{-2} cpkty and 1.8×10^{-3}

cpkty for the 2–4 keV and around 2039 keV energy regions, respectively. The cosmogenic background is dominated by ^{57}Co , ^{54}Mn and ^{60}Co after one year of cooling underground. Compared with the total background budget of CDEX-100, the cosmogenic contribution in copper components can be ignored for dark matter search in the region of 2–4 keV. Cosmogenic activation in germanium crystals, another crucial background source for CDEX-100, is being studied following the procedure established in CDEX-1B [12].

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Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

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