Fast-Neutron Activation of Long-Lived Nuclides in Natural Pb

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Abstract

We measured the production of the long-lived nuclides ^{207}Bi , ^{202}Pb , and ^{194}Hg in a sample of natural Pb due to high-energy neutron interactions using a neutron beam at the Los Alamos Neutron Science Center. The activated sample was counted by a HPGe detector to measure the amount of radioactive nuclides present. These nuclides are critical in understanding potential backgrounds in low background experiments utilizing large amounts of Pb shielding due to cosmogenic neutron interactions in the Pb while residing on the Earth's surface. By scaling the LANSCE neutron flux to a cosmic neutron flux, we measure the sea level cosmic ray production rates of 8.0 ± 1.3 atoms/kg/day of ^{194}Hg , 120 ± 25 atoms/kg/day ^{202}Pb , and $< 0.17 \pm 0.04$ atoms/kg/day ^{207}Bi .

Keywords: neutron activation, double-beta decay, lead, cosmic ray

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1. Introduction

An observation of neutrinoless double-beta decay ($0\nu\beta\beta$) is critical to understanding the neutrino's absolute mass scale and particle-antiparticle nature [1, 2, 3, 4, 5, 6, 7]. The signature of this rare nuclear decay process would be the detection of a mono-energetic line within the the material containing an isotope subject to this decay mode. The potential success of these experiments relies on the ability to mitigate backgrounds that would mask the rare

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signal of interest through a reduction of internal radioactive backgrounds and ability to shield the detectors from external sources of radioactivity. Trace levels of radioactive impurities from natural decay chain isotopes within the detector and shielding components have been the limiting backgrounds in previous experiments. The γ -ray emissions from internal and external backgrounds can overwhelm the potential $0\nu\beta\beta$ signal by producing a continuum of energetic deposits inside the detector. Ongoing and future experiments continue to identify and reduce potential backgrounds to the signal of interest. A background level goal of 1 event/ton-year, however, is an ambitious improvement over the currently best achieved background level [8, 9]. With the efforts to reduce the natural decay chain isotopes improving, previously unimportant or unknown background contributions must be understood and eliminated. To that end, the contribution from long-lived isotopes produced by cosmic-ray neutrons in detector and shielding materials must be considered. For example, the work of Ref. [10] measured cosmic activation of ^{76}Ge . Several $0\nu\beta\beta$ experiments utilize large amounts of low background Pb as a means to passively shield external radiation. If the Pb shielding resides on the Earth's surface prior to deployment at an underground laboratory, cosmic activation occurs. No measurements of cosmic activation in Pb exists or lines identified as such by low background experiments. It is not clear if the lack of prior observations is due to short surface exposures of low background Pb or due to a small activation rate. Therefore, it is important to understand the production of long-lived activated products in Pb for future $0\nu\beta\beta$ decay experiments

We exposed a Pb foil sample to a broad-energy neutron beam with an energy spectrum similar to the cosmic-ray neutron flux on the Earth's surface. After neutron exposure, the sample is counted on a low-background γ -ray spectrometer to detect the decays of produced isotopes. From these data we measure the production rate of activated products due to fast neutrons in the Pb sample. Combining with the neutron-beam and cosmic-neutron energy spectra, we make an estimate of the cosmic ray production of long-lived isotopes in Pb. The measured production rate is compared to a production rate from a cross-section calculation over the appropriate energy range and cosmic-ray neutron flux measurements. This article describes our determination of the production rate of these isotopes.

2. Experiment

The natural Pb sample was placed in the Weapons Neutron Research (WNR) facility Target 4 Flight Path 60 Right (4FP60R) [11] neutron beam at the Los Alamos Neutron Science Center (LANSCE). The broad-spectrum, pulsed neutron beam at this flight path strikes the Pb target produce outgoing γ rays to interact with the GERmanium Array for Neutron Induced Excitations (GEANIE) spectrometer [12]. The GEANIE spectrometer data and resulting $(n,n'\gamma)$ analysis are presented in a separate publication [13]. The GEANIE sample is located a distance of 20.34 m from the natural tungsten spallation target.

The neutron target at the center of GEANIE was five stacked foils of natural Pb ($^{\text{nat}}\text{Pb}$) angled 20° from the normal to the beam direction. The neutron beam had a diameter of 1.905 cm. Each foil measured nominally $5\text{ cm} \times 5\text{ cm}$ in area and 0.475 mm in thickness. The Pb sample obtained from GoodFellow has a purity of 99.95%. Typical impurities for this sample include 20 ppm Ag, 100 ppm Bi, 2 ppm Cd, 20 ppm Cu, 2 ppm Fe, < 1 ppm Mg, 70 ppm Sn, and 5 ppm Tl.

The neutron exposure occurred over two periods with 2 foils exposed in the year 2003 and all 5 foils later in year 2006. The first 2-foil exposure was performed between Aug. 18 and Aug. 22, 2003 (3.72 days). The full 5-foil exposure occurred between Oct. 13 and Oct. 30, 2006 (16.70 d), Nov. 9 and Nov. 13, 2006 (3.66 d), and on Nov. 22 (0.33 d). The neutron beam is pulsed allowing a measurement of the neutron energy and flux by time of flight with an in-beam fission chamber containing ^{238}U foils [14]. As seen in Fig. 1, the neutron energy spectrum at 4FP60R in 2006 shows good spectral agreement over an energy range between 20-300 MeV with the Gordon [15] parameterization of the sea level cosmic-neutron energy spectra.

For convenience, we provide a parametrization of the neutron spectrum impinging our Pb target should an alternate cross section or cosmic neutron flux be investigated. The spectrum can be described as:

$$\Phi(E) = (1.832 \times 10^{10}) \times e^{(5.166 \ln E - 3.815 \ln^2 E + 0.895 \ln^3 E - 0.071 \ln^4 E)} \quad (1)$$

where Φ is in units of neutrons/MeV and the energy (E) is in MeV.

The TALYS [16] nuclear reaction code calculates excitation functions for producing nuclides in Pb. Of the nuclides predicted by TALYS, only ^{194}Hg

Figure 1: The energy spectrum of the neutron beam at 4FP60R for the 2006 exposure periods. The solid curve represents the sea level cosmic-neutron flux from Ref. [15].

Figure 2: The production cross sections for ^{202}Pb (top curve) and ^{194}Hg (lower curve) calculated by TALYS for neutrons impinging on natural Pb. The neutron energy range of TALYS is limited to 250 MeV.

and ^{202}Pb are long-lived ($t_{1/2} > 1$ yr) and support radioisotopes with a decay mode to an excited state so that γ -ray emission is possible. ^{194}Hg supports the shorter lived ^{194}Au ($t_{1/2} = 38.0$ h) and ^{202}Pb supports the shorter lived ^{202}Tl ($t_{1/2} = 12.3$ d) [17]. The production cross sections for these two nuclides are shown in Fig. 2. The production cross section of ^{194}Hg includes feeding from the short-lived nuclides ^{194}Tl and ^{194}Pb . Note that the cross sections are peaked within the energy range of 20-300 MeV, which is the same energy region where there is agreement in the shape of the neutron flux between sea level cosmic neutrons and the 4FP60R neutrons (Fig. 1).

The experimental setup at GEANIE is optimized for measuring prompt γ rays due to neutron reactions in a target sample. During periods of time while the data acquisition system is offline, neutron data is not collected by the in-beam fission chamber. Therefore, a direct measurement of the total

neutron yield on target is not possible. Instead, we rely on an upstream proton current monitor that continuously logs the beam current directed at WNR spallation target. This monitor provides a better handle on the total neutron current delivered to the flight path if the fission chamber neutron monitor experiences periods of downtime (e.g. in between data runs, during DAQ malfunctions and debugging, etc.). A subset of the irradiation period while the neutron fission chamber is online can be used to normalize the proton current to delivered neutron current.

After irradiation, the Pb foils were stored for an extended period and therefore any radioactivity had decayed to an extremely low level (<10 Bq) before counting began. Therefore, the activity of the sample was negligible to sample handling and transportation requirements. The sample was taken to our low background High Purity Germanium (HPGe) underground counting facility at the Waste Isolation Pilot Plant (WIPP) near Carlsbad, NM. The HPGe detector and shielding have been underground at WIPP since 1998. This n-type semi-coax detector has a height of 41 mm and a diameter of 51 mm. The cryostat is made from ≈ 1 -mm thick Cu cryostat. The shield comprises 5 cm of oxygen-free, high-conductivity Cu and 10 cm of Pb.

The sample was counted over a period of 126.6 days between Aug. 4 and Dec. 8, 2010 with a break in data collection resulting in a total live time of 109.07 days. The γ -ray energy spectrum is shown in Fig. 3. Three lines from ^{194}Au , a line from ^{202}Tl , and two lines from ^{207}Bi are observed in the activated spectra in addition to background lines. The γ -ray detection efficiency is determined by Monte Carlo simulations using the GEANT4-based MaGe framework [18]. The simulated geometry includes five sheets of Pb placed on the HPGe detector. The simulated γ rays originate uniformly throughout a cylindrical section of the Pb sheet consistent with the beam spot diameter. Therefore, the γ -ray detection efficiency also takes into account self shielding of the Pb foils in addition to the shielding of the HPGe cryostat and the detector response.

3. Measured Production Rates

The observed peaks in the γ -ray spectrum (Fig. 3) are fit to determine the measured counts (C) that are tabulated in Table 1. The measured energy spectrum shows evidence of the expected long-lived activated isotopes. In some of the peaks, the same lines are observed in the background spectra. In the case of ^{207}Bi , the 570-keV and 1064-keV γ ray lines are also

Figure 3: The energy spectrum of γ rays from the Pb sample (red) as measured by a HPGe detector and a background spectrum (blue) taken with no sample present.

found in the background spectra at a lower rate. The 294-keV γ -ray line attributed to ^{194}Au is also found in the background spectra due to naturally occurring ^{214}Pb . The measured counts in Table 1 represent the net counts after background subtraction for these three lines. Note that the 329-keV line could be due to decay of naturally occurring ^{228}Ac . However, the background spectrum shows no evidence of this ^{228}Ac line and no other ^{228}Ac lines are observed in the sample spectrum. Therefore the entire 328-keV line is attributed to the decay of ^{194}Au .

The γ -ray detection efficiency (ϵ_γ) was determined by Monte Carlo simulations benchmarked against measurements of calibrated radioactive standards ($^{57,60}\text{Co}$, ^{54}Mn , ^{22}Na and ^{137}Cs). We use a random pulser to verify the event-rate dependence of the data-acquisition system dead time.

In addition to ϵ_γ , an additional correction factor accounts for the decay of the isotope since the end of exposure and during the counting period. This latter correction factor (ϵ_c) listed in Table 1 depends on the half-life of the isotope of interest.

It is straight-forward to calculate the number of atoms of each isotope that were present at the end of neutron exposure (November 22, 2006) and the production rate. The measured number of counts (C) is related to the number of atoms (N_i) on the reference date at the end of exposure by:

$$C = \epsilon_c \sum_i N_i \epsilon_{\gamma_i}, \quad (2)$$

where the sum is over the four irradiation periods. Since the first irradiation period only included two of the Pb foils, atoms produced from that run have a different γ -ray detection efficiency than those produced in the latter three runs with all five Pb foils. The γ -ray counting occurred over two periods of time with a short break in between. The decay of the isotope before and during the two counting periods is corrected with a sum over the two periods by

$$\epsilon_c = \sum_j \left(e^{-\lambda T_j^{start}} - e^{-\lambda T_j^{end}} \right), \quad (3)$$

where T_j^{end} (T_j^{start}) is the number of days since the end of exposure that the counting stopped (started) for each of the j counting periods, and λ is the decay constant of the isotope in question.

Table 1: A summary of the measured cosmic production rates (K^C) and the predicted cosmic production rates (K_{TALYS}^C) for the long-lived isotopes in the Pb sample. Due to multiple γ lines, we report the emphasized rate values given in column 8. If the observed nuclide is short lived, the half-life of the supporting parent (listed in parentheses) is provided. The stated uncertainty on the production rate includes all statistical and systematic uncertainties combined in quadrature.

Isotope	$\tau_{1/2}$ [days]	γ ray [keV]	C	ϵ_γ 2006	ϵ_γ 2003	ϵ_c	K^C [/kg/day]	K_{TALYS}^C [/kg/day]
^{194}Au (^{194}Hg)	1.90×10^5	293.5	105 ± 114	0.0041 ± 0.0002	0.0042 ± 0.0015	3.95×10^{-4}	7.0 ± 7.7	
^{194}Au (^{194}Hg)	1.90×10^5	328.5	842 ± 52	0.0287 ± 0.0014	0.0294 ± 0.0105	3.95×10^{-4}	8.0 ± 1.3	16
^{194}Au (^{194}Hg)	1.90×10^5	622.0	101 ± 18	0.0009 ± 0.00005	0.0010 ± 0.0003	3.95×10^{-4}	29 ± 7^a	
^{202}Tl (^{202}Pb)	1.93×10^7	439.5	188 ± 28	0.0444 ± 0.0022	0.0453 ± 0.0108	3.90×10^{-6}	120 ± 25	77
^{207}Bi	1.17×10^4	569.7	316 ± 37	0.0341 ± 0.0017	0.0346 ± 0.0055	5.94×10^{-3}	$< 0.17 \pm 0.03$	
^{207}Bi	1.17×10^4	1063.7	146 ± 26	0.0153 ± 0.0008	0.0153 ± 0.0011	5.94×10^{-3}	$< 0.17 \pm 0.04$	

^asee explanation of this outlier in text

The number of atoms can also be expressed in terms of the reaction rates during an exposure period:

$$N_i = \frac{K_i^L M_i}{\lambda} \left(1 - e^{-\lambda T_i^{irrad}}\right) e^{-\lambda T_i^{decay}} \quad (4)$$

where we have corrected for the decay during the exposure and the decay after the exposure until the reference date. In Eqn. 4, T_i^{irrad} is the duration of irradiation of the i^{th} exposure, and T_i^{decay} is the time between the end of exposure i and the reference date. K_i^L is the production rate [atoms/kg/day] during the LANSCE exposure i , and M_i is the mass of the sample within the beam spot.

The production rate of the identified long-lived nuclides in Pb occurs in an energy range where the shapes of the LANSCE and cosmic neutron fluxes are similar. Hence, a simple scaling of the LANSCE production rate is sufficient to estimate the cosmic production rate, given by

$$K^C = K_i^L \frac{f^C}{f_i^L} \quad (5)$$

$$= K_i^L \frac{f^C}{S_i I_i / A_i / T_i^{irrad}} \quad (6)$$

where K^C is the cosmic production rate [atoms/kg/day], f^C (f^L) is the energy-integrated neutron flux [n/cm²/day] for cosmic (LANSCE) neutrons over the mutual energy range 20-300 MeV, S_i is the normalization between proton current and neutron current, and A_i is the cross-sectional area of the Pb target in the neutron beam. The sea-level cosmic neutron flux of Ref. [15] is assumed where $f^C = 228$ n/cm²/day.

Eqn. 4 can be written as

$$C = \frac{K^C \epsilon_c}{f^C \lambda} \sum_i \frac{n_i S_i I_i}{T_i^{irrad}} \epsilon_{\gamma_i} \left(1 - e^{-\lambda T_i^{irrad}}\right) e^{-\lambda T_i^{decay}} \quad (7)$$

where the M_i/A_i term is replaced by the areal density (n_i) of the Pb target parallel to the beam direction. The beam spot covers most, but not all, of the sample geometry.

Table 1 lists the isotopes found and the number of measured counts after irradiations. Eqn. 7 allows a direct determination of the cosmic production rate K^C , also listed in Table 1. Also listed is the cosmic activation rate

calculated with the TALYS excitation functions (Fig. 2 and the sea level cosmic neutrons [15]).

The production of long-lived ^{194}Hg is found through the presence of characteristic γ rays from supported progeny ^{194}Au . The production rates found by the three ^{194}Au do not all agree: The 294-keV and 329-keV lines agree while that found by the 622-keV line is nearly a factor of four higher. Of the three lines, two have low branching ratios and so we place the most confidence on the stronger 329-keV line. Further, the 622-keV line can occur as a lone γ ray or as a sum of the 293.5-keV and 328.5-keV γ rays. The Monte Carlo of the γ -ray detection efficiency is based on the decay of ^{194}Au and hence the cascade is modeled in the efficiency calculation and likely not the sole cause of the observed excess. There are at least four other γ -ray lines expected from ^{194}Au in the observed energy range with non-negligible intensities: 364.8 keV, 482.8 keV, 528.8 keV, 645.2 keV. No peaks are observed at these energies and the background level is used to set production rate limits (90% confidence level) of 17(3), 11(2), 9(1), and 7(1) atoms/kg/day. These limits rule out the larger production rate found by the 622-keV line. Its excess may be due to a blend from another source at the same energy. One likely candidate is the 622-keV γ ray from ^{106}Rh , which is progeny of fission product ^{106}Ru predicted by TALYS to be activated at a rate of 0.05 atoms/kg/day. If we assume the excess of the 622-keV line is due to ^{106}Rh , we find a cosmic activation rate of 0.18 ± 0.06 atoms/kg/day for that isotope.

The production of ^{202}Pb is observed through the lone γ ray from ^{202}Tl . We mentioned above that the irradiated Pb sample may have impurities, which could also be targets for neutron activation. Of the listed impurities, only Bi provides an additional means to activate ^{202}Pb and ^{194}Au . A TALYS calculation of the production in Bi at the stated impurity levels is found to be negligible compared the the rate found in natural Pb.

While the production of ^{207}Bi in natural Pb is observed, some comments on its measured production are necessary. First, the Bi impurity in the natural Pb sample is large enough to have a significant effect on the production of ^{207}Bi . A TALYS calculation finds the production of ^{207}Bi in 100 ppm Bi to be 10% of that measured in natural Pb. Therefore, the measured production rate may be partly due to the Bi impurity of the sample and therefore we report our result as an upper limit only. Second, Bi has a higher atomic number than Pb and hence the reaction requires an incoming proton. It is likely that Bi is produced from a secondary reaction from residual protons left from incident neutrons. This secondary production is not directly pre-

dicted by TALYS. To quantify the likelihood of secondary reactions involving proton capture, a two-step TALYS calculation is made. The first predicts the proton flux from residual protons emitted from incident cosmic neutrons on Pb. Next, the proton flux generates secondary reactions on Pb that produce ^{207}Bi at a rate of 0.04 atoms/kg/day, which is a factor of 5 less than measured here. Therefore, it is plausible that the suspected secondary proton capture reactions cause the ^{207}Bi production found up to the rate reported.

The aerial densities and total neutron yield for the two irradiation periods are listed in Table 2. The low neutron yield and lower Pb thickness in the 2003 run make it a small contribution to the total production of nuclides.

Table 2: A summary of the total neutron yield for the two irradiation years.

Year	n [g/cm ²]	Neutrons on target ($S \times I$) between 20-300 MeV
2003	1.215	$5.73 \pm 5.46 \times 10^{10}$
2006	3.038	$6.77 \pm 0.28 \times 10^{11}$

The uncertainty in the production rate is dominated by the counting statistics of weak peaks (Table 1), the counting efficiency, the exposed neutron yield, and the cosmic ray neutron flux. The γ -ray efficiency for the full five Pb foils exposed in 2006 is known to 5%, which is based on source data and Monte Carlo agreement. The placement of the two Pb foils exposed in 2003 within the five foils during γ counting is unknown, though they were adjacent. Therefore, the 2003 γ -ray detection efficiency has a larger systematic uncertainty (Table 1). The uncertainty on neutron yield is due to the statistical uncertainty on the proton beam current and the uncertainty on the proton to neutron normalization. The 2006 total neutron yield is known to 4.2%. For the 2003 neutron yield calculation, the limited fission chamber data available suggests a lower proton to neutron normalization, though no physical explanation can explain the discrepancy with the 2006 normalization. We assume the nominal 2006 normalization for the 2003 neutron yield, but conservatively include a larger uncertainty of 95% required to account for the difference found when using the limited fission chamber data. The 2003 exposure represents 3.3% of the total neutron exposure so the large uncertainty on the 2003 neutron yield has a small effect on the net production rate.

The precision to which the cosmic ray neutron flux is measured is claimed to be within 10-15% [15] and so we assume an uncertainty of 12.5%. The total cosmic flux also includes contributions from subdominant proton and pion interactions. This neglected contribution is approximately 10% [19] of the total production rate. These charged particles are much less penetrating than neutrons and therefore their impact on any given sample is very geometry dependent. Hence we assume a 50% uncertainty on this correction for an effective uncertainty of 5% on the neglected contribution. Since the two contributes to the cosmic uncertainty are uncorrelated, we use a net estimated systematic uncertainty on the cosmic neutron flux of 13.5%. The counting durations live times are known to a small percentage and are negligible contributions to the uncertainty. This certainty is also true for the irradiation times. The sample had been stored on the Earth's surface for many years prior to exposure to the neutron beam. Any isotopes produced by cosmic ray neutrons would be below saturation after this extended period is ignored given a cosmic contribution $\ll 3\%$ than that produced by the intense neutron beam. The half-lives and branching ratios of the produced isotopes are known to high precision and are a negligible contribution to the total uncertainty.

4. Discussion

Previous low background experiments using Pb shielding have not found lines in their detector's energy spectrum attributed to long-lived activated products in Pb. For example, the HEILDEBERG-MOSCOW (H-M) [20] neutrinoless double-beta decay experiment does not present evidence of ^{194}Au or ^{202}Tl . The H-M spectra does show the presence of ^{207}Bi , though its attributed to anthropogenic rather than cosmic production in Pb shielding. The ^{207}Bi may be partially cosmogenic in origin in the Pb shielding. If cosmogenic, the decay rate of ^{207}Bi observed here suggests that ^{194}Au and ^{202}Tl would also be observed. However, the activation of ^{207}Bi here is due to secondary reactions involving protons and the net cosmic production would be greater due to the flux of cosmic protons.

The work of Mei *et al.* [21] using a Pb shield surrounding a HPGe detector did observe an unidentified line at 438.9 keV, which could be attributed to the ^{202}Tl line at 439.5 keV. However, a long term spectra obtained by Ref. [22] also observed a line at 438.8 keV but attributed it to a ^{40}K double escape peak. The relative likelihood of this line being attributed to either

cosmogenic ^{202}Pb or ^{40}K should be further investigated. If cosmogenics in Pb are present, its more likely that the more abundant ^{194}Au would be present. The two experiments see the 329-keV line attributed to ^{228}Ac , which could be blending with cosmogenic ^{194}Au . Instead, the rate of the 329-keV line is consistent with the nearby 338-keV line from ^{228}Ac when scaled by their branching ratios suggesting little evidence for the cosmogenic ^{194}Au .

Neutrinoless double-beta decay experiments typically choose target nuclei with a high (≥ 2 MeV) double-beta Q value in order to stay above most γ rays from natural sources. The activated products discovered here do pose a potential background contribution due to their high decay Q values and high energy γ rays. While ^{194}Au has a decay Q-value of 2501 keV with γ rays up to 2413 keV and ^{207}Bi has a decay Q-value of 2398 keV with γ rays up to 1770 keV, ^{202}Tl is less of a concern with a lower Q value of 1363 keV [17]. The production rates of these cosmically activated radioisotopes in Pb can be used to estimate background contributions and surface exposure limitations for the next generation neutrinoless double-beta decay experiments.

With its high energy γ rays, ^{194}Au is likely the most problematic cosmogenic in Pb shielding. The highest intensity γ -ray line above 2 MeV occurs at 2043.7 keV. This transition is close to the 2039-keV Q value of double-beta decay target ^{76}Ge . Using the production rate measured here, cosmogenic ^{194}Au will saturate at a concentration of 93 $\mu\text{Bq/kg}$. With a decay intensity of 3.81%, the 2044-keV γ ray would be produced at a rate of 0.0035 mHz/kg at saturation. As stated earlier, the H-M experiment did not see evidence of ^{194}Au . To estimate their sensitivity to a 2 MeV γ ray, they did observe the 2.6-MeV γ ray from ^{208}Tl in the ^{232}Th decay chain and determined the concentration to be 12.3 $\mu\text{Bq/kg}$ of ^{232}Th (4 $\mu\text{Bq/kg}$ of ^{208}Tl) in the Pb shielding [23]. Therefore, they would have sensitivity to ^{194}Au at saturation indicating the Pb shielding had little exposure to cosmic neutrons at the surface of the Earth prior to use underground. Current and future experiments are finding shielding materials with improved contamination levels. Pb suitable for shielding has been obtained at <4 $\mu\text{Bq/kg}$ of ^{232}Th (<1.3 $\mu\text{Bq/kg}$ of ^{208}Tl) [24]. Since ^{232}Th contamination in Pb is currently unavoidable, it can be used to set the scale of other contaminate goals. In order to keep the decay rate of the 2044-keV γ ray at or below the decay rate of the ^{208}Tl 2614-keV γ ray, Pb surface exposure should be limited to 350 yr. A limitation of 30 yr of surface exposure is required to stay a factor of 10 less than the ^{208}Tl decay rate.

5. Conclusion

We measure the production of ^{194}Hg , ^{202}Pb , and ^{207}Bi in a sample of Pb due to high-energy neutron interactions within a neutron beam with a spectrum similar to that of the cosmic-ray neutron flux at the Earth's surface. These results are used to predict sea level neutron activation of long-lived nuclides in Pb shielding. Measurements of γ -ray emissions reveal the presence of long-lived isotopes and we set production rates of 8.0 ± 1.3 atoms/kg/day of ^{194}Hg , 120 ± 25 atoms/kg/day ^{202}Pb , and $< 0.17 \pm 0.04$ atoms/kg/day ^{207}Bi . Neutron activation of natural Pb can be a source of background for rare event searches such as neutrinoless double-beta decay experiments. Based on previous low background γ ray spectrometers using Pb shielding and given the production rates measured here, we can estimate the surface exposure necessary for neutron-activated products to be a significant background source. The cosmogenic background would require a sea level exposure of 350 yr to be commensurate with the background from ^{208}Tl in ultra-pure Pb shielding. A shorter exposer would guarantee the cosmogenic background is less than the naturally occurring radioisotopes in Pb. Therefore, it is not strictly necessary to provide ultra-pure Pb shielding short-term protection from cosmic rays in the execution of low background, rare event searches using Pb.

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