

Cosmogenic and primordial radioisotopes in copper bricks shortly exposed to cosmic rays

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Abstract. Cosmogenic activation is the most common source of radioactivity in copper, being ⁶⁰Co the most significant because of its long half-life (5.27 y) and saturation activity at sea level of 1 mBq/kg. Copper bricks, which had been exposed to cosmic rays for 41 days after their casting, were used to replace the internal 10 cm of the lead shielding of a HPGe detector placed at the Canfranc Underground Laboratory. We describe the outcome of the new shielding and the cosmogenic and primordial radioisotopes observed.

1. Introduction

Shielding has an important impact in the results of any ultra-low background experiment. Usually, inner layer is made of copper or lead, provided that the latter is old enough to contain a low specific activity of ²¹⁰Pb ($T_{1/2} = 22.3$ y [1]). This isotope, which is a daughter of the ²³⁸U in the lead ore, can reach more than 1 kBq/kg in processed lead [2]. In copper, ⁶³Cu(n, α) captures produce ⁶⁰Co ($T_{1/2} = 5.27$ y [1]), that can reach specific activities of about 1 mBq/kg at sea level and 2 mBq/kg at 1 km altitude [3]. Metallurgic processes reduce ⁶⁰Co in Cu, but cannot reduce ²¹⁰Pb in Pb; therefore, the concentration of ⁶⁰Co depends on exposure to cosmic rays after casting date, whereas concentration of ²¹⁰Pb depends on quantity of uranium in ore and on elapsed time after extraction of lead from ore.

Choice of copper or lead depends on radio-purity, availability, price and application. For instance, the main gamma radiation from ²¹⁰Pb contained in lead is due to the bremsstrahlung of the beta rays emitted by her daughter ²¹⁰Bi. This background is very important around 200 keV, but very small above 600 keV and zero above 1.16 MeV, the end-point of beta rays [1]. In the following sections we describe the observed changes in the measured spectra of a hyperpure germanium (HPGe) detector placed at 2450 meter water equivalent underground when the innermost Pb bricks of its shielding were replaced by Cu bricks.

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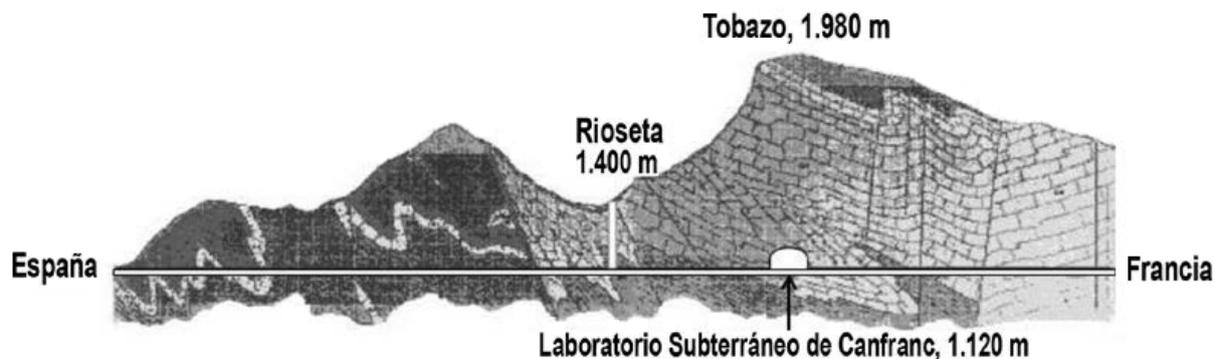


Figure 1. Profile of the Canfranc Underground Laboratory (LSC)

2. Quality control of copper

Primordial and cosmogenic isotopes were searched for in a sample of 18 kg of copper before purchasing the total amount of Cu bricks. Cosmogenic isotopes were expected because, according to company records, the copper had been exposed at 250 m above sea level for 1 year after casting. This elapsed time was useful to compare our data with other measurements [3], to check the storing conditions of copper (altitude, possible cosmic ray attenuation by basement storage, etc.) as well as to estimate the allowed copper exposure to maintain a negligible (less than $\sim 10 \mu\text{Bq/kg}$) ^{60}Co content.

2.1. Experimental set-up

Measurements were done in the Canfranc Underground Laboratory (Laboratorio Subterráneo de Canfranc, figure 1) with a HPGe detector of ~ 1 kg and 41% relative efficiency, inside a shielding of 25 cm lead thickness. The radon was purged with the nitrogen gas evaporated from the HPGe dewar (~ 10 l/h) and the stability of the acquisition system was weekly checked by calibration with a weak ^{22}Na source.

Table 1. Results of copper sample measurement.

Cosmogenic radionuclide	$T_{1/2}$ [1]	Measured specific activity (mBq/kg)	Specific saturation activity at sea level (mBq/kg)[3]
^{56}Co	77 d	0.080 ± 0.018	0.110 ± 0.014
^{57}Co	272 d	0.52 ± 0.10	0.86 ± 0.19
^{58}Co	71 d	0.72 ± 0.06	0.790 ± 0.043
^{60}Co	5.27 y	0.082 ± 0.016	1.00 ± 0.09
^{54}Mn	312 d	0.14 ± 0.03	0.10 ± 0.01
Primordial radionuclide			
^{238}U		< 3.9	
^{226}Ra		< 0.39	
^{232}Th (equilibrium)		< 0.12	
^{40}K		< 0.40	

2.2. Results of quality control of the copper sample

Table 1 shows the results of the measurement. Cosmogenic radionuclides measured activities are consistent with exposure time communicated by company, see specific saturation activities in the last column of table 1. In particular, the ^{60}Co specific activity implies an exposure (at sea level) of 8 ± 2 months after casting. Consequently, the maximum time for copper storage above ground is about 6 weeks. Primordial isotopes were not observed in the sample, their upper limits at 95% C.L. are listed in table 1.

3. Shielding upgrade

Once we assessed the quality of copper, we purchased a batch of bricks that, according to company records, had been exposed to cosmic rays for 41 days after casting. The internal 10 cm of lead, containing ~ 1 Bq/kg of ^{210}Pb , were replaced (figures 2 and 3) by copper.



Figure 2. Initial shielding (25 cm lead).



Figure 3. Final shielding (15 cm lead and 10 cm copper).

Figure 4 shows the total rate between 20 keV and 5.4 MeV for 1 year data. The rate decreased clearly for 6 months because of the decay of shorter lived cosmogenic radionuclides (^{56}Co and ^{58}Co). The measured rates of the peaks from primordial radionuclides remained constant over time.

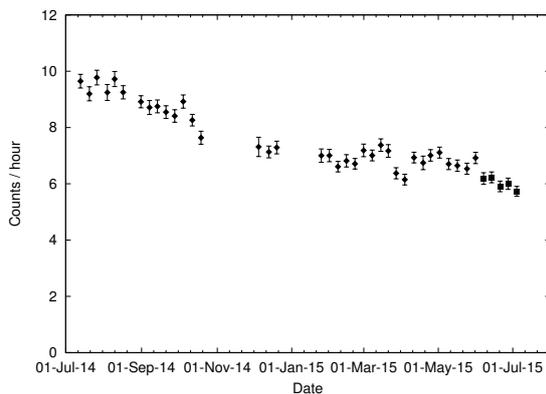


Figure 4. Total rate between 20 keV and 5.4 MeV for the Cu+Pb shielding.

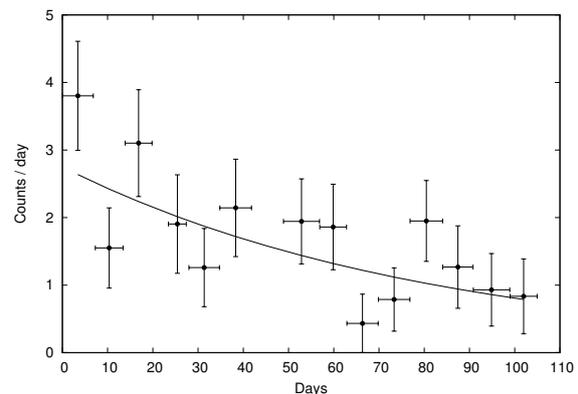


Figure 5. Rate of 811 keV line from the beginning of data acquisition.

After 6 months the rate is compatible with a constant value except for the observed decrease around the beginning of April, which coincided with a ^{222}Rn decrease in the laboratory. The

rate decreased also the last month (full square points of figure 4) because we obtained a further reduction of ^{222}Rn inside the shielding by increasing, with gas provided by LSC, the nitrogen purge up to ~ 30 l/h.

The only visible line from a cosmogenic radionuclide was 811 keV (^{58}Co). It decreased (figure 5) with a half-life of 57_{-15}^{+24} days, compatible with her 71 days half-life [1]. The specific activity of ^{58}Co indicated that the exposure had been 66 ± 10 days (at sea level), roughly similar to the time communicated by the company. After 3 months, the 811 keV line was not longer visible. The data of last month were used to give upper limits to cosmogenic and primordial radionuclides (table 2).

Table 2. Upper limits (95% C.L.) to radionuclides in copper shielding (last month data).

Cosmogenic radionuclide	mBq/kg	Primordial radionuclide	mBq/kg
^{56}Co	< 0.050	^{238}U	< 2.9
^{57}Co	< 0.26	^{226}Ra	< 0.080
^{58}Co	< 0.053	^{232}Th (equilibrium)	< 0.083
^{60}Co	< 0.028	^{40}K	< 0.26
^{54}Mn	< 0.047		

4. Shielding comparison

The spectrum of the last month, when cosmogenic radionuclides were not observed, is plotted in figure 6. Figure 7 shows the spectrum with the shielding of lead, taken before the change of the inner Pb bricks by Cu bricks.

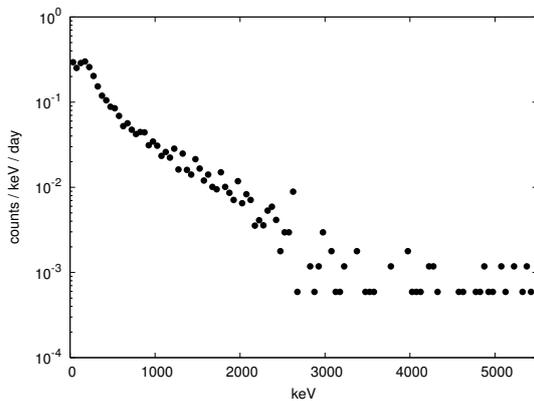


Figure 6. HPGe spectrum with the Cu+Pb shielding. Last month of data.

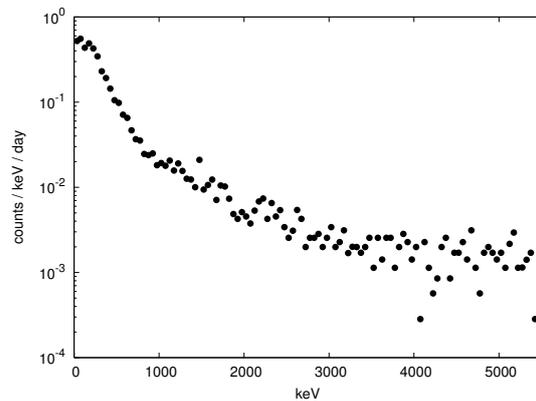


Figure 7. HPGe spectrum with the Pb shielding before the change.

Their differences can be observed in three regions (figures 8 and 9):

- Below 0.6 MeV the spectrum of the Pb shielding is higher than the Cu+Pb one, because of the bremsstrahlung from beta rays of ^{210}Bi and Pb X-rays from the inner lead [4].
- From 0.6 MeV to 2 MeV the spectrum of the Cu+Pb shielding is higher because of the lower attenuation of copper to the gamma radiation coming from the external lead and the outside. This is observed also in the 2615 keV line.

- Above 2.6 MeV the rate in the Cu+Pb shielding is 0.38 ± 0.04 times the rate in the Pb one. This factor matches the ratio of the atomic numbers of Cu and Pb ($29/82=0.35$). It means that the bremsstrahlung [5] of muons in the inner shielding is, likely, the dominant interaction.

The rate is roughly the same from 2 to 2.6 MeV, more probably due to the slow transition between two adjacent regions of opposite behavior.

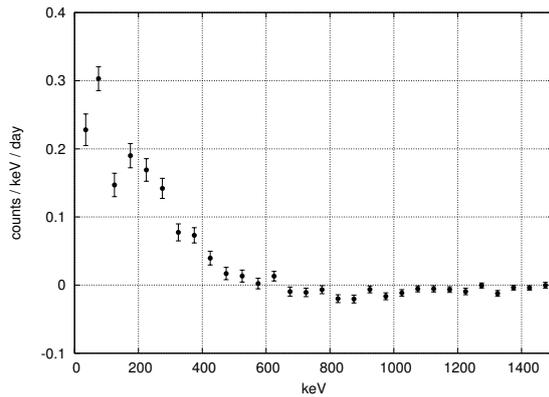


Figure 8. (Pb shielding)-(Cu+Pb shielding) difference spectrum up to 1.5 MeV.

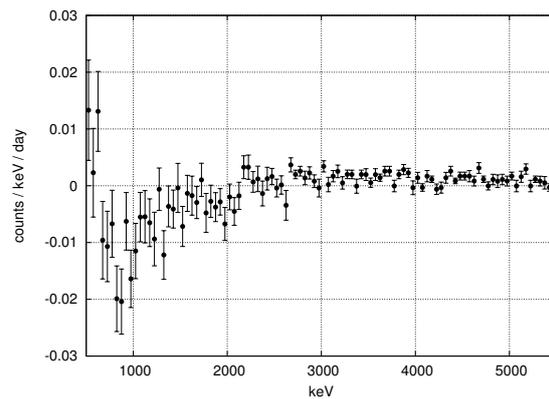


Figure 9. Same difference as figure 8 from 0.5 MeV to 5.4 MeV.

5. Conclusions

The background of our HPGe detector has decreased after the replacement of the inner lead bricks (containing ~ 1 Bq/kg of ^{210}Pb) by those made of copper shortly exposed to cosmic rays. According to the mean free paths of 2615 keV photons (3.0 cm in Cu and 2.1 cm in Pb [6]), the Cu+Pb shielding would be advantageous also in the region from 0.6 to 2 MeV by increasing the inner copper thickness to 15 cm.

Acknowledgements

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