



Ultrasensitive detection method for primordial nuclides in copper with Accelerator Mass Spectrometry



N. Famulok*, T. Faestermann, L. Fimiani, J.M. Gómez-Guzmán, K. Hain, G. Korschinek, P. Ludwig, S. Schönert

Technische Universität München, James-Frank-Strasse 1, 85747 Garching, Germany

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ABSTRACT

The sensitivity of rare event physics experiments like neutrino or direct dark matter detection crucially depends on the background level. A significant background contribution originates from the primordial actinides thorium (Th) and uranium (U) and the progenies of their decay chains. The applicability of ultra-sensitive Accelerator Mass Spectrometry (AMS) for the direct detection of Th and U impurities in three copper samples is evaluated. Although AMS has been proven to reach outstanding sensitivities for long-lived isotopes, this technique has only very rarely been used to detect ultra low concentrations of primordial actinides. Here it is utilized for the first time to detect primordial Th and U in ultra pure copper serving as shielding material in low level detectors. The lowest concentrations achieved were $(1.5 \pm 0.6) \cdot 10^{-11}$ g/g for Th and $(8 \pm 4) \cdot 10^{-14}$ g/g for U which corresponds to (59 ± 24) and (1.0 ± 0.5) $\mu\text{Bq/kg}$, respectively.

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1. Introduction

For the past decades, the need for ultra pure material with a very low contamination of radionuclides has been increasing. Rare event experiments located underground require a low radioactive background of the detector surroundings [1–4]. Different materials like lead, steel or copper are used for passive shielding to suppress external radioactivity. However, these materials can also contain radionuclides themselves, e.g. primordial actinides. The decaying daughter nuclides of their natural decay chains can create interfering signals in the detector. Therefore a careful selection and purification of the shielding material [5] is performed to reduce the amount of primordial radionuclides as much as possible. If low contaminations with radioactive impurities and cosmogenic isotopes are ensured, copper can be one of the cleanest shielding materials available [4]. It is the material of choice for detector support and innermost shielding. For the GERDA-experiment [3], the requirements for copper purity are very stringent, especially for copper parts located very close to the detectors.

The main methods for measuring the amount of radioactive contaminants in shielding material are gamma spectrometry which measures the decay of the daughter nuclides [6] and ICP-MS which detects the nuclides directly [7,8].

We introduce a new method of directly measuring extremely low levels of primordial Th and U in copper samples with Accelerator Mass Spectrometry (AMS). The AMS setup at the Maier-Leibnitz-Laboratory in Garching [9,10] has proven to reach outstanding sensitivities for actinides. Hence, it is well suited for the detection of ^{232}Th and ^{238}U contaminations at trace level.

2. Samples

The material used in the analysis for primordial actinides is copper of different purity. As standard with a known concentration determined by means of gamma spectrometry we use a copper alloy called CU5 or Kuprodur (formerly CuNi60) obtained from the company “Korrodin” [11]. It consists of copper, 1.0%–1.6% nickel and 0.4%–0.7% silicon [12]. For fixation of the top flange of a liquid argon scintillation veto (called “LArGe”) used for GERDA, ~15 cm long bolts of CU5 were used. A sample of 94 of these bolts (~15 kg) was screened in the GeMPI-II gamma spectrometer at Gran Sasso National Laboratory (LNGS) for around 13 days. The results of this screening are shown in Table 1. Significant values for the ^{232}Th -chain were obtained from daughter nuclides at the beginning of the decay chain and an arithmetic average was taken for the concentration of ^{232}Th assuming secular equilibrium. For the ^{238}U chain, gamma lines of only one daughter nuclide lead to a concentration above the expected background. Since no other concentration measurements or information about a possibly

* Corresponding author.

broken secular equilibrium of the CU5 sample were available, we assume this concentration to be correct.

The two samples investigated in this work are firstly conventional copper of industrial standard without any special purity treatment called M-copper. Secondly, a sample called LENS-copper, originally manufactured for the “LENS Low-Level Background Facility” at LNGS [13]. Two sets of this copper (33 mm thick plates, with a total mass of 125 kg) were screened in the GeMPI detector for 100.7 days [14]. The resulting activities were $< 19 \mu\text{Bq/kg}$ for ^{228}Th and $< 16 \mu\text{Bq/kg}$ for ^{226}Ra , which corresponds to concentrations of $< 4.67 \cdot 10^{-12} \text{ g/g}$ for ^{232}Th and $< 1.30 \cdot 10^{-12} \text{ g/g}$ for ^{238}U .

All these samples were manufactured as cylindrical cathodes to be put into the AMS ion source. To exclude any surface contamination, the samples were cleaned with 7 M hydrochloric acid (at 50 °C for about 15 min), rinsed with deionized water followed by ultrasonic cleaning in isopropanol. Because the surface of the sample is sputtered during the sputter process in the ion source, no further sophisticated cleaning and special treatment of the equipment [15] is needed.

3. Setup

The copper sample of interest is put into a Middleton type ion source [16,17] where ionized caesium is used to sputter ions off the sample. For the ion source used in this experiment we paid special attention that no macroscopic amounts of actinides were introduced in the source before or during this experiment. The negative ions are extracted from the source and enter a 90° injector magnet for a first mass separation. An adjacent 18° electrostatic deflector suppresses scattered particles. Afterwards, the ions are injected into the tandem accelerator and reach the terminal which has a voltage of around 10 MV in this experiment. Several electrons of the ions are removed at the terminal by a $4 \mu\text{g cm}^{-2}$ carbon stripper foil, leading to positively charged ions of different charge states. This process suppresses all molecular background and the ions reach another 90° magnet for mass and charge separation.

To get a low background, the magnetic rigidity of charge state 9⁺ is chosen. After passing additional Wien velocity filters, the particles reach the detection system in a dedicated beamline. The detection consists of a time-of-flight (TOF) path and a silicon surface detector, allowing a discrimination of mass and energy. The ions pass a $7 \mu\text{g cm}^{-2}$ carbon foil where electrons are liberated and electrostatically reflected onto a micro channel plate. Thus the start signal of the TOF is created. The stop signal is triggered with the signal of the Si detector at the end. For details on the setup see also [18].

Table 1

Results from the 13 days screening of the CU5-bolts in the GeMPI-II detector. Uncertainties with ~68%CL. The individual nuclides are measured as follows: ^{228}Ra from decay of ^{228}Ac , ^{228}Th from decay of ^{212}Pb , ^{212}Bi and ^{208}Tl , ^{226}Ra from decay of ^{214}Pb and ^{214}Bi [12].

^{232}Th -chain	Radionuclide	Activity (mBq/kg)	Concentration of ^{232}Th (g/g)
	^{228}Ra	0.39 ± 0.12	$(9.7 \pm 3.0) \cdot 10^{-11}$
	^{228}Th	1.5 ± 0.1	$(3.7 \pm 0.4) \cdot 10^{-10}$
^{238}U -chain	Radionuclide	Activity (mBq/kg)	Concentration of ^{238}U (g/g)
	^{226}Ra	0.030 ± 0.007	$(2.4 \pm 0.6) \cdot 10^{-12}$
	^{234}Th	< 22	$< 1.8 \cdot 10^{-9}$
	$^{234\text{m}}\text{Pa}$	< 7.0	$< 5.7 \cdot 10^{-10}$

4. Method

In AMS, Th and U are often extracted as negatively charged oxides. In this experiment no stable conditions were achieved with oxides. The residual air in the ion source varies over time which affects the yield of the negative oxides. Therefore we chose a compound of the element with the matrix material which is formed in the ion source.

Th or U atoms and copper atoms can form a molecular bond when they are ionized. The resulting negative molecules are ThCu^- and UCu^- which were not used in AMS before. We selected the isotope ^{63}Cu because of its higher abundance.

Before the sample of interest is put into the source, all components of the setup are tuned with a beam of gold ions. For the calibration of the energy/TOF-spectra a linear interpolation with two charge states of gold is performed, using an attenuated beam. By choosing a specific terminal voltage, the charge state of an ion can be selected, while keeping the magnetic rigidity constant.

The superimposed spectra of residual energy vs TOF from ^{197}Au measurements with the charge states 7⁺ and 8⁺ are shown in Fig. 1, while Fig. 2 shows the superimposed spectra from a ^{232}Th and a ^{238}U measurement. Each peak in the spectrum corresponds to an isotope that can be identified by its mass and charge state. It can be seen that the discrimination of each isotope is very good and the measurements show no background in the region of interest. This is due to the fact that there are no other primordial nuclides in the mass range of ^{232}Th , ^{235}U and ^{238}U .

The concentration of actinides in the samples is determined as the number of actinide nuclei per number of copper nuclei. To check the tuning in the beam line at the Faraday cup located in front of the accelerator entrance, we choose a cluster of four copper atoms, namely $^{63}\text{Cu}_3^{65}\text{Cu}^-$ with a mass of $A = 254$, not too different to the mass of the actinide–copper molecules. We took this copper cluster also as a reference for the actinides. It seems to us a reliable assumption, to use molecules for comparison and as reference instead of single copper ions. However, future measurements are needed to confirm this supposition.

Concentration measurements in AMS are usually referred relatively to a standard. Therefore material with a known concentration has to be compared to the measured samples of interest. The copper alloy CU5 with established concentrations obtained by gamma spectrometry served this purpose. The concentrations of Th and U in the copper samples were determined relatively to the concentrations in the standard sample.

The concentration c in the samples was obtained using Eq. (1), in which t is the measuring time, I is the current of the copper

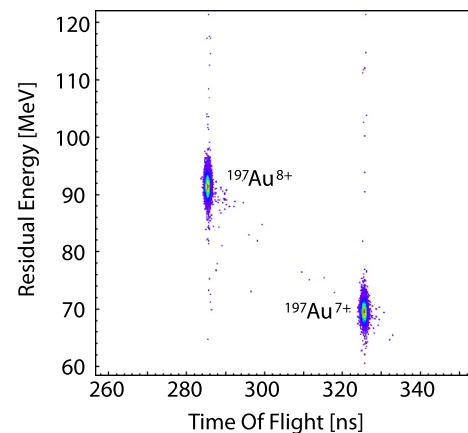


Fig. 1. Residual energy vs. TOF spectra overlaid for a $^{197}\text{Au}^{7+}$ and a $^{197}\text{Au}^{8+}$ run.

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