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# AMS measurement of <sup>10</sup>Be concentrations in marine sediments from Chile Trench at the TANDAR laboratory



BEAM INTERACTIONS WITH MATERIALS

D. Rodrigues <sup>a,b,\*</sup>, A. Arazi <sup>a,b</sup>, J.O. Fernández Niello <sup>a,b,c</sup>, G.V. Martí <sup>a</sup>, A.E. Negri <sup>b,c</sup>, D. Abriola <sup>a</sup>, O.A. Capurro <sup>a</sup>, M.A. Cardona <sup>a,b,d</sup>, E. de Barbará <sup>a</sup>, F. Gollan <sup>a,b</sup>, D. Hojman <sup>a,b</sup>, A.J. Pacheco <sup>a,b</sup>, N. Samsolo <sup>a</sup>, M. Togneri <sup>a</sup>, D. Villanueva <sup>a</sup>

<sup>a</sup> Departamento de Física Experimental, Laboratorio TANDAR, GlyA, Comisión Nacional de Energía Atómica, Av. Gral. Paz 1499, B1650KNA San Martín, Argentina <sup>b</sup> CONICET, Av. Rivadavia 1917, C1033AAJ Buenos Aires, Argentina

<sup>c</sup> Instituto de Investigación e Ingeniería Ambiental, Universidad Nacional de San Martín, 25 de Mayo y Francia, B1650BWA San Martín, Buenos Aires, Argentina <sup>d</sup> Escuela de Ciencia y Tecnología, Universidad Nacional de San Martín, 25 de Mayo y Francia, B1650BWA San Martín, Buenos Aires, Argentina

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#### 1. Introduction

# ABSTRACT

The <sup>10</sup>Be/<sup>9</sup>Be ratios in marine sediments samples from the Southern Chile Trench have been measured using accelerator mass spectrometry (AMS). The samples were measured at the TANDAR accelerator, where the discrimination of the <sup>10</sup>Be radionuclides was achieved by means of a passive absorber in front of an ionization chamber. This setup along with the high voltage available, provided a complete suppression of the <sup>10</sup>B isobar interference. The obtained values for the <sup>10</sup>Be concentrations, of the order of 10<sup>9</sup> - atoms/g, are the first <sup>10</sup>Be measurements from the Southern Chile Trench and offer an excellent tracer to quantitatively study the recycling of sediments in Andean magmas.

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In the last decades, accelerator mass spectrometry (AMS) has proven to be the most sensitive technique for detecting several long-lived radionuclides [1], <sup>10</sup>Be among them. This is the longest-lived beryllium radionuclide with a half-life of  $(1.387 \pm 0.012) \times 10^6$  yr [2], primarily produced in Earth's atmosphere by cosmic-ray induced nuclear reactions on N and O [3]. Detectable amounts of <sup>10</sup>Be are present in rainwater, snow, surface water, and also in marine sediments [4]. It is an ideal radionuclide to be used as tracer in various branches of geosciences [5,6], as well as in astrophysics [7].

Small accelerators exclusively designed for AMS technique offer high transmission and stability. However, the higher beam energies achievable with large tandem accelerators allow the use of a passive absorber to efficiently suppress the interfering isobar (<sup>10</sup>B in this case) and also get a better identification of other interfering ions with a telescope detector.

\* Corresponding author at: Departamento de Física Experimental, Laboratorio TANDAR, GIyA, Comisión Nacional de Energía Atómica, Av. Gral. Paz 1499, B1650KNA San Martín, Argentina.

E-mail address: darodrig@tandar.cnea.gov.ar (D. Rodrigues).

This manuscript reports the measurement at the 20 UD TAN-DAR (TANDem ARgentino) accelerator of the <sup>10</sup>Be concentration in two marine sediments samples from the Ocean Drilling Program (ODP) Site 860 [8]. This site is located at the so-called Chile Triple Junction, where Nazca, Antarctic and South American tectonic plates meet. The bulk of each sample was processed with the aim of quantifying the inventory of total <sup>10</sup>Be in the sediment. This is crucial to estimate the amount of sediment involved in the recycling process produced by the subduction of the oceanic plate underneath the South American Plate [9,10].

In Section 2 we describe the instrumentation used to carry out the AMS measurements. The chemical procedure performed, as well as the AMS measurement procedure are described in Section 3. In Section 4 we present the results and discussion and the conclusions in Section 5.

#### 2. Instrumentation

As part of the AMS program at the TANDAR Laboratory [11,12] several improvements on the machine were carried out during the last years. A new Source of Negative Ions by Cesium Sputtering (SNICS II) was emplaced, the alignment of the experimental lines was carefully checked, the stability of the Generating Volt Meter

(GVM) system was enhanced and the installation of a Wien filter is currently underway.

For AMS measurements, the switching between <sup>10</sup>Be and <sup>9</sup>Be beams was done changing the terminal voltage while keeping the magnetic rigidity at the high energy side fixed. In contrast to most of the facilities used for AMS, the TANDAR accelerator has two electrostatic quadrupoles and one electrostatic steerer along its 33-meters long acceleration column. These optical elements must be also adjusted according to the particle energy during switching between the radionuclide and its stable isotope. In fact, the proper tuning of the electrostatic steerer –placed just after the stripper– was of great importance in order to reduce scattered ions (see Section 3.3).

The main challenges in identifying <sup>10</sup>Be are dealing with molecular (<sup>9</sup>Be<sup>1</sup>H) and atomic (<sup>10</sup>B) interfering isobars. The molecular ions get dissociated at the foil stripper at the terminal of the accelerator. The <sup>10</sup>B ions having the same electric charge state as the one selected for <sup>10</sup>Be after the foil stripper (3<sup>+</sup>) cannot be differentiated by magnetic or electric deflectors. However, the high particle energy available by means of the TANDAR accelerator along with the use of a passive absorber in front of a  $\Delta$ E-E telescope detector, allow discriminating <sup>10</sup>Be radionuclides from its stable isotope (those which manage to pass the analyzing magnet) and from other reaction products (see discussion below) with the complete suppression of <sup>10</sup>B.

Fig. 1 shows a scheme of the detection system built for <sup>10</sup>Be measurements. The passive absorber is a 150 mm long cylinder with a 17 mm diameter, 4  $\mu$ m thick Havar<sup>®</sup> entrance window, based on a similar system developed at the Australian National University (ANU) in Canberra [13,14]. The absorber gas and the one used in the ionization chamber were kept separated by a 2  $\mu$ m thick Havar<sup>®</sup> foil. Thus, choosing the appropriate Ar gas pressure in the passive absorber, the <sup>10</sup>B ions lose all their energy in the absorber, while the more penetrating <sup>10</sup>Be ions enter the  $\Delta$ E-E telescope detector. This detector comprises an ionization chamber 110 mm long filled with 45 Torr P-10 gas followed by a surface barrier detector.

Absorber gas and window material were chosen in order to ensure the minimum content of hydrogen in them [14]. Otherwise, reactions products from the  ${}^{10}\text{B} + {}^{1}\text{H}$  interaction, namely  ${}^{7}\text{Be}$  and  ${}^{4}\text{He}$  ions, could reach the  $\Delta$ E-E telescope detector and contribute to the background [15].

The optimal pressure within the passive absorber is the minimum necessary to completely stop the <sup>10</sup>B ions. This minimizes the energy loss and energy straggling for the <sup>10</sup>Be ions which arrive at the ionization chamber, and thus optimizing their identification. Energy loss calculations using the SRIM code [16] show that in a 150 mm long passive absorber with Ar gas, a pressure of 175 Torr–plus the 4-µm-Havar<sup>®</sup> window– is sufficient to stop <sup>10</sup>B ions having an energy of 27 MeV. Under these conditions the desired <sup>10</sup>Be ions arrive to the ionization chamber with enough energy (about 11 MeV) to pass through the P-10 gas. Finally, these radionuclides reach the surface barrier detector placed behind the ionization chamber with an energy of about 8.5 MeV.

### 3. Experimental procedure

## 3.1. Production and standardization of <sup>10</sup>Be enriched material

The transmission of the system was studied by using commercial BeO powder enriched in <sup>10</sup>Be by neutron activation. This was achieved by irradiating the material with a thermal neutron flux of  $\Phi = (6.1 \pm 0.3) \times 10^{13}$  cm<sup>-2</sup> s<sup>-1</sup> during (60 ± 1) min in the RA-3 reactor at the Ezeiza Atomic Center (Argentina) and producing <sup>10</sup>Be through the neutron capture reaction <sup>9</sup>Be(n,  $\gamma$ )<sup>10</sup>Be.

Both, enriched in <sup>10</sup>Be and non-irradiated powders were dissolved. Beryllium concentrations were calculated from the mass of BeO dissolved in both solutions and confirmed using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) analysis, yielding compatible results within its relative error of 5 %.

Isotopically diluted material (nominal <sup>10</sup>Be/<sup>9</sup>Be =  $(2.2 \pm 0.2) \times 10^{-11}$ ) from the enriched one was measured at ANU in Canberra [14], yielding <sup>10</sup>Be/<sup>9</sup>Be =  $(2.10 \pm 0.06) \times 10^{-11}$ . In this way, we standardize the enriched powder with a ratio of <sup>10</sup>Be/<sup>9</sup>Be =  $(1.87 \pm 0.13) \times 10^{-9}$  traceable to the NIST standard material which was used at ANU.

Mixing in appropriate proportions the solutions of neutronactivated material and the commercial one (our "blank" material henceforth), six different <sup>10</sup>Be solutions with isotope ratios between  $(9.2 \pm 0.8) \times 10^{-15}$  (blank only, see Section 3.2) and  $(1.87 \pm 0.13) \times 10^{-9}$  (pure neutron-activated material) were prepared. Their corresponding isotope ratios were determined with a relative uncertainty of 6 %. The main contributions to the uncertainty come from the measurements of the concentrations and the determination of the volumes used in the mixtures.

#### 3.2. Geological samples

The studied samples were taken from the Core 141-860B, Site 860 (ODP), located at the middle continental slope of the South American plate in the South Pacific Ocean (45°51.972′S, 75°45.101′ W). They correspond to depths of 0.77 mbsf (meter below sea floor, sample 001H) and 2.12 mbsf (sample 002H). The total beryllium content in both sediment samples was also determined by ICP-OES analyses. For chemical processing, 0.67 mg of beryllium carrier material was added to 300 mg of each sample. The isotope ratio of this carrier material had been previously measured at the Vienna Environmental Research Accelerator (VERA) [17], yielding <sup>10</sup>Be/<sup>9</sup>Be = (9.2 ± 0.8) × 10<sup>-15</sup>.

The chemical separation of Be was performed according to the technique reported by Merchel and Herpers [18]. Hydroxides are



Fig. 1. Scheme of the passive absorber and the  $\Delta$ E-E telescope detector used for the identification of <sup>10</sup>Be.

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