

The ^{144}Ce source for SOX

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Abstract. The SOX (Short distance neutrino Oscillations with BoreXino) project aims at testing the light sterile neutrino hypothesis. To do so, two artificial sources of antineutrinos and neutrinos respectively will be consecutively deployed at the Laboratori Nazionali del Gran Sasso (LNGS) in close vicinity to Borexino, a large liquid scintillator detector. This document reports on the $\bar{\nu}_e$ source production and transportation. The source should exhibit a long lifetime and a high decay energy, a requirement fulfilled by the ^{144}Ce - ^{144}Pr pair at secular equilibrium. It will be produced at FSUE “Mayak” PA using spent nuclear fuel. It will then be shielded and packed according to international regulation and shipped to LNGS across Europe. Knowledge of the Cerium antineutrino generator (CeANG) parameters is crucial for SOX as it can strongly impact the experiment sensitivity. Several apparatuses are being used or designed to characterize CeANG activity, radioactive emission and content. An overview of the measurements performed so far is presented here.

1. Introduction

The neutrino sector still shows unexplained discrepancies between observations and theoretical predictions. One of them is an electron neutrino deficit in short distance nuclear reactor based experiments, the so-called reactor antineutrino anomaly (RAA) [1]. A similar problem occurs in the source experiments GALLEX and SAGE [2]. This anomaly could be explained by a short distance oscillation between the three known neutrino states and one hypothetical new state with a squared mass difference of $\Delta m_{new}^2 \approx 1\text{eV}^2$ with the known neutrinos [3]. Due to constraints on Z boson coupling asserted by LEP [4], this state would be sterile.

The SOX experiment (Short baseline Oscillation in BoreXino) is designed to observe this oscillation, which has an expected length of 1 m order of magnitude, using a big liquid scintillator detector, in addition to the possible interaction rate deficit. Borexino is a spherical liquid

scintillator detector, with a fiducial mass of 280 tons, and provides vertex reconstruction with 15 cm precision and 5% energy resolution [5]. With a diameter of 8.5 m, the scintillator vessel allows to probe interaction rate as a function of particle travel distance from the source. $\bar{\nu}_e$ are detected through the inverse beta decay (IBD) process. It has a relatively high cross-section and allows a very good background rejection thanks to its rather unique signature: a time-correlated energy deposit from both the emitted positron and neutron. The study of the flux from a ^{144}Ce - ^{144}Pr $\bar{\nu}_e$ generator (CeANG), deployed in a tunnel right under the center of the detector base, is the first phase of the project.

2. CeANG design

2.1. Source selection

The source design of a β decay antineutrino generator must comply with some challenging constraints. First the source lifetime should be long enough to obtain statistically significant results with a 2 years schedule and to make possible the source manufacturing and transportation in a realistic amount of time. Second, the IBD process has a 1.806 MeV threshold so the source $\bar{\nu}_e$ spectrum must significantly extend beyond this threshold. This statement contradicts the previous one as it leads to a nucleus half-life shorter than the day. To circumvent this difficulty, the retained solution is a nuclei couple, where the mother nucleus satisfies the first requirement, and decays with a long half-life in a daughter nucleus having a high energy decay.

Four nuclei couples hold to this description: ^{42}Ar - ^{42}K , ^{90}Sr - ^{90}Y , ^{106}Ru - ^{106}Rh and ^{144}Ce - ^{144}Pr . The choice of the best suited couple is based on the following considerations: ease of production at the necessary scale, then available energy for the daughter nucleus decay to optimize the event rate. The ^{144}Ce - ^{144}Pr couple is the best candidate, as ^{144}Ce is a relatively abundant fission product from irradiated nuclear fuel and ^{144}Pr exhibits endpoint energy $Q_\beta = 2.997$ MeV well beyond IBD threshold [6]. With a ^{144}Ce half-life of 285 days, the minimal activity for 18 months of SOX data taking is 3.7 PBq.

2.2. Source production

The production of the CeANG requires industrial scale spent nuclear fuel reprocessing. Despite the $\sim 5\%$ cumulative fission yield of ^{144}Ce in commercial reactor fuel, gathering enough material to manufacture the source forces to reprocess tons of spent fuel, depending on the irradiation history and cooling times.

The FSUE “Mayak” PA has demonstrated the ability to handle such radioactive materials and will be in charge of the whole production process. The Cerium extraction begins by the standard PUREX (Plutonium Uranium Redox EXtraction) radiochemical process which allows to separate Uranium and Plutonium from the lighter fission products by liquid-liquid extraction. Then, complex displacement chromatography of rare-earth elements enables to isolate Cerium from the other fission products. The obtained Cerium residue undergoes calcination to form CeO_2 which will be pressed and inserted into a specially designed stainless steel capsule. To satisfy the high activity requirement, the source will contain up to 5 kg of CeO_2 . The expected delivery date of the CeANG is end of 2016.

3. Source handling and transport

3.1. Shielding

The ^{144}Pr decay produces γ rays. Among them the most difficult to handle is a 2.2 MeV ray with 0.7% intensity. Given the PBq activity of the source, it requires shielding both for biological safety and to avoid source-induced backgrounds into the detector. The flux must be suppressed by a factor $\gtrsim 10^{12}$.

The CeANG must also fit into the 1 m-width Borexino tunnel, putting constraints on the shielding compactness. The shielding should then have the highest possible density. For these

reasons, it is built in tungsten-iron-nickel alloy with $\sim 18 \text{ g/cm}^3$ density. The shielding thickness is 19 cm, leading to a $\phi = 54$ and $H = 60$ cm hollow cylinder in which the stainless steel source capsule will be placed. The total mass of the shielding is 2.4 tons, making it the biggest tungsten piece ever built. It is currently being manufactured at Xiamen Honglu Inc. (China). It will be delivered by the end of the year, for testing the different mechanical interfaces (calorimeter, transportation cask) and for some handling training.

3.2. Transportation

Once the capsule will be inserted in the shielding, the transportation will take place from Mayak to LNGS. The route has been planned and taken care of according to national and international regulations by Areva TN (France). The transportation of the CeANG must use a specific certified cask. Areva TN-MTR model, generally used for commercial nuclear fuel transportation, has been selected because of its capacity to handle the weight of the shielding. A custom basket has been designed to fit the shielding into the cask. The accreditations of the cask and basket as a source container for transport have been obtained, as well as transportation authorizations.

The selected route aims at minimizing border-crossing, which is lengthy and tedious to organize for nuclear material transportation. The cask will be delivered by train at Saint Petersburg, then a dedicated boat will bring it to Le Havre. The final step will be done by truck through France and Italy. The whole transportation is expected to take in any case less than three weeks, allowing for less than 5% activity loss. The delivery should take place at LNGS in December 2016.

4. Measurements for CeANG characterization

4.1. Activity

To guarantee the sensitivity of the SOX experiment over a wide range of sterile ν oscillation parameters, one of the key point is to be able to predict the interaction rate into the detector as a function of the $\bar{\nu}_e$ energy and distance to the source. The source activity is a key information to predict this global interaction rate. In order to measure the activity, two calorimeters are being built within the SOX collaboration. They aim at two independent subpercent activity measurements. The calorimeters are detailed in dedicated talks [7],[8].

4.2. β spectroscopy

To convert the thermal power, which is the raw result of the calorimeter measurements, to an activity, one should know the mean decay energy of the couple ^{144}Ce - ^{144}Pr . It depends on the β spectrum shape. This shape also influences directly the $\bar{\nu}_e$ interaction rate prediction as the $\bar{\nu}_e$ spectrum is estimated by converting the β spectrum. To test the parameter space favored by the RAA, the β spectrum shape should be determined at a 0.5% precision. It corresponds to an $\bar{\nu}_e$ interaction rate precision $\leq 1\%$.

The ^{144}Ce and ^{144}Pr spectra contain non-unique forbidden β transitions, for which the theoretical spectral shape is not known at this precision level. Furthermore past measurements of these isotopes show huge discrepancies at the scale of 10 to 15% [9][10]. It is then necessary to make a new β spectrum measurement for the CeANG.

Two β spectrometers are under development for this task, using plastic scintillator. The first have a multiwire-chamber to tag β rays by comparison to γ rays. It is developed by the Technische Universität München and has already been used for ^{238}U fission products spectrometry [11]. The second is designed specifically for SOX by CEA. It aims both at maximizing the light collection and doing fast and repeatable measurements.

FSUE “Mayak” PA has provided a first batch of radioactive samples which are used to check and tune the various apparatuses. Even if the source production process has evolved and the

first samples are not representative of the final CeANG, other samples will be sent during the year of production.

Due to the high lifetime difference, ^{144}Ce and ^{144}Pr are in secular equilibrium in source samples. As the ^{144}Pr $\bar{\nu}_e$ spectrum is the only one contributing to the IBD interactions in Borexino, a chemical extraction will be realized to isolate ^{144}Pr . The second spectrometer will then be used to make repeated hour-scale measurements of ^{144}Pr to achieve the best precision on its shape.

4.3. Source impurity assessment

In addition to calorimetry and β spectroscopy, a whole batch of measurements will help to characterize the content of the CeANG, as the production and handling environments could lead to traces of radioactive leftovers in the final source.

The γ spectroscopy is essential to assert the low background level needed by SOX. The CeANG must not show any gamma emitting impurities with an activity higher than $10^{-3} \text{ Bq/Bq}(\beta)$. This measurement is also done on the same source samples provided by FSUE “Mayak” PA using a high purity Germanium crystal detector. The first sample batch has shown to be within this specification.

To check the relative abundance of Cerium and to quantify the presence of long lifetime impurities and possible neutron emitting impurities, we also rely on α spectroscopy and inductive coupled plasma mass spectroscopy (ICP-MS). α spectroscopy test with gridded ionization chamber was particularly focused on ^{244}Cm and ^{241}Am and underlined neutron activity level lesser than $10^{-6} \text{ Bq/Bq}(\beta)$ in agreement with CeANG specifications. Both these nuclei could generate a problematic background as neutrons they emit can mimic an IBD event in the detector. On ICP-MS again no significant impurities were seen on the first tests, and the relative abundance of isotopes 140, 142 and 144 of Cerium were consistent with the sample origin.

5. Conclusion

The SOX project relies heavily on the feasibility of the radioactive sources production and their characterisation. The CeANG is by various sides an atypical object which has thus required a thorough process of design and validation for the associated tools. Now these early tasks are giving way to the final production phases of the experiment preparation.

During 2016, several characterization measurements will take place on radioactive samples and be cross checked to ensure a deployment with minimal troubles at the end of the year.

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