Towards $^{14}$C-free liquid scintillator

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Abstract. A series of measurements has been started where the $^{14}$C concentration is determined from several liquid scintillator samples. A dedicated setup has been designed and constructed with the aim of measuring concentrations smaller than $10^{-18}$. Measurements take place in two underground laboratories: in the Baksan Neutrino Observatory, Russia, and in the new Callio Lab in the Pyhäsalmi mine, Finland.

Low-energy neutrino detection with a liquid scintillator requires that the intrinsic $^{14}$C concentration in the liquid is extremely low. In the Borexino CTF detector the concentration of $2 \times 10^{-18}$ has been achieved being the lowest value ever measured. In principle, the older the oil or gas source that the liquid scintillator is derived from and the deeper it situates, the smaller the $^{14}$C concentration is supposed to be. However, this is not generally the case and the concentration is probably due to the U and Th content of the local environment.

1. Introduction

The intrinsic $^{14}$C concentration in a liquid is the main source of background at very low energies in high-purity liquid scintillation detectors. Previously measured concentration values are shown in Tab. 1 for scintillators based on PC (Pseudocumene; C$_9$H$_{12}$), PXE (Phenylxylylethane; C$_{16}$H$_{18}$) and Dodecane (C$_{12}$H$_{26}$). There are no published data for the $^{14}$C concentration in the LAB (Linear alkybenzene; C$_n$H$_{2n+1}$, $n=10–16$) being currently the most favourable liquid scintillator in large-volume detectors (e.g. SNO+ and JUNO).

The $\beta$-decay end-point energy of $^{14}$C is quite low, $Q=156$ keV, and the counting rate may be often lowered by setting the appropriate threshold energy. However, too high concentration of $^{14}$C in the liquid may results in pile-ups of pulses. For example, in the Borexino detector at Gran Sasso, Italy, the trigger rate is largely dominated by the $^{14}$C isotope [5] (with the concentration of $2 \times 10^{-18}$).
Table 1. Results of previous $^{14}$C concentration measurements in some liquid scintillators (CTF = Counting Test Facility).

| $^{14}$C concentration ($\times 10^{-18}$) | Liquid scintillator & fluor | Experiment | Ref. |
|----------------------------------------|-----------------------------|------------|-----|
| 1.94 ± 0.09                           | PC + PPO                    | Borexino CTF [1] |    |
| 9.1 ± 0.4                             | PXE + p-Tp + bis-MSB        | Borexino CTF [2] |    |
| 3.98 ± 0.94                           | PC-Dodecane + PPO           | KamLAND [3] |    |
| 12.6 ± 0.4                            | PXE + PPO                   | Dedicated setup [4] |    |

Based on the analysis of the $^{14}$C concentration in liquid scintillators derived from deep oil and gas fields [6], values lower than $10^{-18}$ should be achievable if the source is carefully chosen. The contamination from the reaction $^{14}$N(n, p)$^{14}$C is expected to be the main source of $^{14}$C also deep underground but now neutrons are emitted by the decay chains of U and Th isotopes.

A campaign has been started to measure the $^{14}$C concentration in several liquid scintillator samples (based on oil, gas and coal derivatives of different locations) with the aim of finding out concentrations smaller than $10^{-18}$. Measurements are being carried out simultaneously, with essentially similar instruments and rock overburden, in two deep underground laboratories: in the Baksan Neutrino Observatory, Russia [7] and in the new Callio Lab laboratory in the Pyhäsalmi Mine, Finland [8] at the depth over 4100 m.w.e.

2. Experimental details
In order to study the $^{14}$C concentration in liquid scintillators at the level lower than approximately $10^{-15}$, being currently the lower limit achieved by Accelerator Mass Spectrometry (AMS) method [9], a dedicated experimental setup is required. The central part of the detector setup of the present work consists of two low-activity PMTs (3” ET 9302B), two acrylic light guides and a quartz (or acrylic) vessel of 1.6 litres. The vessel and light guides are wrapped with VM2000 reflecting foil.

The shielding against $\gamma$ and neutron background is complemented using thick layers (10–15 cm) of copper and lead around the central part. Paraffin layer (approximately 10 cm, as the outer layer) may also be used to thermalize neutrons from the rock. The central part of the setup is planned to be flushed with nitrogen to reduce the background from radon.

The DAQ is realized with the DRS4 evaluation board (V5) [10] based on the DRS4 Switched Capacitor Array chip designed at the Paul Scherrer Institute, Villigen, Switzerland. The two PMTs are directly connected to the inputs of the DRS4 board which is connected to the DAQ Laptop via an USB connector. The DRS4 samples the pulse in 1024 bins of the width of 0.2 ns with the maximum sampling speed of 5 GS per second. A 2-channel high-voltage module NHQ 203M of Iseg Spezial Elektronik GmbH is used to power the PMTs.

3. Measurements
The liquid scintillator samples are purified using $\text{Al}_2\text{O}_3$ column and then mixed with $\sim 3$ g/ℓ of PPO and bubbled with nitrogen to remove oxygen. The purification is currently performed in the room atmosphere. A special purification system where the full process could be performed in a nitrogen atmosphere is in the design phase.

The energy calibration is performed with several $\gamma$-ray sources using the position of their Compton edges or the full absorption peak at low energies (around 100 keV). Sources currently in use include $^{57}$Co, $^{109}$Cd, $^{133}$Ba, $^{137}$Cs and $^{241}$Am. Essentially linear calibration curves are expected.
Figure 1. Calibration spectra of one of the ET9302B 3-inch PMTs. The spectra are obtained with voltages of 1100 V (black), 1150 V (red) and 1175 V (blue). The voltage 1150 V results in a gain of $10^7$.

The calibration of 3-inch ET9302B photomultiplier tubes has been carried out in a black box with a led light source. An example with three different high-voltage values for one of the tubes is shown in Fig. 1. In the data processing the digitized waveforms are analyzed and the signal shapes are used to reduce $\alpha$ and neutron induced backgrounds.

4. Results
There are no final experimental results available yet from Baksan or Pyhäsalmi measurements for any sample. However, as a preliminary result, a concentration value close to $10^{-17}$ has been obtained in the first measurement of a LAB samples of Russian origin.

5. Summary
A series of measurements have been started to determine the concentration of $^{14}$C in several liquid scintillator samples. The measurements are carried out in two deep underground laboratories: in Baksan, Russia, and in Pyhäsalmi, Finland. A preliminary result from the first measurement of a sample of Russian origin for the $^{14}$C concentration is close to $10^{-17}$.

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