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Measuring the ^{14}C content in liquid scintillators

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Abstract. We are going to perform a series of measurements where the $^{14}\text{C}/^{12}\text{C}$ ratio will be measured from several liquid scintillator samples with a dedicated setup. The setup is designed with the aim of measuring ratios smaller than 10^{-18} . Measurements take place in two underground laboratories: in the Baksan Neutrino Observatory, Russia and in the Pyhäsalmi mine, Finland. In Baksan the measurements started in 2015 and in Pyhäsalmi they start in the beginning of 2015. In order to fully understand the operation of the setup and its background contributions a development of simulation packages has also been started.

Low-energy neutrino detection with a liquid scintillator requires that the intrinsic ^{14}C content in the liquid is extremely low. In the Borexino CTF detector at Gran Sasso, Italy the $^{14}\text{C}/^{12}\text{C}$ ratio of 2×10^{-18} has been achieved being the lowest ^{14}C concentration ever measured. In principle, the older the oil or gas source that the liquid scintillator is derived of and the deeper it situates, the smaller the $^{14}\text{C}/^{12}\text{C}$ ratio is supposed to be. This, however, is not generally the case, and the ratio is probably determined by the U and Th content of the local environment.

1. Introduction

The intrinsic ^{14}C ($T_{1/2} \simeq 5700$ a) concentration in a liquid causes the main background at very low energies in high-purity liquid scintillation detectors. The measured concentration values are shown in Table 1 for scintillators based on PC (Pseudocumene; C_9H_{12}), PXE (Phenylxylylethane; $\text{C}_{16}\text{H}_{18}$) and Dodecane ($\text{C}_{12}\text{H}_{26}$). The values are essentially between 10^{-17} and 10^{-18} in $^{14}\text{C}/^{12}\text{C}$. There are no published data for the ^{14}C concentration values in the LAB (Linear alkylbenzene; $\text{C}_6\text{H}_5\text{C}_n\text{H}_{2n+1}$, $n=10-16$) being currently the most favourable liquid scintillator in large-volume detectors (e.g. SNO+ and JUNO).

The β -decay end-point energy of ^{14}C is quite low, $Q=156$ keV, and the counting rate from ^{14}C may be often handled by setting the appropriate threshold energy. However, too high concentration of ^{14}C in the liquid may result in pile-ups of pulses. For example, in the Borexino detector the trigger rate is largely dominated by the ^{14}C isotope [5] (with $^{14}\text{C}/^{12}\text{C} \simeq 2 \times 10^{-18}$).

Table 1. Results of previous $^{14}\text{C}/^{12}\text{C}$ concentration measurements in some liquid scintillators (CTF = Counting Test Facility).

$^{14}\text{C}/^{12}\text{C} (\times 10^{-18})$	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 possible if the source is carefully chosen. The contamination from reaction $^{14}\text{N}(\text{n}, \text{p})^{14}\text{C}$ is expected to be the main source of ^{14}C also deep underground but now neutrons are emitted by U and Th isotopes (and their daughters).

In order to measure the ^{14}C concentration in liquid scintillators at the level lower than approximately 10^{-15} , a dedicated experimental setup is most probably required (for example, similar to the Borexino CTF [1] or to the one in Ref. [4], or to the present work) since it is currently the lower limit achieved by Accelerator Mass Spectrometry (AMS) method [7].

A campaign has been started to measure the $^{14}\text{C}/^{12}\text{C}$ ratio in several different liquid scintillator samples (based on oil, gas and coal derivatives obtained from different locations) with the aim of finding out ratios 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 [8] and in the new CallioLab laboratory in the Pyhäsalmi Mine, Finland [9]. In Baksan the measurements have already been started (but no results available yet). In Pyhäsalmi the preparations have been started and the construction of the setup begins in January, 2016.

The present paper describes the procedure and plans for Pyhäsalmi measurements. For comparison, relevant information is also given from the setup in Baksan.

2. Experimental details

The central part of the detector setup consists of two low-activity PMTs (3" ET 9302B), two shaped acrylic light guides and a quartz (or acrylic) vessel of 1.6 litres. The setup is schematically illustrated in Fig. 1. The vessel and light guides are wrapped around by VM2000 reflecting foil.

The shieldings against γ and neutron background are slightly different in the two laboratories: In Pyhäsalmi thick layers (10–15 cm) of copper and lead around the central part are used. 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 flushed with nitrogen for reducing the background from radon. In Baksan the measurements are carried out in a dedicated low-background chamber where the central part is yet surrounded by a thick copper layer. Measurements are carried out deep underground in both sites: 4900 mwe in Baksan and 4000 mwe in the Pyhäsalmi.

The DAQ will be 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. An iseg NHQ 203M HV (2-chn) module is used to power the PMTs.

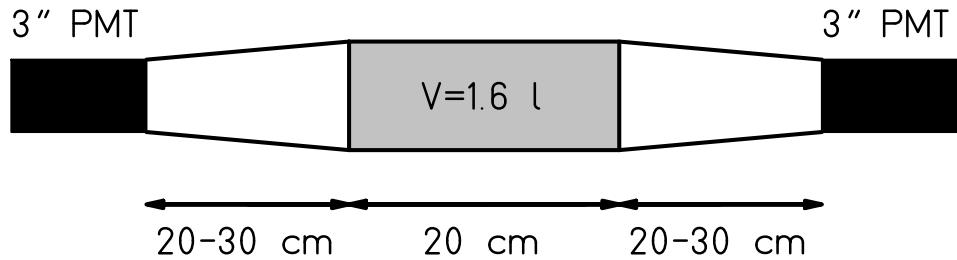


Figure 1. Schematic illustration of the experimental setup. It consists of a cylindrical vessel of 1.6 litres, two shaped light guides and two low-background 3-inch PMTs.

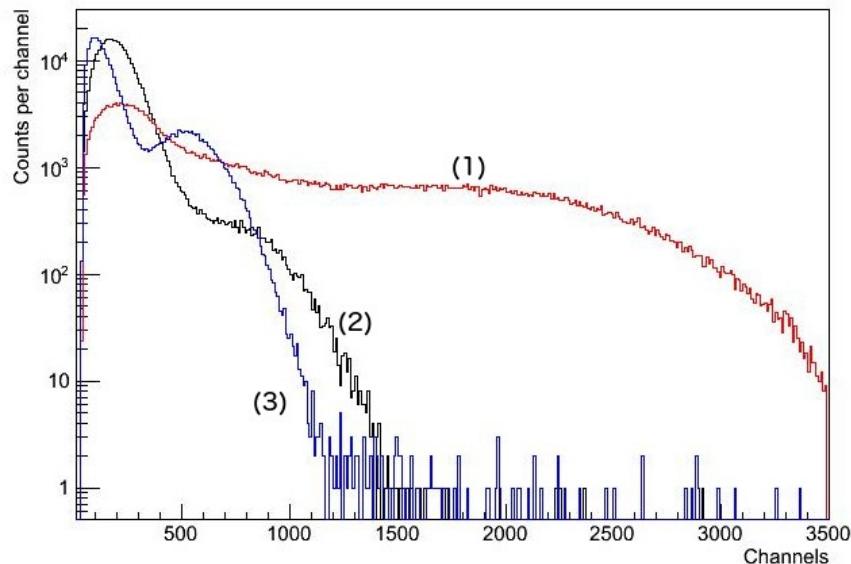


Figure 2. Calibration spectra of (1) ^{133}Ba , (2) ^{109}Cd and (3) ^{241}Am sources for the measurement performed in Baksan. For ^{133}Ba and ^{109}Cd the position of their Compton edges are used while for ^{241}Am the full absorption peak is possible.

3. Measurements

The liquid scintillator samples are purified using Al_2O_3 column and then mixed with $\sim 2 \text{ g}/\ell$ of PPO and bubbled with nitrogen for removing oxygen. The purification is performed in the room air. A special purification system where the full process could be performed in a nitrogen buffer is in the design phase.

The energy calibrations are performed with γ -ray sources using the position of their Compton edges. Three calibration sources are used in Pyhäsalmi (^{57}Co , ^{133}Ba and ^{137}Cs) and four sources in Baksan (^{109}Cd , ^{133}Ba , ^{137}Cs and ^{241}Am). ^{241}Am provides also the full-energy peak. Essentially linear calibration curves will be expected. In Fig. 2 the calibration spectra measured with ^{109}Cd , ^{133}Ba and ^{241}Am sources in Baksan are shown as an example.

In the data processing the digitized waveforms will be analyzed and the signal shape will be used to reduce α and neutron induced backgrounds.

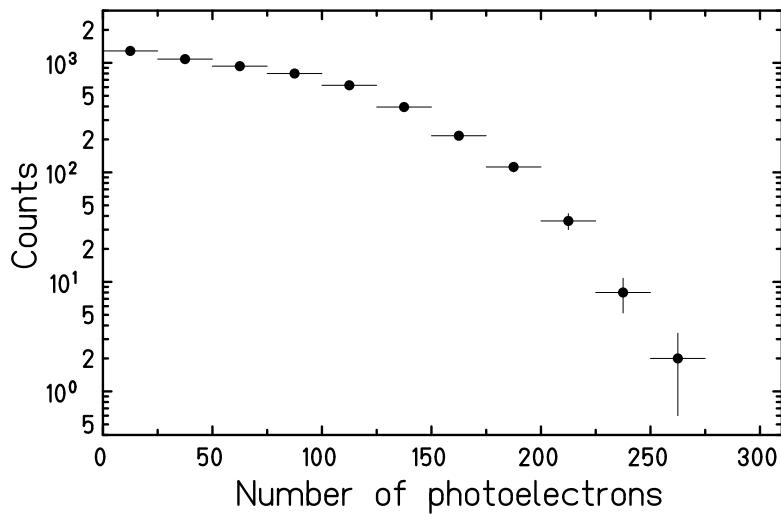


Figure 3. Simulated ^{14}C β -decay energy spectrum in a 1.6- ℓ LAB sample with the concentration of 10^{-17} ($^{14}\text{C}/^{12}\text{C}$) and measurement time of 28 days. Realistic assumptions for the light yield and PMTs were made (see the text for details).

4. Results

There are no experimental results available yet from Baksan or Pyhäsalmi measurements and also the Geant4-based simulation package is not yet complete. We present here a result of a simulation that was performed for studying the ^{14}C energy spectrum and energy resolution, i.e., number of photoelectrons released in the PMT cathodes.

In the simulations LAB (1.6 ℓ , $\text{C}_{18}\text{H}_{29}$, $\rho=0.856$ g/cm³, average light yield 10000 photons/MeV) was selected as the liquid. A realistic case was considered in the simulations by assuming the ^{14}C concentration of 10^{-17} and the measurement time of 672 h (=28 days). The quantum efficiency of $(28\pm 2)\%$ and gain of $(1.0\pm 0.1)\times 10^7$ were assumed for the two PMTs. The vessel and the light guides were wrapped around by the VM2000 reflecting foil in the simulations for increasing the photon collection.

The obtained ^{14}C β -decay energy spectrum (presented as a function of emitted photoelectrons at the cathodes) is shown Fig. 3

5. Summary

A series of measurements have been started where the concentration of ^{14}C will be determined in several liquid scintillator samples. The measurements are being carried out in two deep underground laboratories: in Baksan, Russia and in Pyhäsalmi, Finland.

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