Measuring of the ¹⁴C low abundance in liquid scintillator samples using small volume detector in low background chamber at Baksan

I.R. Barabanov¹, L.B. Bezrukov¹, A.V. Veresnikova¹, Yu.M. Gavrilyuk¹, A.M. Gangapshev¹, V.Yu. Grishina¹, V.I. Gurentsov¹, V.V. Kazalov¹, S.D. Krokhaleva¹,², V.V. Kuz'minov¹, A.S. Kurlovich¹, B.K. Lubsandorzhiev¹, S.B. Lubsandorzhiev¹, A.K. Mezhokh¹, V.P. Morgalyuk², P.Yu. Naumov⁴, G.Ya. Novikova¹, V.B. Petkov¹, A.M. Pshukov¹, A.Yu. Sidorenkov¹, V.V. Sinev¹,*, Sh.I. Umerov¹, E.A. Yanovich¹, T. Enqvist⁵, P. Kuusiniemi⁵, J. Joutsenvaara⁵, A. Virkajarvi⁵ and V.P. Zavarzina¹

¹Institute for Nuclear Researche of Russian Academy of Sciences, Prospekt 60-letia Oktyabrya 7a, 115409 Moscow, Russia, vsinev@inr.ac.ru

²Moscow Institute for Physics and Technology (State University), Vavilova 10, 115409 Moscow, Russia

³A.N. Nesmeyanov Institute of Organoelement Compounds of Russian Academy of Sciences, Vavilova 28, 115409 Moscow, Russia

> ⁴National Research Nuclear University (MEPHI), Kashirskoe shosse 31, 115409 Moscow, Russia

⁵Oulu University, CUPP Pyhäsalmi, Finland

Abstract The scintillation detector was constructed to research ultralow concentrations of ¹⁴C in liquid scintillator samples. The detector is placed in the low background Laboratory BNO INR RAS at a depth of 4900 m.w.e. Measurements of ¹⁴C abundance was done for the samples of liquid scintillator on base of linear alkylbenzene. The detector counting rate was measured and the ratio ¹⁴C/¹²C was extracted from the experimental spectrum the LAB sample. The background model was developed and applied for ¹⁴C abundance analysis. The value obtained is ¹⁴C/¹²C (9.1 ± 1.0) ×10⁻¹⁶.

Keywords: Scintillator, radiocarbon, detector, radioactivity

1. Introduction

To study natural neutrino fluxes, a scintillation detector with a mass of at least 10 kt is required. To determine the component of the geo-neutrino flux from 40 K, the scintillator should be several orders of magnitude cleaner than that used to measure the flux of solar neutrinos [2]. In addition, it is necessary to get rid of the 14 C isotope contained in the scintillator, which prevents the study of solar neutrinos from the pp cycle and the geo-neutrino flux from 40 K.

A program for the search for a liquid scintillator with a reduced content of ¹⁴C is proposed in the Institute for Nuclear Research. For this purpose, an installation with a small scintillation detector for the study of liquid scintillator samples was created in the underground low-background laboratory of the Baksan Neutrino Observatory of the Institute of Nuclear Research of the Russian Academy of Sciences. The possibility of using a small volume detector for measuring the concentrations of ¹⁴C is indicated in [3], where a 1.5-liter detector was used.

The bench can also be used to study the background of a liquid scintillator loaded with neodymium (¹⁵⁰Nd), which is intended to be used to study double beta decay.

2. Low background detector

The detector is located in the underground low-background laboratory of the BNO of the Institute of Nuclear Research of the Russian Academy of Sciences [5] and is intended for measurements of ultralow concentrations of the ¹⁴C isotope in samples of a liquid organic scintillator. The laboratory is located inside the mountain (3,700 m from the entrance to the tunnel), at a depth of 4900 m.w.e., where the muon flux is ~ 0.1 m⁻² hr⁻¹ [6]. To suppress the background from neutrons and gamma quanta of the surrounding rocks, the walls, floor and ceiling of the room where the scintillation detector is installed are consistently made of layers of polyethylene (25 cm), cadmium (1 mm) and lead (15 cm). The detector itself is placed in a box of plexiglass $14.5 \times 14.5 \times 120$ cm³ and is surrounded on all sides by a protection from extremely pure copper 15 cm thick and lead 10 cm thick. The principle diagram of the detector is shown in Fig. 1. The detector includes a quartz cell with a diameter of 100 mm and a length of 200 mm made of quartz glass 3 mm thick (a full volume of about 1.5 L) filled with a sample of liquid organic scintillator, two cylindrical optical fibers made of organic glass (PMMA) with a diameter of 90 mm and length 50 mm, and two low-background photomultipliers (PMT) ET9302B (3"). To increase the light collection, the quartz cell and lightguides are wrapped in a mirror reflective film of VM2000. For better optical contact between the quartz cell, light guides and PMT, a silicone lubricant was used. The sealed polyethylene cover surrounding the detector from the outside, served to protect against radon. From the internal volume, radon was removed by purging with nitrogen gas.

The conditions for measuring ultralow concentrations of radiocarbon make it necessary to use materials with a low content of radioactive impurities in the detector construction. Using a low-background semiconductor detector from HPGe high-purity germanium, measurements were made of the intensity of gamma quanta of a quartz cell and a photomultiplier ET9302B. According to the measurements, calculations were made of the content of radioactive impurities (Bq/kg) in the cell and the photomultiplier in Fig. 1.



Fig1. Low background detector scheme.

To further improve the background characteristics of the scintillation detector, it is planned to use more low-background photomultipliers and optimize the protection of the cell from the radiation from the voltage dividers of the photomultiplier.

3. Scintillator samples

The linear alkyl benzene (LAB) obtained from China was studied. A sample of a liquid

scintillator with an additive of 4 g/l PPO was prepared.

LAB is a mixture of hydrocarbons with the general formula C_nH_{2n-6} , density 0.856 g/l and a flash point of 143° C [8, 9]. LAB has the average formula $C_{17.73}H_{29.46}$ and is a mixture of four isomeric alkylbenzenes with the content: $C_{16}H_{26} - 0.125$, $C_{17}H_{28} - 0.293$, $C_{18}H_{30} - 0.315$, C1 $C_{19}H_{32} - 0.267$, each of which is present as a mixture of isomers of linear structure differing in the position of the phenyl residue in the hydrocarbon chain.

The light yield for a liquid scintillator based on a LAB (~ 8000 photons/MeV) and a coefficient of attenuation of a parallel light beam (15 m at a wavelength of light 420 nm) was obtained. The values obtained allow measurements in the low-energy (<50 keV) region of the beta-spectrum of radiocarbon.

For the measurements we used samples of a scintillator with a volume of 1360 ml, which are completely placed in a 1.5-liter cell.

4. Energy calibration

For the energy calibration of the detector, gamma-ray sources were used: ²⁴¹Am, ¹³³Ba, ¹³⁷Cs, ⁶⁰Co, ²²Na and ²³²Th (²⁰⁸Tl). The recoil energy of the recoil electron is also presented here for the backward scattering of the gamma quantum and the energy at the full absorption peak (FAP) for low-energy quanta. The energy for the maxima in the experimental distributions, which was used for calibration, is given.

5. Detector background

Solvent and copper protection of the detector passed special purification from radioactivity. Therefore, the equilibrium in the decay products of the natural radioactive chains of uranium and thorium is disrupted. If we assume that after the purification only the uranium and thorium isotopes remained, and the products of their decay were removed, then in 5 years, the thorium will again be in equilibrium with its products, and the uranium will have an equilibrium only of the ²³⁴U isotope whose half-life is 2.45×10^5 years. Then the radon background will become independent, which can penetrate into the protection slots and fall into the sample of the scintillator during overflow. There is a background of ⁴⁰K, present in the glass and voltage divider faux. Cherenkov radiation from recoil electrons in the detector's optical fibers, caused by Compton scattering of energetic gamma quanta, can contribute.

Thus, we represent the background of the detector consisting of the following components:

- 1. Internal background from 238 U to 234 U,
- 2. The same external background, coming from the copper shield,
- 3. Internal background from ²³²Th,
- 4. The same external background, coming from the copper shield,
- 5. Internal background from ²²²Rn,
- 6. The same external background, coming from the copper shield,
- 7. External background of glass and faucet dividers from 40K,

8. The background of the Cherenkov light from the light guides together with the background of the pmt itself.

6. Determination of the contribution of ¹⁴C decays to the number of scintillation cell events

The measurement of the ¹⁴C content in the scintillator volume was carried out for 322.9 hours.

A digital oscilloscope National Instruments NI5105 was used to record the charge in the pulse from each photomultiplier. Trigger was the signal from one of the photomultiplier. The measurements were carried out in series, each of which was accompanied by two calibrations: one before the measurement, the other after.

The charge spectrum in the pulse of each series was transferred to the energy spectrum using its average calibration. Then the spectra were added with a weight equal to the measurement time.

The measured spectrum was fitted with simulated background spectra. The remaining part was assigned to the spectrum from ¹⁴C.

At this stage, the scintillator was not blown with nitrogen to saturate it with carbon dioxide containing ¹⁴C in a larger proportion. The figure shows the experimental spectrum, fitted with simulated backgrounds.



Fig2. Experimental spectrum from low background detector. Components of background are shown.

Taking into account the volume of the scintillator (1360 ml), the value ${}^{14}C/{}^{12}C = (9 \pm 1) \times 10^{-16}$ for this LAB sample was obtained. Earlier, for the same sample, a value of $(5.5 \pm 1.0) \times 10^{-16}$ was obtained, and an even earlier measurement yielded a value $(3 \pm 1) \times 10^{-17}$. One can see the effect of saturation of the scintillator with carbon dioxide. Taking into account the limiting solubility of CO₂ 1.18 of the scintillator volume [10], it is possible to estimate the ${}^{14}C$ content in the scintillator itself. In our case, we did not saturate the scintillator to the limit, so we take the amount of CO₂ in half of the limit, that is, 0.5 volume. We get here ${}^{14}C/{}^{12}C < 4 \times 10^{-16}$.

7. Conclusion

An installation for measuring its own background and the content of radiocarbon ¹⁴C in samples of a liquid scintillator was created.

In our work, it is proposed to study samples of a scintillator with a base of a solvent obtained from various petroleum feedstocks to determine the effect of the deposit on the ${}^{14}C$ content. Solvents obtained from coal will also be investigated.

The background of the detector is analyzed in order to be able to further suppress it and lower the detector threshold for more confident separation of the ¹⁴C beta spectrum. A model of the total detector background was created and successfully applied to the description of the experimental spectrum.

The setup will be used to measure the background of the scintillator with dissolved Nd for testing the methods of scintillator purification from natural radioactivity.

Acknowledgments

Authors are grateful to the organization committee of The International Conference SN 1987A, Quark Phase Transition in Compact Objects and Multimessenger Astronomy (held at 2-8 July of 2017, KBR, Terskol (BNO INR); KChR, Nizhnij Arkhyz (SAO)) for their kind invitation to present the talk.

5. References

[1] *I. R. Barabanov, L. B. Bezrukov, A. V. Veresnikova et al.* Large-Volume Detector at the Baksan Neutrino Observatory for Studies of Natural Neutrino Fluxes for Purposes of Geo- and Astrophysics // Phys. At. Nuc., 80, 446, 2017.

[2] Bellini G., et al. (Borexino coll.) // Phys. Rev. Lett. 107, 141302 (2011); O. Yu. Smirnov et al. // arXiv:1507.02432 [hep-ex].

[3] C. Buck at al. // Instrum. Exp. Tech., 55, 34 (2012).

[4] *B. S. Dzhelepov and L. N. Zyrianova* Influence of Atom electromagnetic field on beta-decay. // USSR Academy of Sciences, Moscow 1956.

[5] Ju. M. Gavriljuk, A. M. Gangapshev, A. M. Gezhaev et al. // arXiv: 1204.6424 [physics.ins-det].

[6] V. N. Gavrin, V. I. Gurentsov, V. N. Kornoukhovet et al. // Preprint INR-698 Moscow 1991 (rus).

[7] I. R. Barabanov, G. Ya. Novikova, V. V. Sinev, E. A. Yanovich // arXiv: 0908.1466 [hep-ph].

[8] I. R, Barabanov, L. B. Bezrukov, N. A. Danilov et al. // Journal of Applied Chemistry 84, 385, 2011.

[9] L. B. Bezrukov, N. I. Bakulina, N. S. Ikonnikov et al. // Preprint INR-1382 Moscow 2014 (rus).

[10] I. R, Barabanov, L. B. Bezrukov, B. K. Lubsandorzhiev, V. P. Morgalyuk, G. Ya. Novikova, E. A. Yanovich // Preprint INR-1339 Moscow 2012 (rus).