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BACKGROUND COMPONENTS OF A LIQUID SCINTILLATION COUNTER IN THE ^{14}C WINDOW

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ABSTRACT. We present a broad and detailed study of the background components of a liquid scintillation (LS) detector, using a simple laboratory-built system, ICELS. It was specifically designed for radiocarbon dating and is compact and easily transportable (total weight 35 kg). Its flexible LS detector unit has a dome-shaped vial with 3 mL benzene to which 45 mg butyl-PBD is added. The vial sits on the top of a vertical 28-mm-diameter phototube. The gamma radiation, to which the benzene is exposed under varying conditions, was measured by replacing the vial with a 38-mm-diameter NaI crystal. The pulse-height spectra of the ^{14}C LS background and the NaI gamma background were measured in a surface laboratory and in a deep underground counting room with: 1) a lead shield of varying thickness; 2) lead of normal and low ^{210}Pb concentration; 3) phototubes of 2 different types; and 4) varying benzene volume. The beta emission from the face of the tubes was measured with a low-level Geiger counter.

INTRODUCTION

The background count rate (here simply called "background") of liquid scintillation (LS) counters limits their sensitivity. In spite of 5 decades of LS radiocarbon dating, our knowledge of the background components of these systems is fragmentary (Theodórsson 1998) because systematic studies are lacking as the dominating LS systems with 2 photomultiplier tubes (PMT) working in coincidence are not suitable for studies of this kind. The compact and flexible single-PMT detector unit of the system used here, ICELS (Theodórsson 2005), is ideal for this work. We present a detailed study of both the ^{14}C LS background of 3 mL benzene and the gamma background radiation, to which the vial is exposed, measured by replacing the vial on the top of the PMT with a NaI crystal. The measurements were carried out both in our laboratory and in a deep underground counting room.

EXPERIMENTAL

ICELS comprises a detector unit, a dedicated electronic unit, and a laptop computer (Theodórsson 2005). A multichannel analyzer (MCA) is added in the present study. Figure 1a depicts a schematic diagram of the simple and flexible LS detector unit. A 28-mm-diameter flat-end face PMT is clamped vertically. When the LS ^{14}C background is measured, a dome-shaped quartz vial with 3 mL benzene (with 45 mg butyl-PBD) sits on the top of the tube. The vial is wrapped (except for the bottom, facing the PMT) with 2 layers of a thin Teflon® tape, which gives optimal light reflection.

The stabilized high-voltage supply of ICELS operates the PMT at 500–800 V. At the selected ^{14}C high-voltage balance point (Theodórsson et al. 2003), the ^{14}C counting window spans beta energy ranges from 23.5 keV to 126 keV, which gives about 71% ^{14}C detection efficiency.

To study the gamma flux to which the sample vial (with 3 mL benzene) is exposed (including that coming from the PMT), a 38-mm-diameter, 38-mm-long NaI crystal is put on the top of the vertical PMT, replacing the vial (Figure 1b). The mass of the crystal is about 160 g, 60 times the mass of 3 mL benzene. The crystal was donated to our institute some 10 yr ago by Bicron Corporation for our low-level studies.

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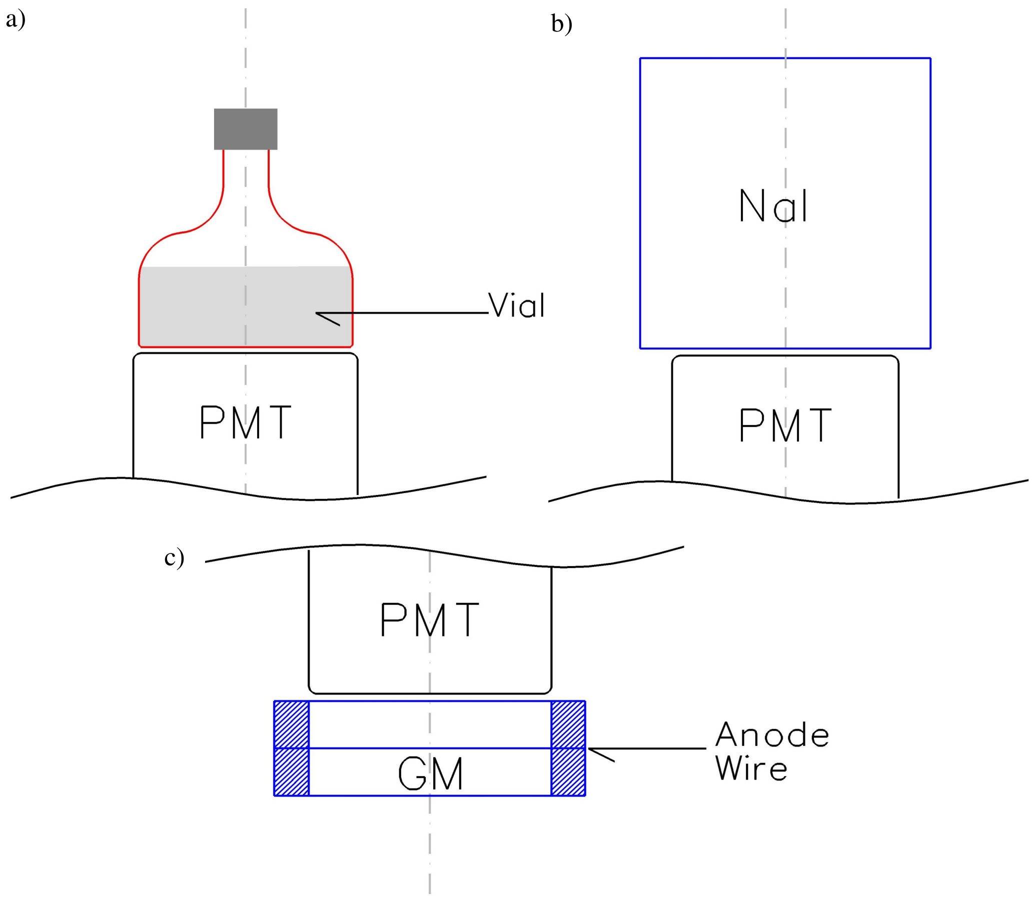

Figure 1 Detectors used in the study: a) 3-mL vial with PMT; b) NaI with PMT; c) low-level Geiger counter for measuring beta emission from the face of the PMT.

Furthermore, the beta emission from the face of the PMTs used was measured with a low-level, thin window (diameter 25 mm) Geiger flow-counter, made in our laboratory (Figure 1c). Its background is 6 counts per hour (cph) in the underground counting room.

Our laboratory has 3 concrete floor plates above it. The underground counting room is a closet in the wall of a road tunnel under a fjord, 37 km from our laboratory. It lies 165 m below the surface of the sea, where the cosmic ray flux is reduced by a factor of about 300 compared to its surface value. The cosmic radiation background component in our LS and NaI detectors is therefore negligible in this room.

BACKGROUND COMPONENTS

The source of the LS ^{14}C background can be divided into the following components (Theodórsson 1996):

1. K/Th/U in detector unit and inner layer of lead shield;
2. K/Th/U in building and other materials in the vicinity of the system;
3. Cosmic radiation;
4. Residual, unexplained background component.

Background Due to K/Th/U in Detector Unit

K/Th/U is always present in PMTs, mainly in their glass bulb. We use here 2 different types of PMTs (Table 1), both produced by ET Ltd (United Kingdom) and made specially for our low-level studies. This service has been an invaluable help in our work.

In this work, we used 2 types of PMTs (Table 1). Type D190XSA, produced in 1992, has a bulb made from the best glass available at that time, designated with the letter X by the producer. Later in the 1990s, glass with considerably reduced K/Th/U concentrations became available, presumably in limited quantity, as a result of a large effort to produce better PMTs for huge LS detectors used in the study of neutrinos and dark matter. In 2003, ET Ltd produced, again specially for our study, a new PMT, D842SA, where the bulb is made of improved glass, B53 (Table 1). The K/Th/U concentrations in the glass of these tubes, as given by the manufacturer, are shown in Table 1. D842SA has been used in the following studies unless otherwise stated.

Table 1 K/Th/U in glass of photomultiplier tubes.

	Year of production	Type of glass	^{40}K (mBq/g)	Th (mBq/g)	U (mBq/g)	Geiger cph[a]
D190XSA	1992	ET-X	9	1.0	1.2	57
D842SA	2003	ET-B53	1.8	0.12	0.3	6
Ratio D190/D842			5	8	3.2	9

[a]cph = counts per hour.

As beta particles from K/Th/U in the face of the PMT may penetrate the bottom of the vial and thus contribute to the background, this beta emission was measured with the low-level Geiger counter, with the front end of the vertical tube touching the window (Figure 1c).

NaI Underground Measurements

The relative K/Th/U concentrations in the 2 types of PMTs used was studied by measuring their NaI background spectrum in the underground counting room inside a 10-cm-thick lead shield, where the background components due to external K/Th/U and cosmic radiation are negligible (see next section). The D190XSA spectrum (Figure 2) shows a clear ^{40}K peak (1460 keV) and a weak one of the uranium series (1720 keV). Only a faint ^{40}K gamma peak is possibly seen in the spectrum of D842SA.

As the K/Th/U gamma peaks of the D842SA NaI spectrum should be a factor of 5, 8, and 3 times lower than for D190XSA, the background gamma contribution of these radioactive materials is practically negligible in the spectrum of D842SA. Nevertheless, we are left with a significant NaI background spectrum. The most likely explanation is that it is coming from beta particles in the face of the D842SA PMT that can penetrate the glass window of the crystal unit.

Another possible explanation is that the background comes from primordial radioactivity in the lead. K/Th/U are separated very effectively from lead in the chemical refining process, but it could come from ^{210}Pb (half-life 22 yr). The main penetrating radiation is then the bremsstrahlung produced in the lead by energetic ^{210}Bi beta particles. Both the NaI and LS background spectra were measured with the inner 5-cm layer of lead with ~300 Bq/kg of ^{210}Pb and then with Boliden lead, produced in the early 1960s, with ~5 Bq ^{210}Pb/kg today. No difference is seen in the 2 background spectra. This background source can therefore be ruled out, and it shows that selecting lead with low ^{210}Pb concentration gives practically no improvement.

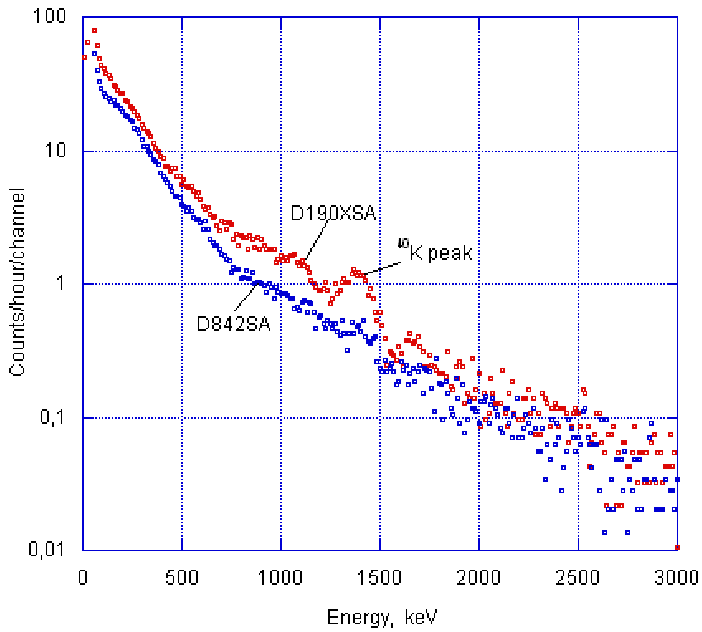

Figure 2 The background spectra with the NaI crystal on PMTs D842SA and D19XSA behind 10 cm of lead in the underground counting room. The spectra show the average values of 5 channels of a 256-channel multichannel analyzer (MCA).

LS Underground Measurements

The LS background spectrum of 3 mL benzene was measured in the energy range 0 to 160 keV with the 2 PMTs under the same circumstances as above. In order to improve the statistics, at the sacrifice of energy resolution, the average value of every 5 channels of a 256-channel spectrum is shown (Figure 3). The difference in the 2 spectra above 80 keV is small, but below 80 keV the difference increases.

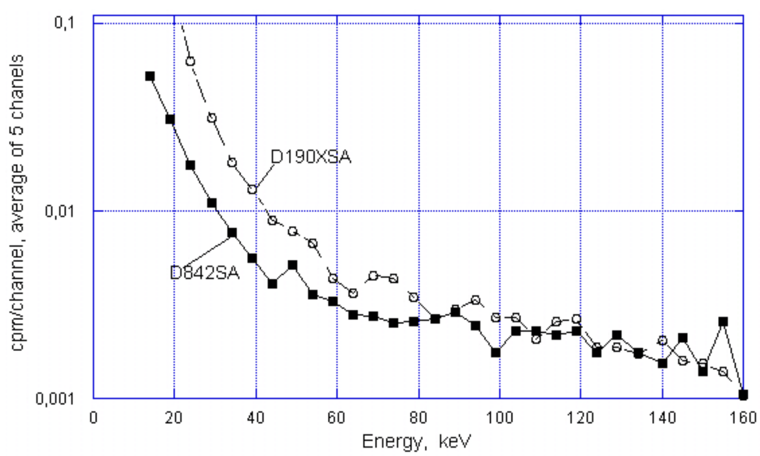

Figure 3 The LS background spectra of PMT D842SA and D190XSA behind 10 cm of lead in the underground counting room.

The ^{14}C window background in the underground counting room is 0.41 cpm for D842SA and 0.72 cpm for D190XSA. The difference, 0.31 cpm, represents an important improvement when cosmic radiation is absent or eliminated with an anticosmic counter. For low-level work, D842SA should be used.

The source of the LS underground background spectrum of D842SA in the ^{14}C window is presumably the same as described above for the NaI spectrum.

External Gamma Radiation

The external gamma flux, to which the benzene samples are exposed, comes from K/Th/U in materials in the vicinity of the detector, outside the shield. This flux was measured with an unshielded NaI crystal (described below). It is similar in the underground counting room (0 cm Pb in Figure 4) and in our laboratory. The LS detector must be shielded from this external gamma radiation. It is important to determine how thick the lead shielding must be to reduce this background component to a negligible value.

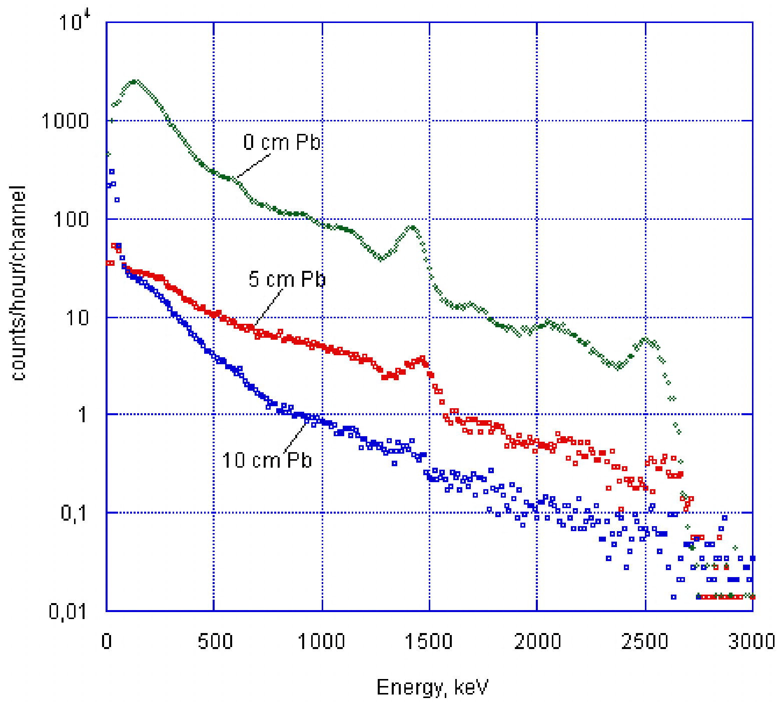

Figure 4 The NaI background spectra in the underground counting room, unshielded (0 cm) and inside 5- and 10-cm-thick lead shields.

The effect of shielding the detector with lead from this radiation was studied underground by measuring 1) the NaI background spectrum (0–3000 keV) without a shield and with 5- and 10-cm-thick shields of lead and 2) the LS ^{14}C background spectra (0–160 keV) under the same shielding conditions.

NaI Measurements

Figure 4 shows the NaI spectra on a logarithmic count rate scale. It shows that 5 cm of lead reduces the spectrum by a factor of about 12, as was expected. Increasing the lead thickness from 5 to 10 cm reduces the background less, by a factor of about 5 above 1000 keV, but below this energy, the difference decreases gradually, and under 100 keV there is practically no difference. The most probable explanation is the same as discussed above, i.e. the emission of beta particles from the face of the PMT.

LS Measurements

Figure 5 shows the LS background spectra (5-channel averages) in the energy range from 0 to 160 keV, measured with the same shielding configurations as above. As to be expected, the background is reduced by a factor of about 12 by 5 cm of lead, but adding a further 5 cm gives practically no further background reduction. Again here, the background is probably dominated by beta particles from the face of the PMT.

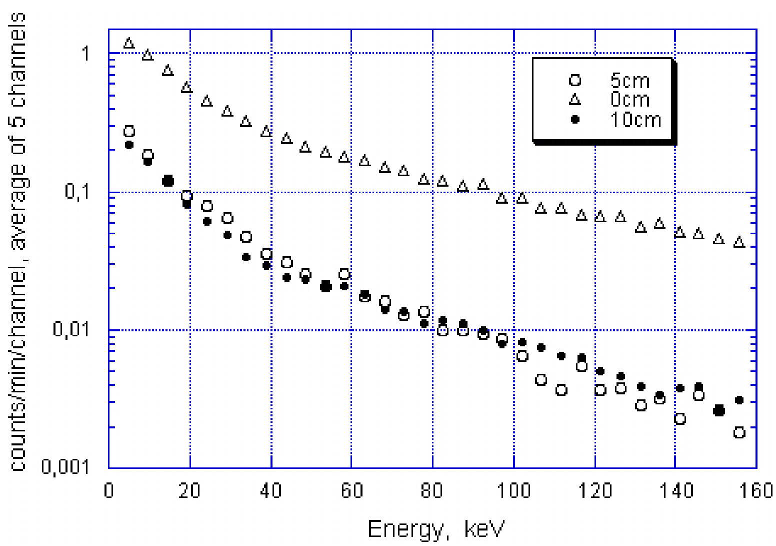

Figure 5 The LS background spectra in the underground counting room, unshielded and inside 5- and 10-cm-thick lead shields.

The result of this study shows that when ^{14}C is measured at our background count rate level (2–3 cpm), using vials of glass, nothing is gained by having a shield of lead that is thicker than 5 cm, and that the selection of lead for the shield is not critical.

^{14}C Background Due to Cosmic Radiation

NaI Measurements

The gamma background spectrum was measured with the NaI crystal (Figure 6) behind 10 cm of lead shield, both in our laboratory and in the underground counting room. In the surface laboratory, only 1 peak is seen, at 511 keV, which is due to the annihilation of positrons produced in the lead by the cosmic radiation. Underground, no gamma peak is seen, except perhaps a very faint ^{40}K peak at 1460 keV due to a trace of potassium in the glass of D824SDA.

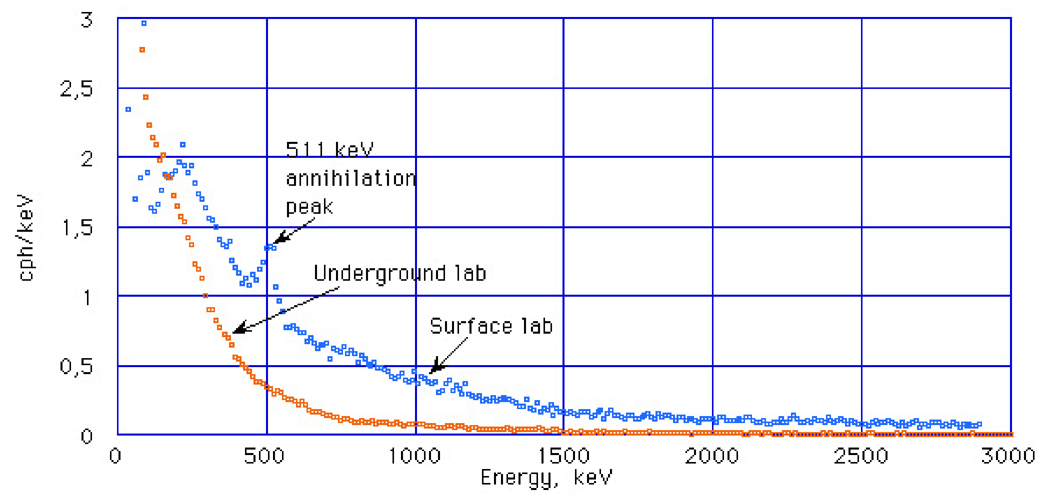

Figure 6 The NaI background spectra, with the crystal inside a 10-cm-thick shield of lead, in the surface laboratory and in the underground counting room.

Figure 7 shows the LS background spectra under the same conditions. The background in the ^{14}C counting window (from 23.5 to 126 keV) in the surface laboratory is 1.62 cpm and 0.41 cpm in the underground counting room. The background contribution of cosmic radiation in the surface laboratory is therefore 1.21 cpm for 3.0 mL of benzene, or 75% of the total background.

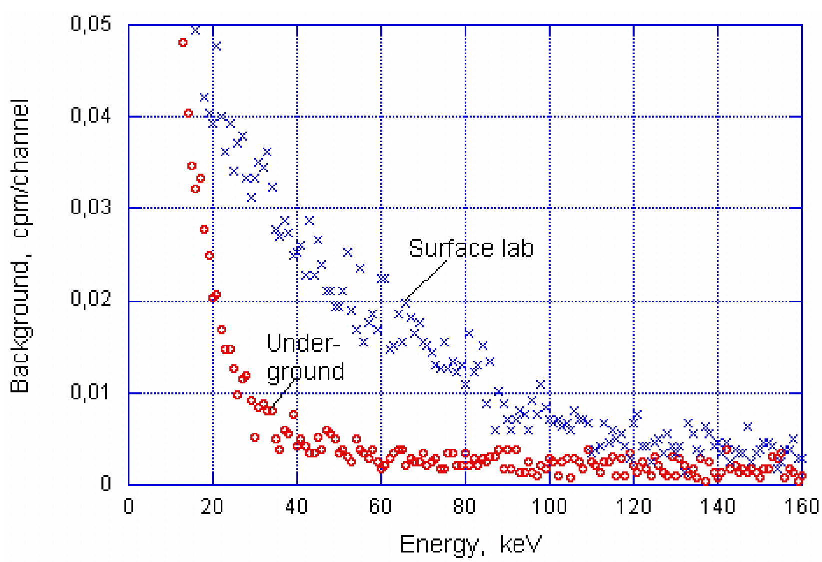

Figure 7 LS background spectra (3 mL benzene) with a 10-cm-thick lead shield in our surface laboratory and in the underground counting room.

A moderate overburden of 15 m of rock reduces the cosmic ray flux by a factor of about 10 (Theodórsson 1996), which would give a ^{14}C background of 0.44 cpm for this detector, a very attractive value. Because of the compactness of the system, it would be easy to operate it in an underground public transportation channel, where an overburden in this range is common.

Background at Varying Benzene Volume

Figure 8 shows the ^{14}C background in our surface laboratory at varying benzene volume, where the detector is shielded by 10 cm of lead. In this late phase of our study, we used cylindrical 4-mL glass vials. The figure shows that the increase in the background count rate varies linearly with volume, but it is not proportional to the volume, because the mass of the vial is a part of the sensing material, as some of the Compton electrons released in the glass may penetrate into the benzene.

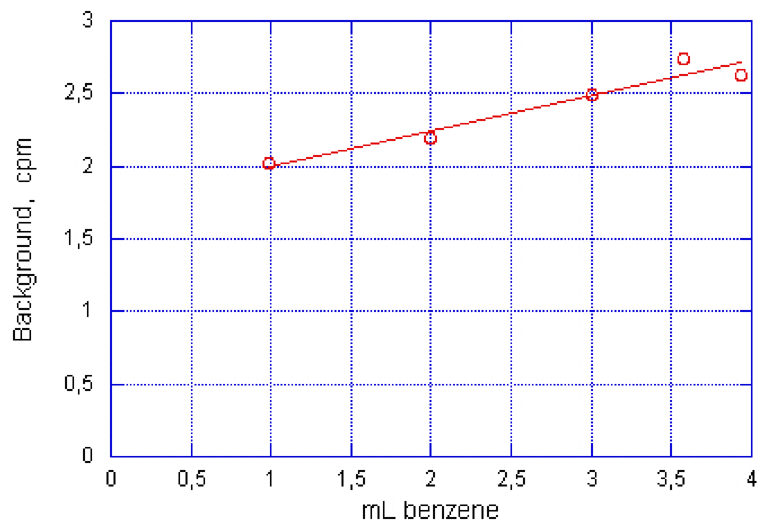

Figure 8 The LS background at varying benzene volume, measured in our surface laboratory.

In Quantulus™ machines, the relation between benzene volume and ^{14}C background with Teflon vials is different; the background is nearly proportional to the benzene volume (Plastino and Kaihola 2004). The most plausible explanation for the difference between the 2 systems is that pulse-shape analysis is applied in Quantulus, which detects ionization in the glass, which produces fluorescence with longer decay time than fluorescence in benzene, causing a tail in these pulses that marks them as external background. Only events causing ionization in the benzene but not in the glass are registered in the ^{14}C channel. The vial thus acts as a guard detector.

CONCLUSION

The background of a single-PMT liquid scintillation counting system with 3 mL of benzene in a specially made vial of quartz has been studied in detail by a variety of methods. Using a PMT (D84SA) made of new high-purity glass, the ^{14}C background was 1.62 cpm in a surface laboratory, of which 1.21 cpm is due to cosmic radiation. The background in a deep underground counting room was 0.41 cpm. Using D190XSA, made in 1992, gives 0.31 cpm higher background. The study shows that increasing the thickness of the lead shield from 5 cm to 10 cm gives a negligible background reduction for our system in the ^{14}C counting window.

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