Applied Radiation and Isotopes 105 (2015) 209–218

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Development of a low background liquid scintillation counter for a shallow underground laboratory

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HIGHLIGHTS

- Graded-shielding can produce an ultra-low-background liquid scintillation counter.
- Location in a shallow underground cleanroom further enhances background reduction.
- A novel light collection design and selected low background materials are utilized.
- The background is predicted to be 10–100 times below typical commercial systems.
- Simulations tentatively predict a background rate of order 10 counts per day.

ARTICLE INFO

Article history:
Received 2 March 2015
Received in revised form
5 August 2015
Accepted 18 August 2015
Available online 20 August 2015

Keywords:
Liquid scintillation counting
Low background
Shallow underground laboratory

ABSTRACT

Pacific Northwest National Laboratory has recently opened a shallow underground laboratory intended for measurement of low-concentration levels of radioactive isotopes in samples collected from the environment. The development of a low-background liquid scintillation counter is currently underway to further augment the measurement capabilities within this underground laboratory. Liquid scintillation counting is especially useful for measuring charged particle (e.g., β and α) emitting isotopes with no (or very weak) gamma-ray yields. The combination of high-efficiency detection of charged particle emission in a liquid scintillation cocktail coupled with the low-background environment of an appropriately designed shield located in a clean underground laboratory provides the opportunity for increased-sensitivity measurements of a range of isotopes. To take advantage of the 35 m-water-equivalent overburden of the underground laboratory, a series of simulations have evaluated the scintillation counter's shield design requirements to assess the possible background rate achievable. This report presents the design and background evaluation for a shallow underground, low background liquid scintillation counter design for sample measurements.

1. Introduction

To further enhance the low-level radiation detection capabilities of the Pacific Northwest National Laboratory shallow underground laboratory (Aalseth et al., 2012), the development of a low-background liquid scintillation counter is underway. The instrument is being developed to measure a range of low-level beta-emitting isotopes collected in the environment such as tritium, ^{14}C, strontium isotopes, and others. Such measurements impact a range of sciences including studies of environmental transport mechanisms, biological radio-isotopic dating, radioactive fallout, or tracking of nuclear accident effluent (Salonen et al., 2012).

Liquid scintillation counting (LSC) identifies radioactive decay through detection of the ensuing scintillation photons produced by charged particles emitted into the scintillation cocktail. The photons are detected and this signal is amplified with one or more photomultiplier tubes (PMTs) for processing in a data acquisition system. Liquid scintillation counting provides a mechanism for counting β and α emitters down to very low concentrations, due

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http://dx.doi.org/10.1016/j.apradiso.2015.08.027
0969-8043/

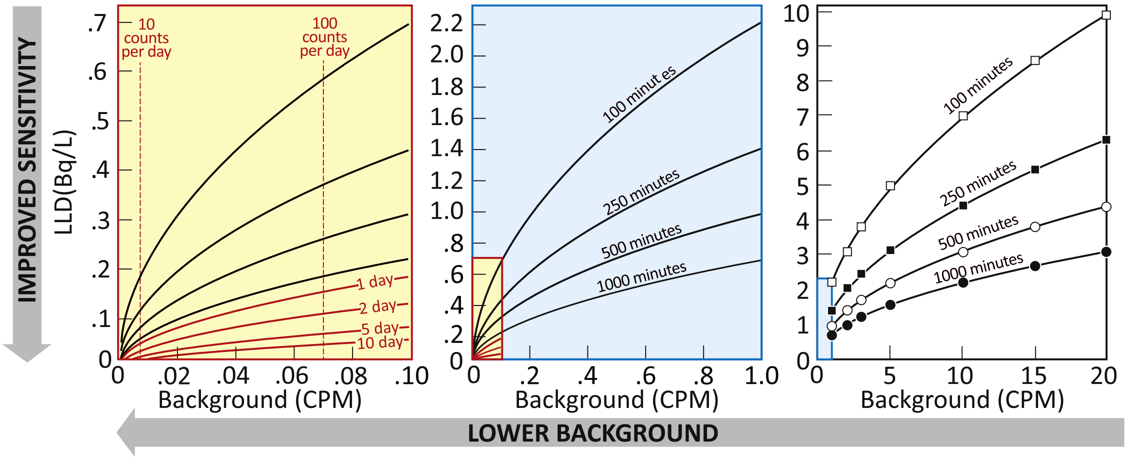

Fig. 1. A “text book” example of the relation between *sensitivity* versus *background & count duration* with extrapolations to a shallow underground, low background liquid scintillation counter design. The “text book” example (right most panel) presents typical sensitivity levels for measurements of a 10 mL tritium sample given typical background rates and count duration and with 35% counting efficiency (LAnnunziata, 2012a). The panels to the left progressively reduce the background rate in the proposed instrument and show the relevant sensitivity levels attained for differing count duration. The count duration curves derived from the Currie formulation (Eq. (1)) are provided to guide the eye and may be impractical for some choices of background and count duration. In these figures the background rate is quantified in counts per minute (CPM) and the minimum detectable activity sensitivity is presented as a lower limit of detection (LLD) value.

primarily to the high detection efficiency achievable through the use of a scintillation cocktail detection medium. Measurement of tritium, ^{3}H, is useful for bench-marking a system's performance. Typical commercial low-background liquid scintillation counters can achieve detection limits on the order of 1 Bq of ^{3}H per liter in the presence of ~1 count per minute (cpm) backgrounds for 1000 minute (min) counting times. For the present study, consider the extrapolation of the decrease in lower limit of detection as a function of decreased background count rate, as shown in Fig. 1 for a series of counting times. The ^{3}H counting efficiency in the extrapolated example is 35% for a constant sample size of 10 mL (LAnnunziata, 2012a). A combination of decreasing the background to the order of 10–100 counts per day and increasing the counting times to days results in a lower limit of detection more than an order of magnitude lower than typically achieved in commercial instruments.

The design presented in this report aims to reduce the background to a range from 0.01 to 0.07 cpm. In the case of tritium (see Fig. 1) this results in detection limits between 0.05 and 0.2 Bq/L depending on the number of days a sample is counted. These estimates employ Currie's equation for minimum detectable activity (MDA), given in Equation (1) (Currie, 1968), where C_b is the background count rate (cpm), T_b is the background count time (min), ε is the counting efficiency, V_s is the sample weight (grams), and T_s is the sample count time (min).

$$\mathrm{MDA} \equiv L_D = \frac{2.71 + 4.65\sqrt{C_b T_b}}{\varepsilon \times V_s \times T_s \times 60} \tag{1}$$

The factor of 60 is used to express the MDA in terms of Bq/g. When the measurement is quantified for liquid volumes, the sample weight is replaced by a volume (milliliters, mL) resulting in a MDA reported in units of Bq/mL. The constant 2.71 is the frequently used value to account for a zero blank case corresponding to a 5% probability of false negatives and 4.65 accounts for a 5% probability of making Type I or Type II errors. The Currie equation provides a consistent way of quantitatively evaluating measurements and is valuable for revealing the potential value of a custom low background LSC system. Foreshadowing the background estimates described in Section 3, the ultra-low-background liquid scintillation counter (ULB LSC) under development is expected to lower detection limits by more than a factor of 10 in comparison to commercial systems located in surface laboratories. In some cases, the result of the background reduction is used to directly improve the MDA of the measurement. In other cases, the 10–100 reduction factor in background rate is used as a trade-off with other factors in the Currie equation (such as sample mass or volume) to reach the same target MDA for a given measurement. For example, an improved MDA may allow greater reach of age-dating in a geochronology scenario while a fixed MDA for expected levels of tritium in water implies reduced sample size.

A companion paper submitted to the J. Radioanal. Nucl. Chem. (Douglas et al., submitted for publication) evaluates in detail a number of specific cases. A summarized selection of three examples from that analysis is presented here to demonstrate the potential use of a ULB LSC system:

(#**1**) A campaign of measurements of tritium backgrounds in natural spring waters was conducted prior to construction of a repository for radioactive waste (Vaupotic et al., 2011). A total of 124 springs were sampled and each sample was electrolytically enriched in tritium (20 × enrichment). Liquid scintillation counting was employed to measure the resulting tritium concentrations using a LSC system having 20–23% detection efficiency and a background rate of 1–2 cpm, resulting in an MDA of 0.3 Bq/L. The electrolytic enrichment step required 139 h/sample and each sample was counted seven times for 30 min. Together this is approximately 6 days of total process time per sample. In the case of a low background LSC system with a background rate reduced to 0.01 cpm, the spring water samples could be counted without the electrolytic enrichment step achieving the same MDA in 195 min of LSC counting. Including the seven-fold replicate counting, the total process time would remain less than 1 day.

(#**2**) A method for determination of ^{90}Sr levels in food samples is comprised of the combustion, dissolution, precipitation, extraction chromatography, ion exchange chromatography, and final precipitation of a 140 g food sample (Heilgeist, 2000). This complex process requires two days to convert the food sample into an LSC sample for analysis with a 1220 Quantulus™ low-background LSC. An MDA of 0.1 Bq/kg was achieved with 100 min count times. Lowering the background to 0.1 or 0.01 cpm would reduce the sample size to 50 or 22 g, respectively, for an equivalent MDA, potentially reducing the scale of the complex sample preparation process.

(#**3**) As part of the ^{235}U decay chain, ^{227}Ac is present in both the water column and ocean sediment. The parent ^{231}Pa is