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Calibration of a liquid scintillation counter to assess tritium levels in various samples

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Abstract

An LKB-Wallac 1217 Liquid Scintillation Counter (LSC) was calibrated with a newly adopted cocktail. The LSC was then used to measure tritium levels in various samples to assess the compliance of tritium levels with the recommended international levels. The counter was calibrated to measure both biological and operational samples for personnel and for an accelerator facility at KFUPM. The biological samples include the bioassay (urine), saliva, and nasal tests. The operational samples of the light ion linear accelerator include target cooling water, organic oil, fomblin oil, and smear samples. Sets of standards, which simulate various samples, were fabricated using traceable certified tritium standards. The efficiency of the counter was obtained for each sample. The typical range of the efficiencies varied from 33% for smear samples down to 1.5% for organic oil samples. A quenching curve for each sample is presented. The minimum detectable activity for each sample was established. Typical tritium levels in biological and operational values are presented. All measured values are far below the recommended international limits. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Liquid scintillation counter; Calibration; Tritium; Bioassay; Smear; Oil

1. Introduction

This study was performed at the Center for Applied Physical Sciences (CAPS), a specialized laboratory at King Fahd University of Petroleum and Minerals (KFUPM). CAPS operates two user facilities for research in areas of nuclear physics and material science using ion beams. The research facilities include a 350 kV high current–light ion accelerator, and a 3.0 MV tandetron accelerator [1].

The zero degree beam line of the 350 kV accelerator is used for elemental analysis using a 14 MeV fast neutron activation analysis technique [2]. The fast neutrons are produced via $T(d, n)$ reactions. The tritium targets consist of tritiated atoms absorbed in titanium layers that are evaporated at 0.25 mm thick copper backing. A continuous flow of a 5 mm thick water layer cools the copper backing. The water flow rate of 5 l/min keeps the target temperature below 25°C.

The handling of tritium involves personnel who may be exposed to contamination, and possible internal intake of tritium. Therefore, the assessment of the tritium levels in the laboratory requires various biological and operational sample analyses

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such as bioassay, smear, oil, environmental, and target cooling water samples. These assessments are evaluated by using the available Liquid Scintillation Counter (LSC). CAPS is equipped with an LKB-Wallac 1217 Rack Beta LSC (LSC) [3], which has a microcomputer that controls four windows for the purpose of counting beta emissions over a wide energy range (from 1 to 2800 keV). The counter includes an automatic window setting for individual samples and batches of samples, a heterogeneity indicator, full interrupt possibilities and multi-user operation. Automatic quench calibration and subsequent calculation of the activity, in disintegration per minute (DPM), values may be obtained by using either the spline function or linear interpolation.

Proper assessment of tritium levels in various samples requires proper calibration of the LSC, by taking into account appropriate sample collection, preparation method, and the counting efficiency. The counting efficiency of the LSC, which varies from sample to sample, depends on the chemical composition, color and original matrix of the sample. Three quench correction methods are commonly used, namely internal standardization, sample channels ratio (SCR), and the external standard ratio (ESR) [4]. Current LSCs are equipped with radioactive sources such as ^{137}Cs for quench corrections. The quenching greatly depends on the color of the sample, and the cocktail being used.

Meehan [5] investigated the color quenching for three different types of cocktails using two different counters. Other types of cocktails were investigated by DeVol et al. [6], in which it was concluded that OptiPhase HiSafe-3 is slightly less quench sensitive than other liquid scintillation cocktails. Additionally, the latter cocktail was found suitable for high-quench samples, and has a high efficiency. In this work, OptiPhase HiSafe-3 cocktail was used to calibrate the LSC. Then the LSC was used to measure tritium levels in various samples in order to assess the compliance with the international recommended levels. Also, the tests were conducted to release items as radiologically clean, to monitor for tritium contamination, and to protect workers from airborne tritium.

2. Methods and materials

2.1. Urine samples

Standard procedure is used to evaluate the HTO intakes by personnel working with tritium. Urine samples were received from personnel, studied, and analyzed on a monthly basis, and according to the Radiation Safety Officer's (RSO) judgment. The counting protocol, suggested by the American National Standards (ANS) is usually followed, and applied here [7]. Under this method, a set of five standards is prepared using 10 ml of cocktail, 1 ml of distilled water, and a traceable certified tritium standard with varying amounts of color quenching agent to cover a wide range of urine color using 20 ml low potassium liquid scintillation glass vials. The calibration standards are certified (by the National Bureau of Standards, U.S.A. with an estimated accuracy of $\pm 1.0\%$). The counting mode of the LSC is adjusted to measure a single label tritium using ESR. Therefore, the counting window is set at 8–110, which covers the beta spectrum energy. Windows three and four, which are assigned for ESR measurement, are set to cover channels 100–135, and 135–185, respectively. The output of the counter is adjusted to give the desired information about a given sample, such as its position, counting time, count rate, quenching parameters, ESR, activity in disintegration per minute (DPM), or in disintegration per second (DPS) and the chemiluminescence percent. A prepared set of standards is used to calibrate the machine in the DPM mode. The efficiency, and the ESR for each standard, are obtained, and a calibration curve is constructed as shown in the results.

2.2. Oil samples

Two types of oil are commonly used in the accelerator, namely, the silicon based oil (fomblin oil) and the typical organic oil. Fomblin oil with a low tritium absorption is used with all roughing pumps, while organic oil is used with all turbo-molecular pumps. The air inside the beam lines is pumped out first via the roughing pumps, and then by turbo pumps. Therefore, any tritium atom produced as a result of degassing from the target will be

removed by the pumps and thus mixed with the oil. As the oil gets used, it loses its viscosity, therefore, it should be changed frequently.

In this study, the contaminated oil which might be a source of tritium hazard was measured and proper waste disposal method was practiced depending on the level of contamination. Detection of tritium in oil by using LSC requires a special calibration for many reasons, such as sample degradation of the scintillation cocktail, high quenching contribution of the oil sample, color change, and the chemiluminescence phenomenon.

A window was calibrated specifically for measuring tritium in organic oil. Additionally, another set of four oil standards was prepared using 1 ml of fresh oil, 10 ml of cocktail and traceable certified tritium standards with varying amounts of quenching agents to assimilate the color range in oil samples. The LSC was calibrated by using the prepared standard set. A calibration curve showing the function of efficiency versus ESR was established as shown in the results.

2.3. Water samples

Water samples are measured to assess the contamination in a target water cooling system and to monitor against airborne tritium (Derived Air Concentration (DAC)). As a result of a D–T reaction, localized heat is generated at the back of the T–Ti target; therefore, it is essential to use water or freon as a coolant. The accelerator's cooling system at the CAPS uses de-ionized water as a coolant at a rate of 5 l/min with a total volume of 25 l. The water gets highly contaminated as a result of flushing the back of the tritium target. Therefore, a periodical check of the water system is applied to observe the contamination level, so that the limit is not exceeded, and the time for replacing the water may be predicted beforehand.

As a requirement, to establish proper personnel protective measures against airborne tritium, room air is bubbled through water or condensed using a cold trap. The water is then measured and the DAC is calculated based on the air flow-rate through the water.

Condensed moisture was collected and analyzed at least twice a week, and after an installation

process of a tritium target. The collected water condensate from air depends on the temperature, pressure, and relative humidity.

For this study, a window was calibrated for measuring tritium in water. A set of three water standards was prepared using 1 ml of distilled water and 10 ml of cocktail, and a certified tritium standard capsule. The corresponding typical efficiency and the ESR values of the three standards were obtained as shown in the results.

2.4. Smear samples

Surface contamination should be monitored regularly, such as bench tops, external glove box walls, floors, and equipment. To assess surface contamination, swipe samples of a 100 cm² area using a filter paper (5.5 cm diameter, Whatman micro-fibre filter, model GF/A) are employed. The amount transferred to the swipe filter represents the removable activity available. The recommended [8] removable efficiency for most swipes is 10% and is dependent on such factors as the surface characteristics, swipe material, and applied pressure. The filter swipe is immersed in a 20 ml cocktail, and the contamination activity is counted using a calibrated LSC.

For this study, a special window was calibrated for this purpose. The channel was calibrated using a clean filter paper with a certified tritium standard, and counted for several times at different filter-to-cocktail configurations.

The minimum detectable activity (MDA) recommended by the American National Standards [7] and implemented by Peterman [9] was estimated by using the following equation:

$$MDA = \frac{4.65\sqrt{N} + 3}{\varepsilon \times T} \quad (1)$$

where N is the background rate in counts per minutes (cpm), T the counting time in seconds, and ε the counting efficiency in count s⁻¹ Bq⁻¹.

According to the criteria of the American National Standards for bioassay tests, the MDA of the counting system should be less than 10⁴ Bq l⁻¹. To account for background corrections, a number of

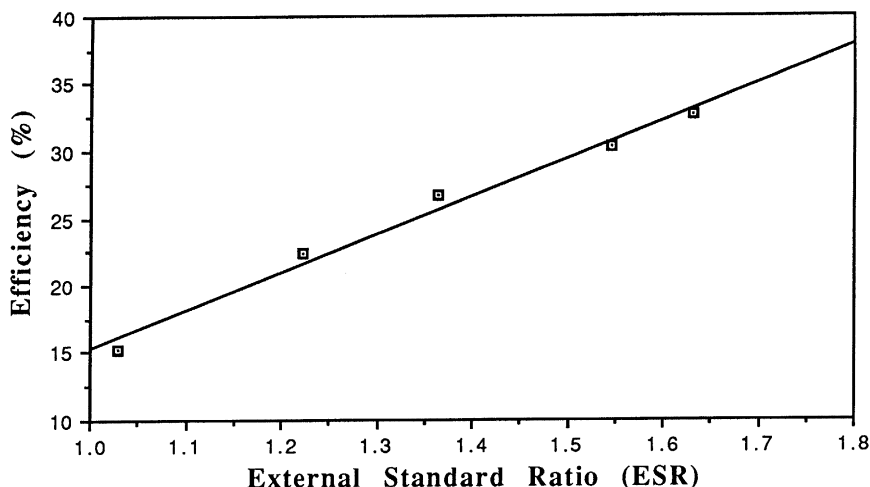


Fig. 1. Calibration of the quenching curve showing the efficiency versus ESR for bioassay samples.

match blanks were fabricated, which is usually used before counting any sample.

3. Results and discussion

3.1. Bioassay samples

Fig. 1 shows the quenching calibration curve for bioassay samples, where a function of efficiency versus ESR was plotted. The range of the efficiency varies from 16% for a high quenching sample up to 32% for a low quenching sample, while the ESR ranges from 1.02 up to 1.65, respectively. The equation of efficiency as a function of ESR is derived using a first order linear regression line to fit the data, with the coefficient of determination (R^2) of 0.98 as follows:

$$\text{Efficiency}(\%) = 28.24 \times \text{ESR} - 12.94 \quad (1 < \text{ESR} < 1.65). \quad (2)$$

Using the calibrated setup, the minimum detectable activity for 5 min counting was calculated using Eq. (1), and was found to be $219 \pm 15 \text{ Bq l}^{-1}$, which is far less than (10^4 Bq l^{-1}), the criteria adopted by the ANS for permissible levels [7]. Personnel handling tritium targets at CAPS were monitored monthly and 3 h after the installation of a tritium

target. Typical tritium concentration of urine samples varied from less than the MDA up to $300 \text{ Bq l}^{-1} \pm 17$. This is far below $1.79 \times 10^7 \text{ Bq l}^{-1}$, which is derived from the annual limit of intake (ALI = $1.2 \times 10^9 \text{ Bq}$) for a normal human being (70 kg) with 67% fluid.

3.2. Oil samples

By using the standard preparation procedure for oil, discussed earlier, a calibration curve for oil as shown in Fig. 2 is obtained, which gives the quenching calibration curve for oil samples of the efficiency function versus ESR. The range of the efficiency varies from 2.0% for low quenching, up to 0.37% for a high quenching sample, while the ESR ranges from 0.05 to 0.35, respectively. This efficiency is very low as compared to bioassay and water samples. This is because of sample degradation in the scintillation cocktail. The equation of the efficiency versus ESR is derived using a first order polynomial fit to the data, with the value of the coefficient of determination (R^2) of 0.99 as follows:

$$\text{Efficiency}(\%) = 4.65 \times \text{ESR} + 0.36 \quad (0.05 < \text{ESR} < 0.35). \quad (3)$$

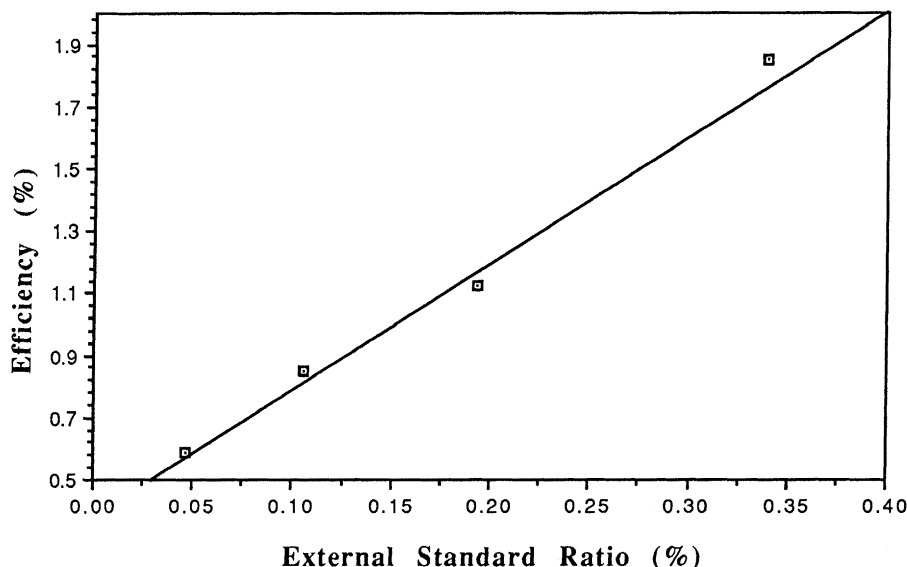


Fig. 2. Calibration of the quenching curve showing the efficiency versus ESR for organic oil samples.

The output of the counter was set to give the chemiluminescence percent, which is a common phenomenon in oil samples. For high chemiluminescence samples, proper assessment of the concentration could be obtained by measuring the activity of the sample after the decay of such an effect, or by increasing the lower energy side of the counting window. Typical concentrations of tritium in organic oil samples which were collected from turbo molecular pumps ranged between 4.23 and 10.68 Bq ml⁻¹ depending on the position of the turbo pump with respect to the tritium target.

3.3. Smear samples

Fig. 3 shows the efficiency of the smear sample versus ESR with respect to other samples. The typical efficiency was found to be approximately 33% with a corresponding ESR of 1.88. This efficiency is due to the low quenching factor, and the proper selection of filter paper, which becomes transparent as it is doused in the cocktail. Contamination levels of various surfaces within the laboratories range from 1×10^{-3} to 4×10^{-3} Bq cm⁻²,

which is less than the (4 Bq cm⁻²) removable surface contamination limit, derived surface contamination level of 30 MBq m⁻² recommended by ICRP 57.

3.4. Water samples

Due to the similarity in the nature of the water standards, the efficiency of all three standards were about the same ($\sim 32\%$), and the ESR values were also about the same (~ 1.63). This matches with the unquenched bioassay samples. Fig. 3 shows the efficiency versus ESR for water, with respect to the values for bioassay, oil, and smear samples. As a result of monitoring, the maximum tritium concentration in the tritium cooling system, for a 3 Ci target, with a total operation load of 9.5 mA h (approximately twice the half life of the tritium target, which is about 2.7 mA h), was found to be 40 Bq ml⁻¹. This is less than 76.1 Bq ml⁻¹, the limit recommended by the IAEA safety series-115 [10]. Thus, while operating these facilities, water is replaced whenever a new target is installed.

Due to the need of continuous monitoring of the airborne tritium, the bubbler or a cold trap method

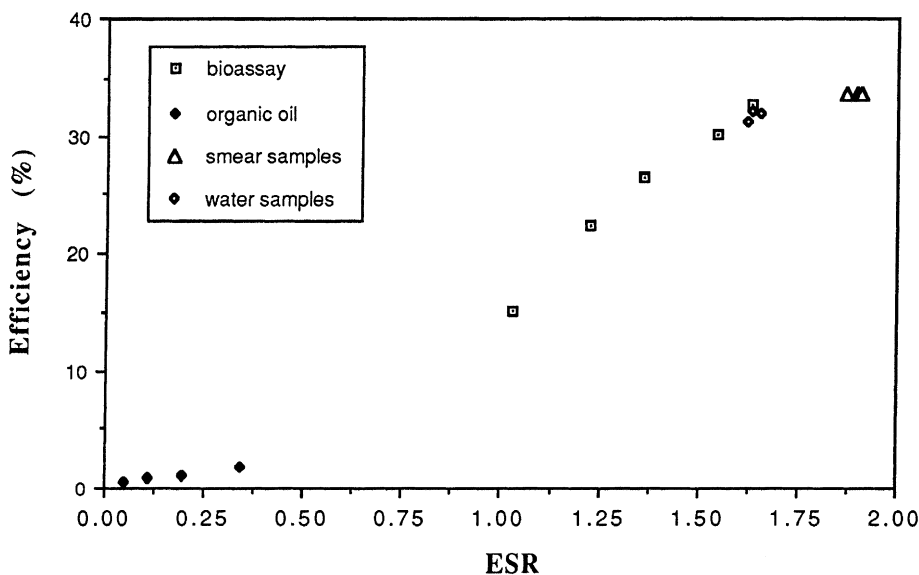


Fig. 3. Overall calibration curve showing the efficiency versus ESR for various samples.

is employed. The collected water sample is analyzed by a calibrated window. The results are converted from Bq to Bq m^{-3} depending on the air flow rate passing through the water.

Typical values of tritium concentration in air varies from 5 to 15 Bq m^{-3} , and these are lower than ($2.3 \times 10^5 \text{ Bq m}^{-3}$) the allowable limit of tritium concentration in air.

4. Summary

The calibration of a Liquid Scintillation Counter for measurements of tritium levels in various biological and operational samples such as bioassay, water, organic oil, and smear samples are described. Typical efficiencies and quenching curves for each type of sample are presented. The minimum detectable activity for each type of sample is established. Typical operational and biological tritium concentration values are presented. None of the measured samples have exceeded the recommended international limits.

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