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Standardization of tritiated water by two improved methods

D. Stanga[a,*], I. Moreau[b], P. Cassette[b]

[a]National Institute of R&D for Physics and Nuclear Engineering "Horia Hulubei" IFIN-HH, P.O.Box MG-6, Bucharest-Magurele, R-077125, Romania
[b]LNE-Laboratoire National Henri Becquerel (LNHB), CEA-Saclay F-91193, Gif –Sur –Yvette CEDEX, France

Abstract

Tritiated water has been standardized in the framework of a French–Romanian cooperation by two improved methods: liquid scintillation counting based on the triple to double coincidence ratio method and the internal gas proportional counting used in conjunction with a tritium generator for chemical reduction of water. The uncertainties of measurement for both methods were smaller than 0.6% and the two results were consistent within these uncertainties, indicating that either method is equally suited for standardizing tritiated water.

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1. Introduction

Tritiated water is a good tracer of nuclear activities in the environment. Its standardization is nevertheless difficult as it decays though pure beta transition with the emission of low-energy electrons (Makepeace et al., 1998). In the framework of a French–Romanian cooperation, tritiated water has been standardized by two improved methods: liquid scintillation counting (LSC) based on the triple to double coincidence ratio (TDCR) method (Broda et al., 1988; Cassette and Vatin, 1992) and internal gas proportional counting (IGPC) used in conjunction with a tritium generator (TG) for chemical reduction of water (Makepeace et al., 1996; Unterweger et al., 1980).

For the application of the IGPC, an improved method of measurement based on the equality between molar activities of the tritiated water and the tritiated hydrogen has been used. For the application of the TDCR method, the anisotropy of the counter was taken into account and the efficiency variation method was used to determine the optimal parameters for the calculation model. Measurement results are presented and discussed in the paper. These results indicate that either method is equally suited for standardizing tritiated water.

2. Description of the methods

2.1. TDCR method

The TDCR method previously described (Broda et al., 1988 ; Grau Malonda and Coursey, 1988) was used for standardizing tritiated water. The method requires the use of a triple coincidence liquid scintillation counter. The LNHB counter uses BURLE 8850 photomultiplier tubes and the discriminator threshold is adjusted under the single photoelectron peak (Cassette and Vatin, 1992). The detection efficiency is calculated using the TDCR07 program that takes into account the different quantum efficiency of photomultiplier tubes by solving a set of three TDCR equations:

$$\frac{T_{abc}}{D_{ab}} = \frac{\sum_{spectrum} S(E)(1-\mathrm{e}^{-\eta_a})(1-\mathrm{e}^{-\eta_b})(1-\mathrm{e}^{-\eta_c})}{\sum_{spectrum} S(E)(1-\mathrm{e}^{-\eta_a})(1-\mathrm{e}^{-\eta_b})}, \tag{1}$$

$$\frac{T_{abc}}{D_{bc}} = \frac{\sum_{spectrum} S(E)(1-\mathrm{e}^{-\eta_a})(1-\mathrm{e}^{-\eta_b})(1-\mathrm{e}^{-\eta_c})}{\sum_{spectrum} S(E)(1-\mathrm{e}^{-\eta_b})(1-\mathrm{e}^{-\eta_c})}, \tag{2}$$

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*Corresponding author. Tel.: +40 1 780 7040; fax: +40 21 4231701.
E-mail addresses: doru@ifin.nipne.ro, stanga_doru@yahoo.com (D. Stanga).

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$$\frac{T_{abc}}{D_{ac}} = \frac{\sum_{spectrum} S(E)(1-e^{-\eta_a})(1-e^{-\eta_b})(1-e^{-\eta_c})}{\sum_{spectrum} S(E)(1-e^{-\eta_a})(1-e^{-\eta_c})}, \tag{3}$$

where T_{abc} is the experimental triple coincidence counting rate, D_{ab}, D_{bc} and D_{ac} are experimental double coincidence counting rates and $S(E)$ is the tritium spectrum discretized over 100 channels. The set of three equations was solved by using the downhill simplex algorithm (Press et al., 2001) and Birks formula

$$\eta_i = \frac{v_i}{3}\int_0^E \frac{A\,\mathrm{d}E}{1+kB(\frac{\mathrm{d}E}{\mathrm{d}x})}, \quad i \equiv a,b,c, \tag{4}$$

where $\mathrm{d}E/\mathrm{d}x$ is the electron stopping power and v_i ($i \equiv a,b,c$) are the respective quantum efficiencies of the photomultiplier tubes a, b and c.

For a low-energy beta emitting nuclide like ^{3}H, the calculation of the detection efficiency is very dependant on the results from the ionization quenching formula and thus the best models and parameters must be used to calculate this formula. The calculation of the electron stopping power was made using the ICRU 37 formula (ICRU, 1984) with a linear extrapolation to zero under 100 eV.The optimum value of the kB coefficient was chosen by instrument defocusing where the detection efficiency is decreased by decreasing the photomultiplier tubes focusing electrode voltage and the calculated activity (i.e. the counting rate divided by the detection efficiency) is plotted against the efficiency. For each value of the kB parameter, in the 0.007–0.015 cm/MeV range, a straight line is fitted to the activity values and the best estimate of the kB parameter corresponds to the minimum slope of this fit. In other words, the best kB value is the value giving a radioactive solution activity independent of the detection efficiency.

2.2. IGPC method

The equipment used for the tritiated water standardization by internal gas proportional counting consists of a TG for the tritiated water reduction and a counting system with length-compensated proportional detectors for tritiated hydrogen measurements. The IGPC system was previously described together with the method of measurement (Makepeace et al., 1994; Lansiart et al., 1993; Stanga et al., 2002). Using this method, the activity per unit of volume of the radioactive gas samples at STP conditions ($T_0 = 273.15$ K, $p_0 = 1013.25 \times 10^2$ Pa) Λ_V^0 can be calculated with the following equation:

$$\Lambda_V^0 = f\frac{V_T}{V_D.V_S^0}R_D, \tag{5}$$

where f is a correction factor for R_D, V_T ($V_T = (4685.9 \pm 6)\,\mathrm{cm}^3$) is the total volume of the IGPC system, V_D ($V_D = (105.85 \pm 0.21)\,\mathrm{cm}^3$) is the difference between sensitive volume of the long and short detectors, R_D is the difference in count rate (corrected for dead time and background) between the long and short counters and V_S^0 represents the volume of the radioactive gas sample in STP conditions.

The conventional method for the tritiated water standardization by IGPC is based on the fact that the total activity of the tritiated water sample is wholly transferred to tritiated hydrogen when the reduction yield is unity. Briefly, it can be described as follows. A tritiated water sample of mass m is chemically reduced to tritiated hydrogen, which is collected in a flask and then is transferred to a volume V where its pressure p and temperature T are measured. A sample of gas is taken from V and its activity per unit volume, Λ_V^0, at STP conditions is measured by IGPC method. The activity per unit of mass, Λ_m , of the tritiated water sample is given by

$$\Lambda_m = \frac{\Lambda_T^{(th)}}{m} = \Lambda_V^0 \frac{V.p.T_0}{m.p_0.T}, \tag{6}$$

where $\Lambda_T^{(th)}$ is the total activity of the tritiated hydrogen.

In this work, an improved method has been used (Stanga and Cassette, 2006). It is based on the equality between the molar activities of the tritiated water and the tritiated hydrogen obtained after the reduction process. This equality can be explained theoretically by taking into account the chemical equations for the tritiated water reduction, which show that one mole of water gives always one mole of hydrogen. Due to isotopic effects (difference in vapor pressure of H_2O and T_2O), the equality is valid only when the reduction yield is unity. Consequently, a tritium generator that always gives a unitary chemical reduction yield and is able to detect very small quantities (~10 μg) of evenly unreduced water has been used (Stanga et al., 2005). Also, the chemical reduction has been achieved at a quite low temperature (450 °C).

Using the equality between molar activities, it can be shown that the activity per unit of mass, Λ_m, of the tritiated water is given by (Stanga and Cassette, 2006)

$$\Lambda_m = \frac{k_m\Lambda_V^0}{1+\Lambda_V^0(2k_m/\lambda_t N_A)}, \tag{7}$$

where N_A is the Avogadro constant, λ_t is the decay constant for tritium and k_m is given by

$$k_m = \frac{RT_0}{p_0\mu_{H_2O}} = (1244.1657 \pm 0.0207)\,\mathrm{cm}^3/\mathrm{g}, \tag{8}$$

with R representing the gas constant and μ_{H_2O} the molar mass of water. In all tritiated water measurements, Eq. (7) was approximated by

$$\Lambda_m = k_m\Lambda_V^0 \tag{9}$$

because $(2k_m/\lambda_t N_A) = 2.32 \times 10^{-12}\,\mathrm{cm}^3/\mathrm{Bq}$ and Λ_V^0 was smaller than 0.1 kBq/cm^3.

Eq. (7) establishes a very simple and direct relationship between Λ_m and Λ_V^0 providing the metrological traceability for tritium gas and tritiated water measurements.

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