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COMPARISON OF TRIPLE TO DOUBLE COINCIDENCE RATIO AND QUENCH PARAMETER EXTERNAL METHODS FOR THE DETERMINATION OF ^{3}H EFFICIENCY BY LIQUID SCINTILLATION COUNTING

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ABSTRACT

The aim of this study is to determine the tritium efficiency by liquid scintillation counting using two methodologies, Quench Parameter External (QPE) and Triple to Double Coincidence Ratio (TDCR), and to compare the results**.** The equipment used was the HIDEX model 300-SL Liquid Scintillation Counter, composed of three photomultipliers coupled with coincidence pulses, discrimination level and MikroWin 2000 software. The efficiency varied from 0.028 to 0.706 cps dps^{-1} for QPE and from 0.061 to 0.703 cps dps^{-1} for TDCR. Different efficiencies were obtained using both methods, in the range from 459 to 572 quenching, above this range the efficiencies were similar. The verification of the efficiencies was performed by participating in the Intercomparison National Program (PNI).

1. INTRODUCTION

The liquid scintillation counting (LSC) is a technique in which the sample is mixed to the liquid scintillation, forming a scintillation solution, capable of converting the kinetic energy of nuclear emissions into light photons. The interaction of the emissions occurs in the liquid solution, producing excitement with emission of photons of ultraviolet radiation. Quench is a reduction in system efficiency, as a result of energy loss in the liquid scintillation solution. Because of quench, the energy spectrum detected from the radionuclide appears to shift toward a lower energy. The three major types of quench are photon, chemical, and optical quench [1,2]. The counting efficiency was determined by the relative quenching of the sample, by using an external source for the determination of quench indicating parameter [3]. Quench Parameter External (QPE) is the most common method for determination of efficiency in LSC, used with external standard generating the parameter of indication of quench [4]. Triple to Double Coincidence Ratio (TDCR) is a primary measurement method based on calculation of the efficiency from the measured ratio of double and triple coincidence counting rates. TDCR does not require radioactive source and quenching curve, therefore it is an absolute measurement [5]. The quenching factor is more critical for the determination of tritium. The tritium is a pure beta emitter, with 18.6 keV energy and half-life of 12.32 years [6], produced naturally in the atmosphere and as a byproduct in nuclear reactors and explosions [7]. This paper describes the determination of the efficiency for tritium using two methodologies, QPE and TDCR**.**

2. MATERIALS AND METHODS

The efficiency for tritium was measured with a LSC, HIDEX model 300-SL, composed by three photomultipliers coupled with coincidence pulses, discrimination level and MikroWin 2000 software. The counts were determined using three different Quenched Standards Sets, ^{3}H NES Quenched Standard Set (NES), ^{3}H UG Quenched Standard Set (UG) and ^{3}H XR Quenched Standard Set (XR), and a reference standard without quench (Unquenched), provided by PerkinElmer. For QPE methodology, an external source of ^{152}Eu was used for producing a parameter of indication of quench. The counting efficiency was obtained by the equation (1).

$$Ef_{\_QPE} = \left(\frac{counts - Bg}{Activity} \right) \tag{1}$$

where
$Ef_{\_QPE}$: counting efficiency (cps dps^{-1})
Activity: source activity (Bq)
counts: source counts (cps)
Bg: background radiation (cps)

For the TDCR methodology, the calculation took into account the differences between the counting ratios on the photomultipliers, based on the efficiency from the measured ratio of double and triple coincidence counting rates, by the expression (2) [8].

$$TDCR = \frac{\int_{0}^{E_{\max}} S(E)(1-e^{-n})^{3}\,dE}{\int_{0}^{E_{\max}} S(E)((3(1-e^{-n})^{2} - 2(1-e^{n})^{3}))\,dE} \tag{2}$$

Where
S(E)(1 - e^{-n})^{3}: triple coincidences
S(E)((3((1 - e^{-n})^{2} - 2(1 - e^{-n})^{3} e^{-n}: all coincidences

The counting time was 20 minutes for each sample. The samples were analyzed in triplicate. The background radiation was determined by using the same scintillating vial with deionized water.

3. RESULTS AND DISCUSSION

For the two methodologies, 24 tritium quench cocktail standards with different quenching agents, with activity of 2567 Bq for NES, 4218 Bq for UG, 4280 Bq for XR and Unquenched with activity 4083 Bq, were used. The efficiency results of QPE and TDCR are presented in Table 1. In the quench indication range from 459 to 572 the efficiencies varied significantly, whereas in the range from 631 to 844 the results were similar.

Table 1: Counting efficiency (cps dps^{-1}) by using the QPE and TDCR methodologies for Tritium determination

^{3}H Quenched Standard Set	Quench indication	QPE Efficiency (cps dps^{-1})	TDCR Efficiency (cps dps^{-1})	QPE/TDCR
UG	459	0.028 ± 0.001	0.061 ± 0.001	0.46
UG	525	0.073 ± 0.001	0.105 ± 0.001	0.70
NES	561	0.120 ± 0.001	0.155 ± 0.001	0.77
UG	572	0.123 ± 0.001	0.148 ± 0.001	0.83
UG	631	0.220 ± 0.001	0.225 ± 0.001	0.98
XR	645	0.279 ± 0.001	0.269 ± 0.001	1.04
NES	666	0.313 ± 0.001	0.303 ± 0.001	1.03
UG	669	0.316 ± 0.001	0.303 ± 0.001	1.04
XR	681	0.381 ± 0.001	0.356 ± 0.001	1.07
NES	699	0.397 ± 0.001	0.378 ± 0.002	1.05
UG	701	0.403 ± 0.001	0.380 ± 0.001	1.06
XR	720	0.482 ± 0.002	0.451 ± 0.001	1.07
NES	724	0.456 ± 0.005	0.434 ± 0.002	1.05
UG	728	0.468 ± 0.001	0.441 ± 0.001	1.06
UG	752	0.520 ± 0.001	0.495 ± 0.001	1.05
NES	757	0.523 ± 0.006	0.498 ± 0.003	1.05
XR	759	0.563 ± 0.002	0.536 ± 0.001	1.05
UG	777	0.565 ± 0.001	0.543 ± 0.001	1.04
NES	785	0.573 ± 0.001	0.556 ± 0.003	1.03
XR	794	0.627 ± 0.002	0.609 ± 0.001	1.03
UG	800	0.610 ± 0.001	0.590 ± 0.001	1.03
XR	824	0.671 ± 0.002	0.663 ± 0.001	1.01
XR	839	0.694 ± 0.002	0.687 ± 0.001	1.01
Unquenched	844	0.706 ± 0.001	0.703 ± 0.001	1.00

The Figure 1 presents the relationship between the TDCR efficiency and the QPE efficiency, for ^{3}H standards with different quenching agents, calculated in the present study.

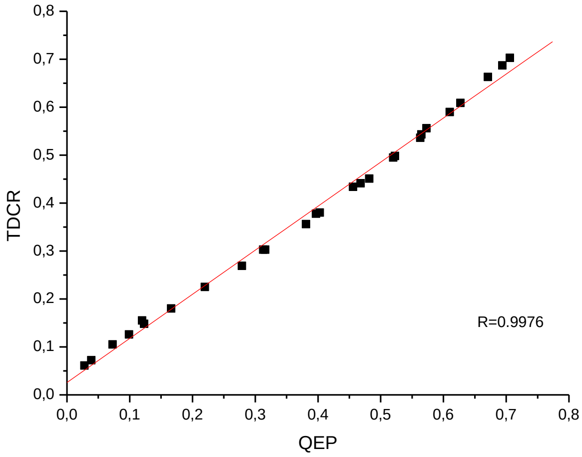

Figure 1. Relationship between the TDCR efficiency and the QPE efficiency for ^{3}H

The adjustment showed a good correlation, of 0.9976, between the TDCR efficiency and the QPE efficiency.

The verification of the TDCR and QPE efficiencies was performed by measuring samples provided by the Brazilian National Intercomparison Program (PNI). Each sample was analyzed in triplicate. The average of the quench indication obtained was 735 ± 1, the mean efficiencies were 0.465 ± 0.002 and 0.467 ± 0.014, for the QPE and TDCR methodologies, respectively. The results are presented in table 2.

Table 2: Concentrations and uncertainties of ^{3}H determination by using QPE and TDCR efficiencies

^{3}H reference value Bq L^{-1}	^{3}H concentration Bq L^{-1} (QPE)	^{3}H concentration Bq L^{-1} (TDCR)	Relative Error % (QPE)	Relative Error % (TDCR)	Relative Standard Deviation (QPE)	Relative Standard Deviation (TDCR)
252±51	247±4	248±3	-2.2	-1.9	1.5	1.1
553±111	584±1	608±8	5.5	9.8	0.1	1.3
202±40	187±16	183±11	-7.6	-9.4	8.4	6.2
450±90	404±1	415±5	-10.2	-7.9	0.1	1.2
141±28	135±2	132±2	-4.3	-6.6	1.2	1.6

In general, relative standard deviations and relative errors were lower than 10%, proving that the two methodologies were satisfactory for the determination of ^{3}H by liquid scintillation technique.

Since the efficiencies varied significantly in the quench indication range from 459 to 572, a new verification was performed by using the standard solution XR, with corrected activity of

3938 ± 63 Bq. The results of the activity in the critical range of the quench are shown in table 3.

Table 3: Concentrations and uncertainties of ^{3}H using QPE and TDCR efficiencies in the Quench Indication critical range

Quench indication	H-3 concentration (Bq) (QPE)	H-3 concentration (Bq) (TDCR)	Relative Error % (QPE)	Relative Error % (TDCR)	Relative Standard Deviation (QPE)	Relative Standard Deviation (TDCR)
580	3742±61	3572±20	-5.0	-9.3	1.6	0.6
528	3668±61	2989±15	-6.9	-24.1	1.7	0.5
463	3654±60	2052±48	-7.2	-47.9	1.7	2.4

The TDCR method gave worse results with the increment of the quench indication. The QPE method was more appropriate in the quench indication range from 463 to 580.

4. CONCLUSIONS

The TDCR method requires equipment with three photomultipliers, whereas the QPE method requires equipment with only two photomultipliers, but needs different quenching standards of the element to be analyzed.

The TDCR method is in general faster and simpler, and does not require radioactive standard and quench curve, since it is an absolute measurement.

For the verification of ^{3}H efficiencies by using the QPE and TDCR methods, samples of PNI were analyzed. The values obtained for the relative standard deviation and relative error were lower than 10%, proving that the precision and accuracy of the liquid scintillation technique are appropriate for the determination of tritium.

The QPE and TDCR methods gave results with a good precision and accuracy in the range from 580 to 844 for the quench indication. In the range from 463 to 580 the accuracy was not satisfactory for the TDCR method. The QPE method was more appropriate for the entire quench indication interval studied.

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