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Standardization of ^{32}P radioactive solution

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ABSTRACT

The standardization solution using three different methods is presented. The disintegration rate was determined by the CIEMAT/NIST and TDCR methods in liquid scintillator systems and self-absorption extrapolation method using 4π(PC)-β system. The results obtained for the activity of the ^{32}P solution were compared and they agree within experimental uncertainties.

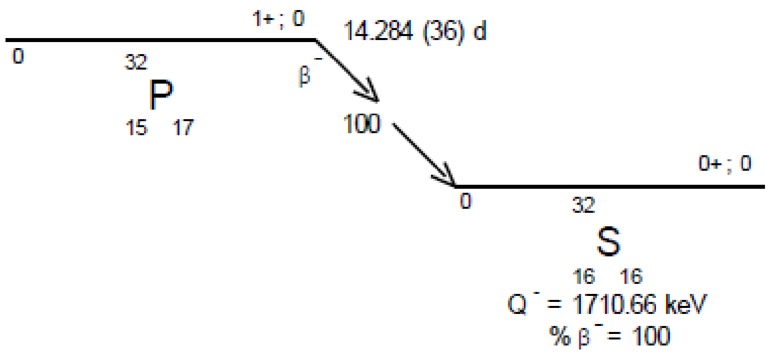
Keywords: standardization, liquid scintillation, phosphorus-32, radioactivity.

1. INTRODUCTION

The Nuclear Metrology Laboratory (LMN) at the IPEN, São Paulo, Brazil, has developed different methods for the standardization of pure beta emitter radionuclides such as ^{90}Y, ^{35}S, ^{14}C [1-3] by means of liquid scintillators systems and 4π(PC)-β coincidence systems in order to establish standard methodologies. In this paper, the procedures applied to standardize a ^{32}P radioactive solution are shown. For this standardization, two liquid scintillator systems and a 4π(PC)-β system were used.

The radionuclide ^{32}P may be produced in nuclear reactors mainly by $^{32}S(n, p)^{32}P$ and $^{31}P(n, \gamma)^{32}P$ reactions. The former reaction gives rise to ^{33}P or ^{35}S impurities and must be checked by analyzing the liquid scintillator beta spectra. One of the most important applications relies on its use in nuclear medicine for treatment of polycythemia vera, leukemia for distinct subgroup of elderly patients and treatments of skin [4]. The ^{32}P has a half-life of 14.284 (36) days and decays by beta particles with endpoint energy of 1710.66 keV to ^{32}S [5]. The ^{32}P decay scheme is shown in figure 1.

Figure 1: *^{32}P decay scheme*



Source: reference [5]

The measurements in Liquid Scintillation Counting (LSC) systems were carried out in two systems. The first one was a TRICARB 2100 Packard system using the CIEMAT/NIST method [6], where a ^{3}H standard solution is used as tracer. This is a standard technique used in most metrology laboratories around the world. The other was HIDEX 300SL system using the TDCR (Triple to Double Coincidence Ratio) method [7].

The disintegration rate was also determined in a 4π(PC)–β system by applying the self-absorption extrapolation method [8]. A Time Amplitude Converter (TAC) method [9] was used to register the beta events detected in the proportional counter.

2. METHODS

2.1. CIEMAT/NIST Method

The CIEMAT/NIST method consists of relating the experimental efficiency of a standard ^{3}H solution, obtained from samples with different quenching, with the theoretical ^{3}H efficiency, which is obtained in function of Factor of Merit (FM). This results in a relationship between quenching and Factor of Merit, which is called universal curve. By means of this curve, it is possible to determine the theoretical efficiency as a function of FM for the radionuclide under study, related to the corresponding experimental quenching. Code CN2001 [10] was used to obtain ^{3}H and ^{32}P theoretical efficiencies.

2.2. TDCR Method

TDCR method [7] uses three photomultipliers tubes (PMT) positioned at relative angle of 120 degrees (A, B and C) and operated in coincidence. Through a special electronic system, the coincidence of the three PMT pairs, which are N_{AB}, N_{AC}, N_{BC}, and N_D and N_T, the logical double and triple coincidence rates are collected. The arithmetic relationships among these rates [7,11] are given by:

$$\boldsymbol{N_{AB} + N_{BC} + N_{AC} = 2N_T + N_D} \quad \textbf{(1)}$$

$$\boldsymbol{N_T = N_{ABC}} \quad \textbf{(2)}$$

where N_{ABC} is the triple coincidence count rates for the three photomultipliers A, B e C; N_{AB} is the double coincidence count rates for the photomultipliers A and B; N_{AC} is the double coincidence count rates for the photomultipliers A and C and N_{BC} is the double coincidence count rates for the photomultipliers B and C. The double and triple counting rates are given by:

$$N_D = N_0 \varepsilon_D \quad (3)$$

$$N_T = N_0 \varepsilon_T \quad (4)$$

where ε_D and ε_T are the double and triple counting efficiencies, respectively, and N_0 is the activity. For a large number of detected events, the ratio of N_T/N_D converges to the ratio $\varepsilon_T/\varepsilon_D$ given by [12]:

$$N_T/N_D = \varepsilon_{TDCR} \quad (5)$$

The HIDEX 300SL system assumes that, for beta pure emitters, the $\varepsilon_{TDCR} = \varepsilon_D$ [12] and the activity is obtained by:

$$N_0 = N_D/\varepsilon_{TDCR} \quad (6)$$

2.3. Self-Absorption Method

The self-absorption method consists of measuring solid sources of the beta pure emitter, prepared in Collodion subtracts, with various aliquots of the solution, in a 4π(PC)–β system. From these measurements, a curve is obtained between N_β counting rate (cps mg^{-1}) *vs.* sources mass (mg) which, extrapolated to mass zero, gives the disintegration rate. A straight line using covariance methodology fits this curve. The intercept of this line yields the activity N_0.

2.4. Experimental Set Up

2.4.1. Tricarb 2100 Liquid Scintillator Counting System

TRICARB Mod 2100 TR is a liquid scintillator counting system which detects the photons emitted from the scintillation vial by means of two photomultipliers tubes operated in coincidence. For this measurement, the CIEMAT/NIST method was applied. Owing to it is necessary determine the quenching indicator parameter that was obtained by means of the tSIE (transformed Spectral Index of External Standard), which is calculated from the Compton spectrum induced in the scintillation cocktail, by an external source of ^{133}Ba, placed near the measurement system [13].

2.4.2. HIDEX 300SL Liquid Scintillator Counting System

HIDEX 300SL is a commercial liquid scintillator counting system that uses the TDCR method. The HIDEX software, associated to the system, provides the TDCR efficiency and the activity in disintegration per minute (dpm). This system does not require samples with different quenching values.

2.4.3. 4π(PC)-β system

The 4π(PC)-β system used was a 4π proportional counter filled with P-10 gas (90% Argon plus 10% methane) operated at 0.1 MPa. The measurements in the proportional counter were performed in the integral mode, using a single channel analyzer (SCA) to discriminate the electronic noise. The events detected in the proportional counter were registered by a method developed by the LMN, which makes use of a Time Amplitude Converter (TAC), associated with a Multichannel Analyzer. In this method, after to be selected in the SCA, the detector pulses are sent to two gates and delay generators that will give the start and stop information to the TAC. The observed counting rate N_β was corrected for background, dead time and decay in the usual way.

2.4.4. Source preparation

The sodium phosphate (^{32}P) solution was obtained from the Radiopharmaceutical Center (CR) at IPEN. The radioactive solution was diluted in distilled water. No impurities were detected by liquid scintillation measurements. The sources, to be measured in the proportional counter, were prepared by dropping aliquots with different masses of the solution on a 20 μg cm^{-2} thick Collodion film. This film has been previously coated with a 10 μg cm^{-2} gold layer, in order to make the film conductive. A seeding agent (CYASTAT SM) was used to improve the deposit uniformity and the sources were dried under a nitrogen jet at 45°C. A total of eight sources were prepared with masses ranging from 11 to 35 mg.

The samples for liquid scintillator were prepared by pouring 15 mL of Ultima Gold scintillator cocktail, by means a calibrated pipette, and 1 mL of distilled water, in polyethylene 20 mL vials. For the present measurements, five samples of ^{3}H and ^{32}P were prepared. After dropping the radioactive solution into the scintillator cocktail, the samples were mixed and stirred to obtain a homogeneous solution. Different amounts of nitro-methane solution were added in five samples in order to obtain different quenching factors and therefore different efficiencies. One extra sample with liquid scintillator plus water (blank) was prepared to measure the background.

The masses of radioactive material determination were performed using the pycnometer technique [14] in a Mettler balance model XP56. Buoyancy correction has been applied to all masses.

3. RESULTS AND DISCUSSION

The self-absorption extrapolation curve obtained with 4π(PC)-β system is presented in figure 2. The fit of a straight line was performed by least squares by means of code LINFIT [15], which incorporates covariance matrix methodology.

Figure 2: *Self-absorption extrapolation curve of ^{32}P 4π(PC)-β system. The black marks correspond to experimental points and the continuous line to the fitting.*

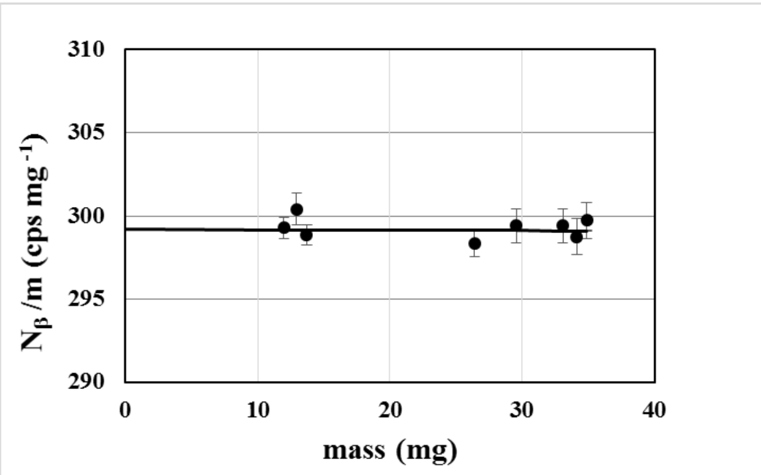


Table 1 presents the sample mass, TDCR efficiencies and ^{32}P activity results with total uncertainties obtained using the HIDEX 300SL liquid scintillator system.

Table 1: Activity results obtained with TDCR method in HIDEX 300SL system.

Sample	Experimental data		
	Mass (mg)	**ε_{TDCR}* (%)**	**Activity* (Bq mg^{-1})**
1	17.026	99.7 (4)	300.8 (16)
2	9.147	99.6 (5)	301.6 (21)
3	11.224	99.5 (5)	301.1 (19)
4	8.119	99.3 (6)	294.8 (22)
5	8.627	99.1 (6)	299.4 (22)

*Absolute uncertainty is given in parentheses

In table 2, the amount of carrier, the quenching parameter (tSIE), ^{32}P efficiencies and the activity obtained by the application of CIEMAT/NIST method are shown.

Table 2: Activity results obtained with CIEMA/NIST method in TRICARB 2100 system.

Sample	Experimental data			
	Nitro (μL)	**Q***	**ε ^{32}P* (%)**	**Activity* (Bq mg^{-1})**
1	10	489 (3)	99.8 (5)	297.3 (56)
2	20	375(1)	99.7 (5)	297.8 (57)
3	30	314 (1)	99.7 (5)	296.9 (50)
4	40	216 (3)	99.6 (5)	289.2 (54)
5	50	160 (3)	99.5 (5)	293.9 (64)

*Absolute uncertainty is given in parentheses

The uncertainties of the three methods are presented in table 3. For the 4π(PC)-β system, the main uncertainties involved were: fitting procedure, counting statistics from N_{β} and beta background, which were considered uncorrelated, and the uncertainties in weighing, dead time, decay correction and resolving time, considered correlated. The uncertainty due to count statistics is included in the extrapolation curve fitting.

For the TDCR method, the uncorrelated uncertainties considered were counting statistics, TDCR efficiency and background; the uncertainties in weighing and decay correction were considered correlated. For the CIEMAT/NIST method, the counting statistic and quenching parameter were considered uncorrelated and the ^{3}H and ^{32}P efficiencies, weighing and decay correction were considered correlated

Table 3: Typical partial uncertainties in the activities for the three methods, in percent (k=1).

Component	**Uncertainty (%)**		
	4π(PC)-β	**TDCR**	**C/N**
Counting statistics	-	0.38	0.60
Weighing ^{32}P	0.10	0.10	0.10
Tracer (^{3}H)	-	-	1.6
Quenching (Q)	-	-	0.54
Efficiency C/N	-	-	0.5
Efficiency TDCR	-	0.54	-
Decay correction	0.21	0.22	0.22
Resolving time	0.05	-	-
Dead time	0.10	-	-
Extrapolation self-absorption curve/ least squares fit error	0.23	-	-
Combined uncertainty	0.35	0.70	1.88

The mean activity obtained for the solution considering the three methods were:

(299.9 ± 1.2) Bq mg^{-1}, with the 4π(PC)-β system, (299.8 ± 1.5) Bq mg^{-1}, with TDCR method and (296.0 ± 4.7) Bq mg^{-1}, with CIEMAT/NIST method, respectively. As can be seen, the final values obtained with the three methods agree within the experimental uncertainties.

4. CONCLUSION

Absolute activity measurements of ^{32}P were carried out by means of three distinct methods and the results obtained from these methods agree within the experimental uncertainties, confirming their feasibility.

However, the TDCR method proved being more convenient for ^{32}P standardization compared with the method of self-absorption due to sample preparation and it is more convenient than CIEMAT/NIST method because it does not require neither the use of a standard solution as tracer, nor the use of quenching agents.

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