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Development of Movable Detection System for Efficiency Measurement in 3-PM Liquid Scintillation Counting

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Abstract

We developed an improved 3-PM liquid scintillation counting (3-PM LSC) method in which three detectors can be displaced to back and forth directions, and a data acquisition system being able to provide the values for all parameters required for the method. The detectors are entirely located in a 20-mm lead chamber of an inner diameter of 500 mm. A saw-toothed gear ties up all detectors so as to move them uniformly, up to 50 mm with unit of 1 mm. The data acquisition system was designed in an integrated circuit to perform the necessary works such as fast amplification, discrimination, coincidence and logic analysis. It generates values of nine parameters among twelve's generated in the 3-PM LSC method. The dead time of each counting channel is of extending type, varying from 10 to 100 μs. We measured the TDCR values with an unquenched liquid scintillation source ^{14}C by displacing the detectors with a step of 2.5 mm away from counting vial. Their values were derived on the range from 0.9 to 0.6. The extent is three times wider than those regions observed by applying the defocalization technique.

Key Words : 3-PM LSC, movable detetion system, defocalization, TDCR, ^{14}C

1. Introduction

The 3-PM LSC method, using three photomultiplier tubes as detectors, has been widely applied in determining the activity of pure beta emitters [1-5]. Since the efficiency parameter is yielded entirely from triple to double coincidences ratio in the method, the activity is obtained using the efficiency-parameter function derived by means of the adequate technique. Thus the method is known as TDCR (Triple to Double Coincidence Ratio) method alternatively. In the conventional technique using two photomultiplier tubes as detectors, it needs to prepare some quenched samples so as to vary the detection efficiency. Thus it is necessary to take into accounts the corrections due to the complicated chemical effects such as chemical quenching, colour quenching etc. [6,7]. Whereas the activity in the new method is obtained directly from the triple and double coincidences ratios, it enables to apply the mechanical techniques pertinent to yield their variations on the wide ranges only with single unquenched liquid scintillation sample.

For this aim, the defocalization method, forces to change the focus voltages loaded between photocathodes and first dynodes of each photomultiplier tube, was developed and has been widely used up to now.

However, the detectors such as photomultiplier tubes are very sensitive even the slight change of applied voltages, and it is undesirable to change the voltage frequently during the experiment. Further since the voltage of the photomultipliers should be maintained above the operating voltage to let them work, the extent of the efficiency-variations is not being capable of reach to wide regions, for example at most 10 % for the case of ^{14}C according to our experiences.

Another candidate that enables to get over those difficulties can be found in the geometry-variation method. Since the geometry of an array of detectors can be displaced mechanically, one can obtain the detection efficiencies on the wide ranges. In addition, the method does not give any external influences to counting instruments. We developed such a 3-PM LSC system in which all the detectors can be displaced close to or away from counting vial at a same time. The advantages of the method are demonstrated by measuring the TDCR values with an unquenched liquid scintillation sample ^{14}C.

2. Experiment

Figure 1 shows the detection system developed for this work. The detection part is entirely located in the cylindrical chamber with an inner diameter of 500 mm, mounted in a 20-mm lead shield. The detectors used in this work were Hamamatsu R1847-07 photomulitiplier tubes, 14 stage fast-linear focussed type with bialkali photocathode of 51 mm diameter. These are symmetrically placed around the centrally located counting vial. A saw-toothed gear ties up all detectors so as to move them uniformly, up to 50 mm with unit of 1 mm by a step-motor. Computer controls all the

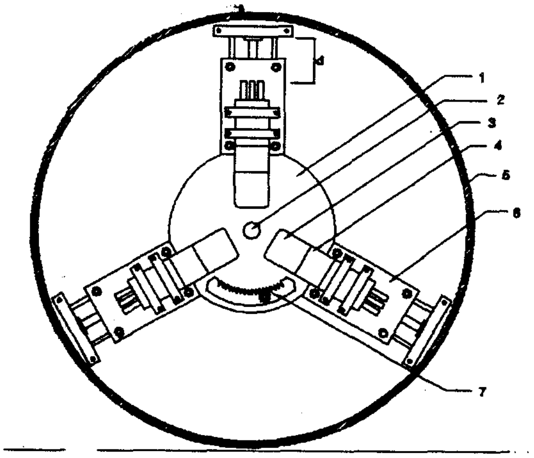

Fig. 1. Schematic Diagram of the Detection System Developed at the Present Work. The Detection Part is Entirely Located in the Cylindrical Chamber with an Inner Diameter of 500 mm, Mounted in a 20-mm Lead Shield. 1. Rotating Wheel to Push or Pull the Detectors 2. Counting Vial 3. Photomultiplier Tube 4. Preamplifier 5. Lead Shield of 20 mm 6. Detector Holder 7. a Saw-toothed Gear and d = 50 mm

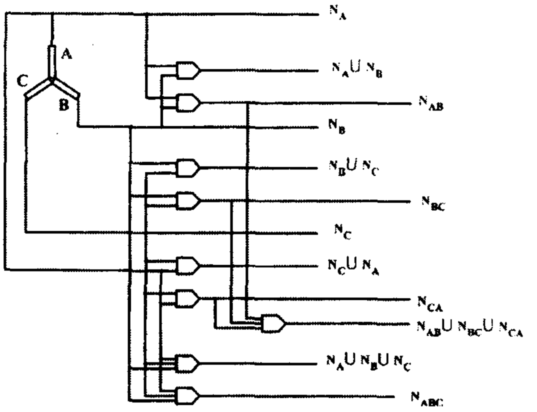

Fig. 2. Outputs Generated in the 3-PM Liquid Scintillation Counting Experiment. $N_{i,\ i-1,2,3}$; Single Counts of ith Detector. N_i ∪ $N_{j,\ i,j-1,2,3}$;Logic Sum of N_i and N_j. N_{ij}; Coincidence Counts of ith and jth Detector. N_{AB} ∪ N_{BC} ∪ N_{CA}; Logic Sum of Three Double Coincidences. N_{ABC}; Triple Coincidence Counts

measurement process such as the selection of displacement intervals or their direction and the number of intervals to be measured.

Figure 2 shows the possible outputs in the 3-PM LSC experiment. There are totally twelve parameters. The measurement of these parameters are necessary both for preliminary work to check the performance of counting devices and for data acquisition. It was designed in an integrated circuit to be able to carry out all the necessary work such as fast amplification, constant fraction discrimination, double and triple coincidence and logic sum analysis and so forth. It provide the values for nine parameters among the twelves shown in figure 2, i.e., three single counts N_A, N_B, N_C and their logic sum $N_A \cup N_B \cup N_C$, and three double coincidence counts N_{AB}, N_{BC}, N_{CA} and their logic sum $N_{AB} \cup N_{BC} \cup N_{CA}$ and a triple coincidence counts N_{ABC}. Especially, these parameters are generated at their own circuit prepared independently so as to neglect the time delay usually taken place in the analysis process itself. The type of dead time, extending or non-extending [8], can be chosen with their lengths varying from 10 to 100 μs. The experiments were carried out for nineteen intervals by displacing the detectors with a step of 2.5 mm away from the counting vial, an unquenched ^{14}C liquid scintillation solution of ultima gold contained in 20 ml standard glass vial. The activity of the sample was about 320 Bq. The dead time of each detector was extending type and adjusted to the same value of 10 μs, long enough length to suppress the afterpulses [9]. The measurement was continued for 5000 sec at each interval.

3. Analysis

The basic idea of the 3-PM LSC experiment for standardization of pure beta emitters is standing at the fact that the counting probability for double coincidence event should be larger than that of the triple coincidence at any experimental environment and their values would converge to the activity of the considering nuclide at a same time as far as the detection efficiency of all the detectors become 100 %. The detection efficiency parameter K, so TDCR value, is determined from the ratio of triple coincidence rates N_T to logic sums of double coincidence rates N_D, so $K = N_T / N_D$. And their counting probabilities converge to unity or zero values , namely, $K = 1$ or 0, respectively. In addition, since the logic sums of double coincidence rates N_D vary lineally with K around the regions where $K = 1$ according to the theory, the activity of the sample under consideration is obtained by fitting the data of N_D to least squared linear regression against K;

$$N_D(K) = a + b \times K. \tag{1}$$

where a and b are constants.

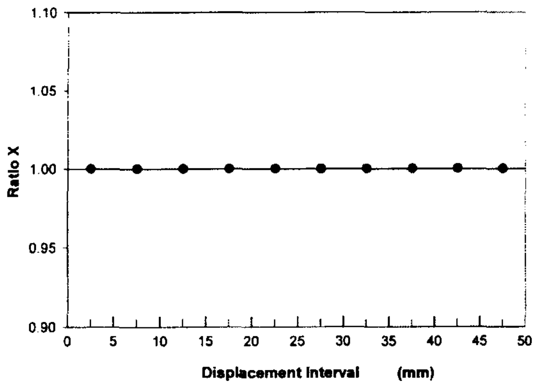

Fig. 3. Plots of $X=(N_{AB} + N_{BC} + N_{CA})/(N_D+2*N_T)$ versus Displaced Intervals Measured Away from Counting Vial with a Step of 5 mm

Thus the activity N_O is obtained by extrapolating the obtained function to $K = 1$,

so $N_O = N_D(K = 1)$.

In order to apply the method correctly, it should satisfies the following relations;

$$N_{AB} + N_{BC} + N_{CA} = N_D + 2\ N_T. \qquad (2)$$

note that $N_D = N_{AB} \cup N_{BC} \cup N_{CA}$

The eq.(2) can be derived easily from basic principle of the set theory. More details can be found elsewhere [10-12]. Eq.(2) can be used to estimate the degree of the precision and the accuracy of the measurement process itself. We measured the values of N_{AB}, N_{BC}, N_{CA}, N_D and N_T with the data acquisition system developed for this work by displacing the detectors away from counting vial. Since the activity of the sample is considerably low and the coincidence resolving time was as short as 50*ns* at most, the corrections due to the dead times and the accidental coincidences were neglected in the present work. The result is shown in figure 3. For convenience, we use notation X for the ratio of $(N_{AB} + N_{BC} + N_{CA}) / (N_D + 2\ N_T)$ and *D* denotes the displaced distances away from counting vial. The functional relation of X and D is found to be;

$$X = 1.000 - 1.870 \times 10^{-6} D \quad \text{for } 0 \leq D \leq 50 \text{ mm}$$

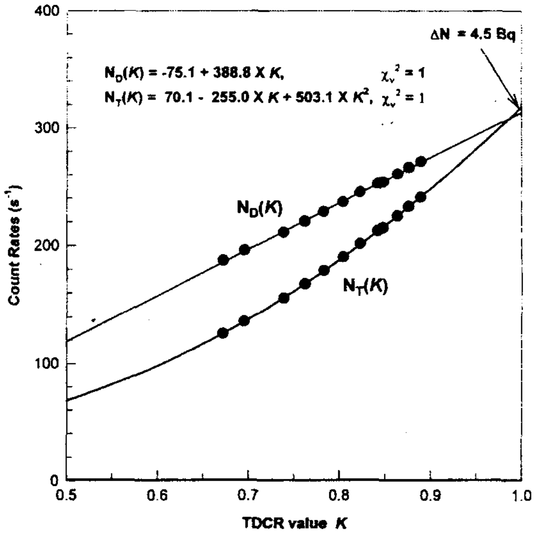

Fig. 4. Plots of $N_D(K)$ and N_T (K) Obtained by Displacing th Detectors with Steps of 2.5mm from Counting Vial. Where $\Delta N = N_T(K=1) - N_D(K=1)$.

In fact the observed values of *X* are uniform in overall the detector-displaced ranges. Thus the 3-PM LSC system developed at the present work satisfies the fundamental relation of eq.(2). Figure 4 shows the plots of $N_D(K)$ and $N_T(K)$ versus K obtained by measuring their values at each nineteen interval. The values of $N_T(K)$ decreases faster than the values of $N_D(K)$ as the detectors were moved away, and they approach each other as the detectors are more close to the vial. TDCR values for the case of unquenched ^{14}C was from 0.9 to 0.6 with the method. This extent is three times wider than those obtained by applying the defocalization technique to ^{14}C according to our earlier experiences. The plots of $N_D(K)$ varies

Table 1. Relative Uncertainty Component in %.

Component	Uncertainty(%)	Remarks
Counting Statistics	1.50	$(N_T(K=1)-N_D(K=1)/N_D(K=1)$
Weighing	0.10	$\Delta m/m$
Background	1.50	N_{BT}/N_T
Efficiency Curve extrapolation	0.50	
Ion quenching factor	1.00	P(E<1 keV)
Combined Uncertainty	2.40	Square root of summed squares

N_{BT} ; background triple coincidence counts.

P(E<1 keV) ; β decay probability with energy below 1 keV at ^{14}C

linearly on the above-described region, while that of $N_T(K)$ changes by quadratic order of K. Both plots are well fitted with least squared linear regressions for $0.6 \leq K \leq 0.9$. (see the results written in the figure 4.) Their difference at K = 1 is at most 1.5 %. The activity of ^{14}C sample was found to be 314 Bq at the reference date of November 12, 2002 UT with combined uncertainty of 2.4 %. The uncertainty was calculated from the square root of the summed squares of each standard uncertainty component tabulated in Table 1. Since the activity of the sample is on a low level, the uncertainty due to the triple-coincidence backgrounds become dominant. Otherwise the combined uncertainty can be of an order of 1 % with the method developed at the present work.

4. Summary

It is widely acknowledged that the TDCR values obtained by using the 3-PM LSC can be of true detection efficiency parameter. The advantage of the method is lying at the fact that one can determines the whole TDCR values by means of pertinent their variation method with single sample, without using additional quenched samples. Since the TDCR values are determined entirely from the triple and double coincidences, it offers the opportunity to develope various kinds of mechanical techniques which yield TDCR values varying in the wide range. However the defocalizarion method provides a few changes due to the strong restriction that the applied-voltage should be above the operating level to count the event. The geometrical variation method, developed through the present work at first, shows that it enables to derive the variation curve in the wide range of $0.6 \leq K \leq 0.9$ by displacing the detectors only 50 mm away from counting source ^{14}C, without giving any external influences arising from the counting instruments. Thus the data can be obtained by giving a few corrections commonly encountered in experiment such as statistical and instrumental uncertainties. It is three times wider extent than that observed by applying the defocalization technique according to our work. Both plots for $N_D(K)$ and $N_T(K)$ vary as they are predicted in the theory. And the derived data of $N_D(K)$ is fitted with a least squared linear function in that region definitely. Thus the activity of the considered nuclide can be obtained straightforwardly with good precision and high accuracy.

TDCR method is in developing technique as a metrology tool in the field of liquid scintillation counting. It need to have further studies to apply the method in determining the activity for electron capture decay nuclide with precision as much as good resulted in the case of pure beta emitters. We expect that the geometry-variation technique provide more insight than those being attainable with the earlier method for the case of electron capture nuclide. There were some difficulties encountered in the practical work with the method due to insufficient preliminary arrangements in designing the system. For example since the motors generate much intensive electromagnetic field during the work, they provide many noises for that moment whatever the detectors are covered with magnetic shielders. It is more convenient and cost-benefit to displace the detectors manually than using motors according to our experience.

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