Journal of the Korean Nuclear Society
Volume 36, Number 2, pp.121 ~ 126, April, 2004

Application of Geometry-Efficiency Variation Technique to Activity Measurement of ^{204}Tl for 3-PM Liquid Scintillation Counting

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(Received September 30, 2003)

Abstract

3-PM liquid scintillation counting using the geometry-efficiency variation technique has been applied to the activity measurement of ^{204}Tl, which decays to ^{204}Hg and ^{204}Pb by β^- and E.C., respectively. The TDCR values K have been derived over a wide range, $0.78 < K < 0.97$, by displacing the detectors up to 50 mm away from an unquenched liquid scintillation sample ^{204}Tl. The derived plots of the logic sums of double coincidences $N_D(K)$ very K vary linearly in the observed regions. The fractions of losses due to electron capture decay have been taken into account by employing a PENELOPE Monte Carlo simulation. The calibrated activity is 102.3 kBq at a reference date of July 1st, 2002 (UT) with a combined uncertainty of 0.63 %. This is consistent with the value determined by means of the CIEMAT/NIST method at KRISS.

Key Words : 3-PM LSC, geometry-efficiency, TDCR, defocalization, 204Tl

1. Introduction

The 3-PM Liquid Scintillation Counting (3-PM LSC) method has been successfully applied in determining the activity of pure beta emitters and some radionuclides which decay by electron capture[1-11]. With this method, the efficiency parameters K ($=N_T/N_D$), or TDCR value K, can be obtained by the ratio of triple coincidence rates N_T to logic sums of double coincidence rates N_D [1-3]. (hence the 3-PM LSC method is alternatively called the TDCR(Triple to Double Coincidence Ratio) method). Thus the activity of the considered sample is obtained directly, without preparing several quenched sources or tracers so as to calibrate the detection efficiencies. Owing to this mojor advantage, this method is under development at many laboratories.

In the 3-PM LSC method, a defocalization technique that changes the focus voltage, loading

between the photocathe and first dynode, has been developed in order to vary the efficiency parameters. However, the focus voltages should be maintained above certain values in order to count the events. Usually, from +800 V to +650 V, the efficiency parameters are automatically changed slightly. For example, it is of a few percent, even in the case of ^{14}C, according to our earlier work.

Recently, we have been developing an improved technique in order to ouercomp the aforementioned difficulties. It achieves the efficiency variations by altering the geometry of the detection position, namely by uniformly displacing the detectors from the counting vial. The virtue of this method has been demonstrated by studying the activity of a pure beta emitter ^{14}C [12]. In our earlier work, the TDCR value K was deriviable in the regions of $0.6 < K < 0.9$ by displacing all detectors up to 50 mm from an unquenched ^{14}C liquid scintillation sample. The variable extent for K with the method is three times wider than those derived by applying the defocalization method. Interestingly the plot for N_D versus K converges linearly to $K = 1$.

Eventually we will improve the geometrical efficiency variation method as a pertinent technique for the 3-PM LSC method. To realize our intent, we should first rerify whether i) it is well applicable even for the cases of electron capture decay nuclei, and ii) the plot of $N_D(K)$ converges to $K = 1$ by a first order linear function.

For these aims, we prepared an unquenched liquid scintillation sample ^{204}Tl of which activity was certified with the CIEMAT/NIST method developed by *KRISS (Korea Research Institute of Standards and Science)*. It decays to its daughters, ^{204}Hg and ^{204}Pb, by means of electron capture (2.6 %) and β^-(97.4 %) decay, respectively, And, since the beta end point energy is much larger than that of ^{14}C, we could expect clear observation of the convergent shape of $N_D(K)$ around $K \simeq 1$.

2. Experiment

We briefly describe the 3-PM LSC system using the geometry-efficiency variation technique developed at our laboratory. More details can be found elsewhere [12-14].

The detection part is entirely located in a chamber with an inner diameter of 500 mm, shielded with lead 20 mm in thickness, Each detector is symmetrically equipped around the centrally located counting vial. Since a vice ties up all detectors, they are displaced uniformly at the same time. A computer controls all the measurement processes such as the selection of displacement intervals and their direction and the number of intervals to be measured. For data acquisition, we developed a specially designed integrated circuit with which all the necessary tasks, such as fast amplification, constant fraction discrimination, double and triple coincidence and logic sum analysis, can be performed at a same time. It provides the output values of nine parameters, 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 $N_D(=N_{AB} \cup N_{BC} \cup N_{CA})$ and a triple coincidence counts N_{ABC}. The type of dead time, extending or non-extending[15], can be chosen at will. And both the length of dead times and the coincidence resolving times are variable from 10 to 100 μs and 10 to 1000 *ns*, respectively. Fig. 1 shows the schematic diagram of the data acquisition method specially developed for this work.

The experiments were carried out at eleven intervals by displacing the detectors with a step of 5 mm away from the counting vial. We prepared an unquenched 204Tl liquid scintillation solution of ultima gold contained in a 20 ml standard glass vial. It is KRISS (Korea Research Institute of Standards and Science) certified radionuclide by means of the CIEMAT/NIST method. The

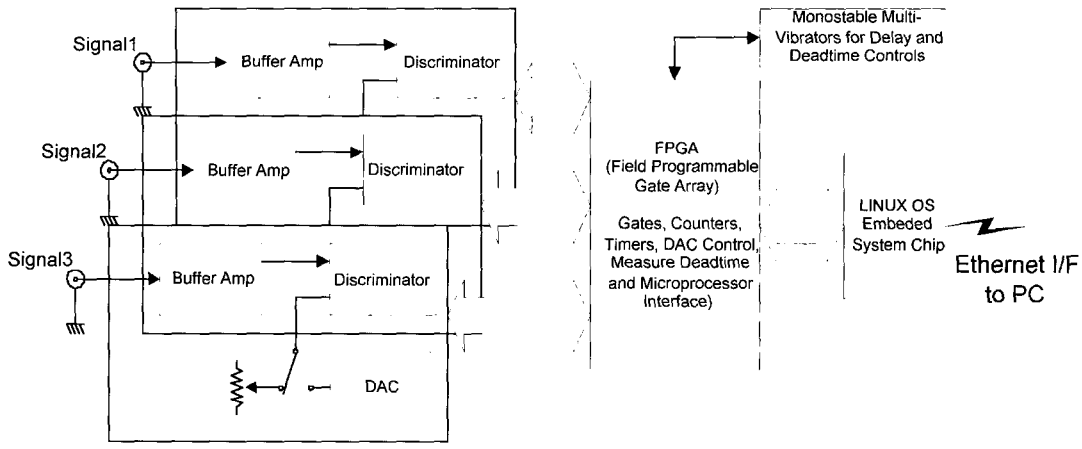

Fig. 1. Schematic Diagram of TDCR Control Circuit

calibrated activity is 101.8 kBq at a reference date of July 1st 2002 (UT). The dead time of each counting channel was non-extending and adjusted to be sufficiently long, 15, 25, 35 and 40 μs, respectively, so as to supress afterpulses[16]. At each chosen dead time, measurement was continued for 2500 sec for each counting interval and the backgrounds were counted in the same manner as for the for counting sample.

3. Analysis

The basic principle of the 3-PM LSC method is that the TDCR value K converges to unity whenever the detection efficiencies of all detectors become 100%. In fact, the TDCR value K, determined from the ratio N_T to N_D, is not an exact detetetion efficiency, but the activity of the considered sample can be obtained at $K = 1$ by extrapolating the fitted function to the observed data of $N_D(K)$. Thus, the activiry N_o of the considered sample is given by $N_o = N_D(K=1)$;

$$N_D(K) = a + b \cdot K$$
$$N_o = N_D(K = 1) \qquad (1)$$

where a and b are constants.

In order to apply the method for a practical experiment, the following relation should be satified:

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

where N_{ij} is the observed double coincidences between counting channels i and j and N_D is the logic sums of double coincidences $N_D = N_{AB} \cup NBC \cup NCA$.

Since eq.(2) is a fundametal relation according to set theory, it should hold always regardless of the experimental emvironment. Therefore, it can be a good testing ground to judge if it is possible to obtain data with good precision with the geometry-efficiency variation technique. To this end, we investigated the relation shown in eq.(2) with the counting sample ^{204}Tl before the practical work. Parameters such as N_{AB}, N_{BC}, N_{CA}, N_D and N_T were measured by displacing the detectors away from the counting vial for each dead time varying from 15 to 40 μs.

As a result, the functional relation of X and D is observed to be;

$$X = 1 - 1.00019 \times 10^{-6} D \quad \text{for } 0 \text{ mm} \leq D \leq 50 \text{ mm}.$$

Where X denotes the ratio of $(N_{AB} + N_{BC} + N_{CA}) / (N_D + 2*N_T)$ and D denotes the distance from the measuring vial to detector window.

The result shows that the observed values of X are constant in the range of the given source-to-detector distance. Thus, the 3-PM LSC technique, developed at our laboratory satisfies eq.(2) in considering radionuclide, regardless of the displacement of the detectors and the length of dead times. The actual experiments were carried out as done in the preliminary experiment but the dead time of each counting channel was varied from 15 to 40 μs. The TDCR value K was obtained from the ratio of N_T to N_D measured at each counting interval. Since the activity of the considered nuclei is of a relatively low level and the resolving time is as short as 100 ns, corrections due to accidental coincidences have not been considered in the coincidence counting channels. For each selected dead time, the plots of N_D and N_T are drawn as a function of K, respectively, and they are shown in Fig. 1 for the case of a dead time of 35 μs. For the detector position of 0 to 50 mm, $0.78 < K < 0.97$ according to our data.

In the earlier work for ^{14}C, the derived extent for K was $0.6 \leq K \leq 0.9$ with an unquenched sample, but K_{max} was too low to predict the convergent shape of $N_D(K)$.

However, in the case of ^{204}Tl, since the maximum beta end point energy is four times larger than the case of ^{14}C, K_{max} is increased by 7%, and is sufficiently high to see the convergent shape of $N_D(K)$.

The plot of $N_D(K)$ varies linearly over the observed region $0.78 \leq K \leq 0.97$, while that of $N_T(K)$ changes by quadratic orders of K. Both plots are close to each other whenever K converges to unity, as predicted in the theory. We concluded that $N_D(K)$ converges linearly to $K = 1$ definitely in the region of $0.78 < K$ according to our data.

^{204}Tl decays to its daughter nuclides by both beta and electron capture decays[17]. The branching ratios of the latter case are small, but their effects should be considered in order to determine the activity of the considered sample with high accuracy. The electron capture decays take place with accompaning X-rays and Auger electrons. Since X-rays are extremely penetrative, they scarcely deposit any energy in the solution. Their effects on the detection efficiency are calculated for each TDCR value K by modeling the principal physical phenomena occurring in the detector. Although this model is mainly used for pure-beta emitting radionuclides, it can be extended to

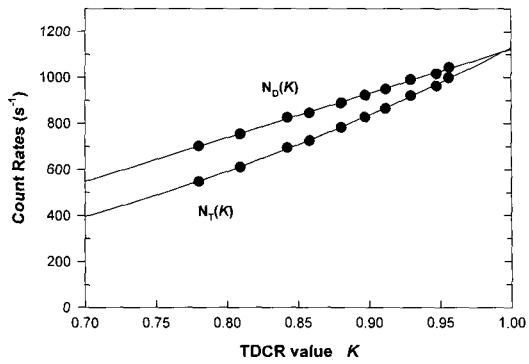

Fig. 2. Plots of $N_D(K)$ and $N_T(K)$ as a Function of K with an Unquenched Solution of ^{204}Tl

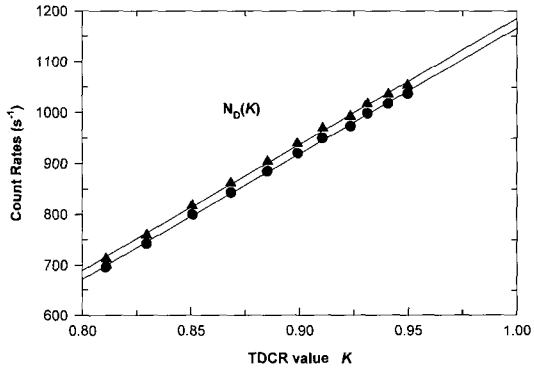

Fig. 3. Plots for ND(K) of which Resulted with Consideration of the Electron Capture Effects (▲) and Obtained in the Present Experiment(●)

Table 1. Relative Uncertainty Components in %

Component	uncertainty(%)	Remarks
Counting Statistics	0.04	$1/N_D(K_{max})^{1/2}$
Weighing	0.09	$\Delta m/m$
Deadtime	0.05	
Resolving time	0.02	
Half-Life	0.02	Calculated from Decay Data
Displacement	0.04	$(N_{AB}+N_{BC}+N_{CA})/(N_D+2*N_T)$
Decay Parameters	0.01	Simulated Output
Background	0.04	N_{BD}/N_D
Extrapolation of efficiency curve	0.62	
Combined Uncertainty	0.63	quadratic sum of uncertainty component

include electron-capture radionudlies such as 204Tl. The probability of interaction of photons in the scintillator is obtained by employing a PENELOPE [18] Monte Carlo simulation.

Fig. 3 shows both plots of $N_D(K)$, including the electron capture effects(▲) and that obtained from the experiment(●). For both cases, $N_D(K)$ varies linearly in the range of $0.78 < K$ and is well fitted with the linear least square for $0.78 < K < 0.97$, respectively. The activity of ^{204}Tl was obtained by extrapolating the fitted function obtained with each selected dead time. and it is 102.3 kBq/g at a reference date of July 01, 2002 (UT). This is consistent with the calibrated value of 101.8 kBq/g at *KRISS* by means of the CIEMAT/NIST method. The combined uncertainty was found to be 0.63 %. The uncertainty was calculated from the square root of the summed square of each standard uncertainty component, as tabulated in Table 1. Regarding the uncertainty components, the largest contribution was due to the curve fitting.

4. Conclusions

The object of this work was to verify that the geometrical efficiency variation technique, developed in our laboratory for 3-PM LSC, is an effective technique for determining the activity of a radionuclide, even for the cases when the rodionuclides to their daughters by both β^- and electron capture. We have successfully demonstrated the effectiveness of our approach by applying the method in determining the activity of ^{204}Tl.

The result is consistent with that obtained by means of the CIEMAT/NIST method using two photomultiplier tubes as detectors. The CIEMAT/NIST method has been acknowledged as an advanced technique for activity measurement, [19,20], but it sequires a combination of measurements and theoretical calculations, both for ^{3}H, used as an efficiency tracer, and the nuclide being calibrated. With 3-PM LSC, it is possible to obtain the activity directly from the consideried sample itself. However, the defocalization technique cannot provide the efficiency parameters when one varied to a wide extent to due the reasons nofed in section 1.

The geometrical efficiency variation method enables us to extend K into a wide range $0.78 < K < 0.97$ by simply displacing the detectors only 50 mm away from the counting source 204Tl. The newly observed range is several times wider than that with the defocalization method. The obtained data of $N_D(K)$ is fitted by a first order linear regression on that region. Thus, the activity can be determined straightforwardly with high precision and accuracy.

The 3-PM LSC method is a technique that is seeing ongoing development as a metrology tool in the field of liquid scintillation counting. It is widely acknowledged for the cases of pure beta emitters that the TDCR value K can be a true efficiency parameter enabling determination of the activity directly from the consideried sample. However further studies are needed to apply the method to cases of electron capture decay or

delayed transition radionuclides. We expect that the geometry-efficiency variation technique will offer more insight into than the defocalization method for the cases of such radionuclides.

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