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Activity Determination of ^{14}C and ^{204}Tl by Applying the 3-PM LSC Technique

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Abstract

We have developed an improved LSC(Liquid Scintillation Counting) method using a three photomultiplier tubes as detectors and developed a three dimensional data acquisition method with which pulse heights from an array of detectors can be multi-scaled with dwell time 10 ns. Since the method enables to obtain the absolute detection efficiencies directly overall the regions of interests by measuring the triple to double coincidence counting ratios, the method is particularly suitable for experiments such as both double and triple coincidence distributions are required. Virtues of the method are demonstrated by measuring the activity of ^{14}C and ^{204}Tl. The computer discrimination together with the MCTS (multi-channel time scaling) technique is applied to determine the TDCR efficiency functions in the analysis. Fuller details of accounts of applying the technique is given in this report.

I. Introduction

The LSC(Liquid Scintillation Counting) method has been recognized as the pertinent method in determining the activity of pure beta decay nuclides, so has been in use over 30 years. However there are some difficulties to get over by applying the commonly used technique and those might be summarized by two categories as following; i) since the method use the PM tubes as detectors, one usually encounters with lots of random events such as dark currents and afterpulses. The double coincidence technique can be of alternative method which enables to distinguish them by using the off-the-shelf electronics, as far as the activity of the sample is sufficiently low to neglect the accidental coincidences in the analysis. Ii) and it requires to prepare several quenched samples or the other substitutable materials to obtain the absolute detection efficiency on the regions available to obtain the activity of the considering sample.

Recently, an improved technique, known as TDCR(triple to double coincidence ratio) method[1-5], which enables to overcome the above mentioned difficulties has been developed and successfully applied on the measurement of activity of pure beta emitters. The TDCR method uses three PM tubes as detectors, so also called by 3-PM LSC. Since the strong advantage of the method is lying on the facts that the triple to double coincidence ratio can be of absolute detection efficiency, thus the activity of the considering sample is determined directly by slightly change the electronic conditions, without using the quenched samples or the other materials which play similar roles. For example, the efficiency can be varied by defocalizing the first dynode at PM tube.

We have been developed a noble data acquisition method with which pulse heights from an array of detectors can be digitized and clocked with dwell time as short as 10 ns.[6-8] Since the all necessary parameters such as pulse heights and clock-time information, are stored sequentially by using the method, pulse heights, clock time and resolving time selections can be done at will. Thus the overall of TDCR efficiency can be derived by applying the computer discrimination together with multi-channel time scaling(MCTS) method at the same time from the single data, not multiple data set.

In this report, the virtues of applying the TDCR method, using the data acquisition technique developed at our laboratory, is demonstrated by measuring the activity of pure beta emitters, ^{14}C and ^{204}Tl.

In section II, both the theoretical foundations for TDCR and the data acquisition method are described. The experiment and both the MCTS and the computer discrimination method to obtain the overall TDCR efficiencies and the use of Cox and Isham formulae[8,9] for corrections, due to dead time and accidental coincidences, are described in section III and section IV, respectively. Consequence of the present work is at section V.

II .Theory and Data Acquisition Method

II-1. Theoretical Foundations of TDCR method.

Since the TDCR method uses three PM tubes as detectors, there are totally twelve output signals, three are from each counting channel (N_A,N_B,N_C), and three from logic sums(N_A+N_B, N_B+N_C, N_C+N_A) and coincidence counts(from each couple of detector(N_{AB}, N_{BC}, N_{CA}) respectively, logic sums of each single count (N_A+N_B+N_C) and double coincidence counts(N_{AB}+N_{BC}+N_{CA}), respectively and triple coincidence counts(N_{ABC}). If the detection efficiency for each counter converges to unity, then all the listed output counts approaches to activity of considering sample, respectively, as can be seen in figure 1, which shows efficiency probability functions corresponding to each output signal. However all the outputs stem from each single counter contain large amounts of random events due to dark currents and afterpulses. The random events can be monitored and reduced drastically by applying the coincidence counting method.

According to the TDCR method, since the triple to logic sums of double coincidence ratio can be of direct detection efficiency K, and so it is; $K = N_T / N_D$, where N_T (= N_{ABC} =N_o □ e_T) is for triple coincidence rates with detection efficiency e_T and N_D (= N_{AB} + N_{BC} + N_{CA} = N_o □ e_D) logic sums of double coincidence rates with detection efficiency e_D for considering sample with activity N_0. As the detection efficiency converges to unity value, so K = 1, each value of N_T and N_D becomes activity of the sample, respectively. According to the TDCR theory, the values for relatively higher

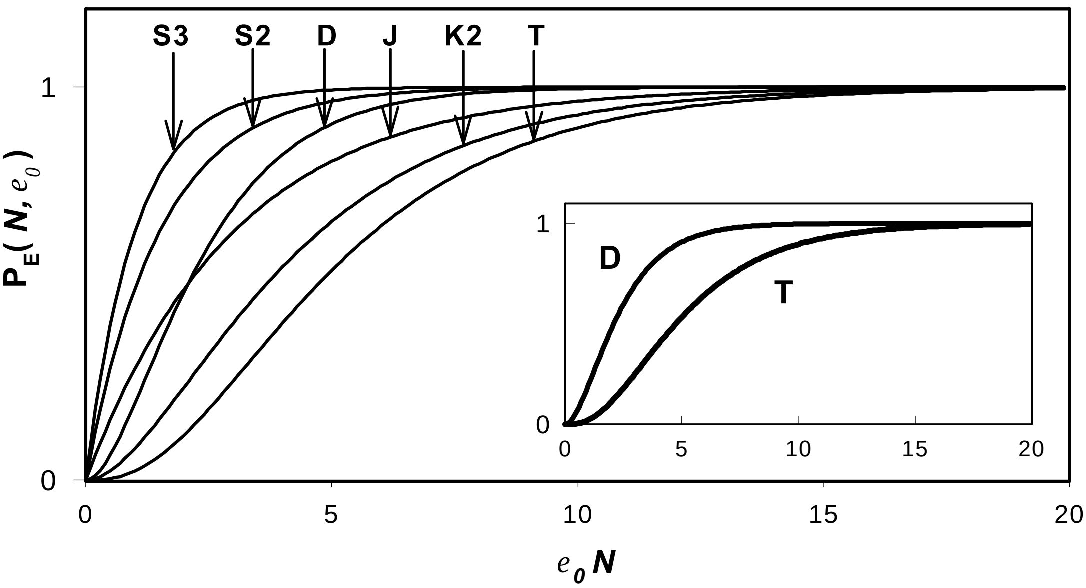


Fig. 1 Detection probability functions of each output pulse of 3-PM LSC system, where J= N_A or N_B or N_C ,S2= N_A + N_B or N_B+N_C or N_C+N_A, S3= N_A + N_B + N_C ,K2= N_{AB} or N_{BC} or N_{CA} ,D =N_{AB} + N_{BC} + N_{CA} ,T= N_{ABC}

detection efficiencies. Thus the activity is obtained by fit the data to a linear function of K with which lying on the just mentioned regions, i.e.,

$$N_D(K) = a \bullet K + b.$$

Consequently the activity of the considering sample is obtained by extrapolating the data to $K = 1$, $N_D(K = 1) = N_0$

II-2. Data Acquisition Method.

We have developed three dimensional data acquisition(3D-DAQ) method with which pulse heights from an array of detectors can be digitized and clocked with a resolution of

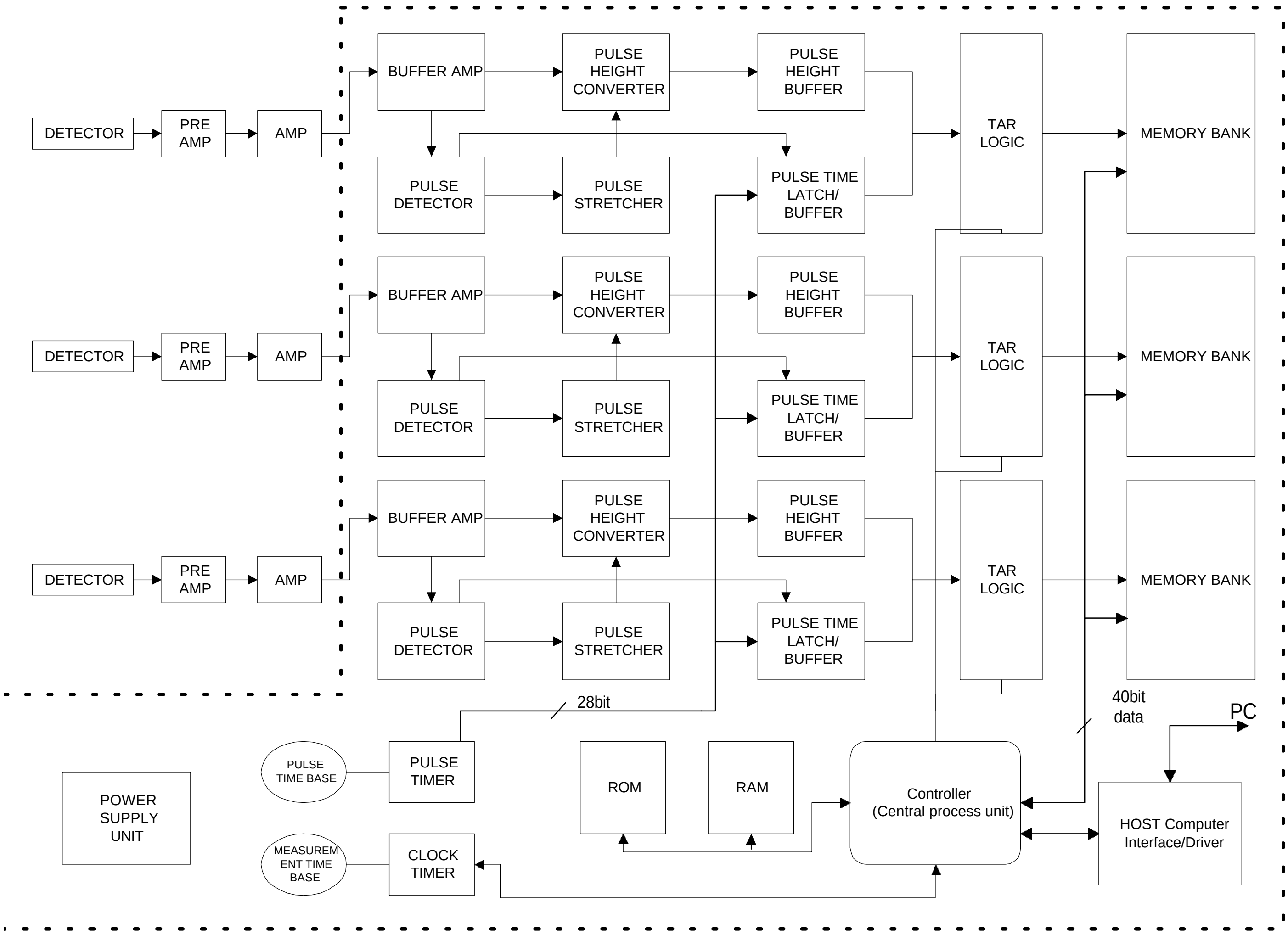


Fig. 2 Simplified block-diagrams of 3D-DAQ system

an order of 10 ns. The method is particularly suitable on experiments both the double and triple coincidence distributions are required such as TDCR experiment. We will briefly describe the data acquisition method. More details can be found at our earlier reports. The main part of the 3D-DAQ system consists of three ADCs of SAR (Successive Approximation Register) type, a 100 MHz oscillator clock and DRAM (Dynamic Random Access Memory) of 192-Mbyte capacity. Figure 2 shows the simplified block diagram of the system. Linear pulses from an array of detectors are transferred to each *Buffer Amplifier*, that is prepared to maintain the pulse shape with original one. The pulse is differentiated first and then inverted immediately at the P*ulse Detector*. Then both pulses are input to C*omparator* and strobe pulse is generated at crossing position of each other, thus at the peak position of the original pulses. Then the strobe pulse latches output of *Timer* which consists of 28 bits synchronous counter and a calibrated 32 MHz oscillator. Thus clock time is obtained at an uniform position without
dealing with pulse height.

And the peak position is just the rising position of strobe signal. The pulse height is detected and digitized by applying the 12 bits ADC of SAR type. The P*ulse Stretcher* generates a stretched pulse at the peak position for the longer period than the ADC conversion time. Then the pulse height is converted to the binary number for duration of a stretched pulse. Briefly, the linear pulses from the detectors are digitized and latched by the pulse generated at peak position and then immediately transferred together with clock time to an array of 5.37 ´ 10^8 bits DRAM, of 40 bit width, 12 bits for pulse height and 28 bits for clock time. When the data acquisition ends, the DRAM content is transferred to a larger storage device, DRAM is cleared and the fast accumulation cycle starts again. For example, when the count rate is 1000 s^{-1}, one can obtain at most 1.34×10^7 events each of which consists of a digitized pulse height and the sequentially recorded clock time from 0 s to 1.34×10^4 s. Concerning the dead time selection, the TDCR method uses extending dead time technique. However there are disadvantages in case for using the method. There are two types of dead times, extending and non-extending type, commonly applied for most experiments. Using the former type with high activity samples, since the every input pulses can be accepted even the analyzer is being busy, so the dead time may be extended significantly large values from time to time. Thus it usually looses lots of true events. On the other hand, since the every input pulses are rejected during the analysis in case for applying the non-extending, it is pertinent to

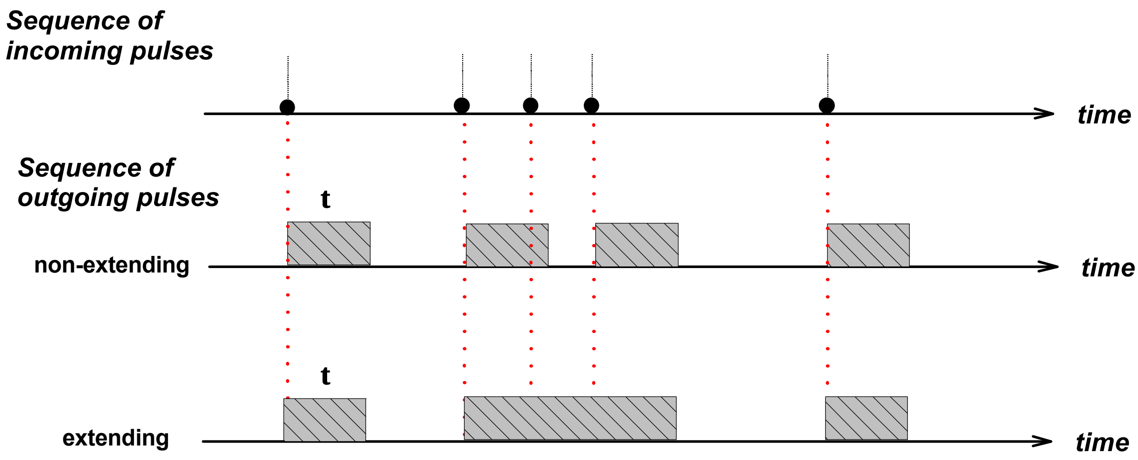


Fig. 3 Illustrations of two types of Dead Times commonly applied in most experiments

use for higher activity sample. The two types of dead times are showing in figure 3. And there is another advantage with non-extending type. Since the coincidence channel usually counts the accidental coincidences, thus those should be subtracted from the observed coincidences. Fortunately, there has been found exact formulae available for this case.

In the present work, the dead time of each counting channel is non-extending type, determined by the width of stretched pulse described above and can be variable from 10 ms to 50 ms in this study.

III. Experiment

In the conventional TDCR method, it requires the fast electronics using an off-the-shelf coincidence analyzers and other ancillary electronics such as fast discriminators. However, since all the necessary parameters can be stored sequentially by using a 3D-DAQ, it needs only three linear amplifiers for pulse shaping as shown in figure 1. The detection part is placed in the cylindrical chamber with inner radius of 400 *mm*, mounted in a 20 *mm* lead shield. The detectors used in this work were HAMAMATSU R1847-07 photomulitiplier tubes, a 14 stage fast-linear focussed type with bialkali photocathode of 51 mm diameter in each. These are symmetrically equipped around the centrally located counting vial, and maintained at 30 *mm* distance from the center, respectively. The radionuclide sample used for this work is unquenched ^{14}C and ^{204}Tl solutions contained in 20 *ml* standard glass vial. The dead time of each channel was of the non-extending

type and adjusted to the same value of 25 ms, respectively. The experiments were carried out four times for one sample, and the measurement time was set to 10^3 s for one measurement.

IV. Analysis and Results

TDCR(Triple to Double Coincidence Ratio) method in liquid scintillation counting is one of the technique which can directly determine the activity without requiring information of such as counting efficiency. The activity can be obtained by extrapolation of data points of N_D as a function of K factor which is N_T / N_D, where N_D denotes the logic sums of three double coincidence rate and N_T is for the triple coincidence rate.

We show the methods to derive the values of K and the activity from the data files obtained by using a 3D-DAQ.

The true count rate r_i of the pulses from the ith detector is related to the observed rate R_i and dead time t_i by

$$r_i = \frac{R_i}{1 - R_i \cdot t_i}. \quad \text{where} \quad i = A,B,C$$

And the observed double and triple coincidence rates can be written by;

$$R_{ij} = R_{cij} + R_{fij}. \quad \text{where } i,j = A,B,C$$

$$R_{ABC} = R_{cABC} + R_{fABC}.$$

where R_{ij} denotes the observed coincidence rate derived by comparing the events detected on i^{th} channel with j^{th} channel. R_{cij} (R_{fij}) is the genuine (accidental) coincidence rate. R_{ABC} is the observed triple coincidence rate and R_{cABC} (R_{fABC}) is the genuine(accidental) triple coincidence rate.

Since the data obtained with our 3D-DAQ system consist of chronologically ordered records of pulse height and the corresponding clock time for each channel, the pulse-height, clock time and the discrimination-range selections can be done at will from the same data. To determine the R_{ij} in eq.(2), the recorded clock time of each i^{th} event is

compared with clock times of the j[th] data. Events are selected in which the time difference of each compared event is within the resolving time r, and set to be same value of 1.2 ms for all channels.

Thus each double coincidence rate, R_{AB}, R_{BC} and R_{CA} is derived, and its corresponding true coincidence rate r_{ij} can be determined by using a Cox and Isham formula. Since $r_i = r_j = r$ and $t_i = t_j = t$ for all i and j in this study, it becomes;

$$r_{ij} = \frac{R_{ij} - 2rR_iR_j}{(1-R_i \cdot t)(1-R_j \cdot t)X(t, r, r) + R_{ij}tY(t, r)}.$$

The function $X(t, r, r)$ is to correct for the accidental coincidences included in R_{ij} and the function $Y(t, r)$ is for the dead-time corrections. An exact solution is known for the case where $t_i = t_j = t$. Fuller details can be found elsewhere.[10]

Thus the values of $N_D = r_{AB} \cup r_{BC} \cup r_{CA}$ can be directly obtained by using the simple algebraic relation;

$$r_{AB} + r_{BC} + r_{CA} = N_D + 2 \bullet N_T.$$

Triple coincidence rate r_{ABC} (= N_T) can be obtained by; first creates the data file {R_{AB}} of which each line consists of the pulse height and clock time for both of A[th] and B[th] events those are in coincidence relation. And secondly, the recorded clock time of each C[th] event compares with both registered time in each line of {R_{AB}}, and select the events in which the time difference of each comparison is within the resolving time, respectively. The method for collections of triple coincidnece events are summarized in Table 1.

Therefore R_{ABC} is derived and r_{ABC} can be obtained by using a Cox and Isham formula[9,10], but the fraction of dead time losses P_{AB} for set {R_{AB}} must be taken into account in the formula. Since there are totally for possible states, for example counter A and B. In order to count the coincident events, then both counter should be open simultaneously, otherwise fail to count the true event. It is definitely depends on the length of dead times adjusted to each channel, respectively. It is illustrated in figure 4.

Table 1. Selections of triple coincidence events.

R_{AB}					
CH-A	TIME A	CH-B	TIME-B	CH-C	TIME-C
158	1286402405	136	1286402220	111	1286402042
579	1286599217	457	1286599095	384	1286598886
197	1287247999	291	1287247939	515	1287247698
168	1287402967	397	1287402877	336	1287402636
385	1287521780	196	1287521720	159	1287521479
301	1288251780	225	1288251720	99	1288179042
332	1288610655	255	1288610595	335	1288251511
		197	1288610354		
Triple Coincidences					
7	1288919842	385	1288919939	46	1288694323
85	1289638436	74	1289638345	84	1288730417
95	1289891249	60	1289891158	216	1288919698
110	1289943342	141	1289943283	0	1289010042
297	1290277592	463	1290277627	91	1289638104
272	1290650592	394	1290650595	135	1289890917
39	1291419624	10	1291419689	65	1289943104
75	1291736342	171	1291736439	52	1290083292

The probability P_{AB}, for both of A and B counters are open simultaneously, is obtained as;

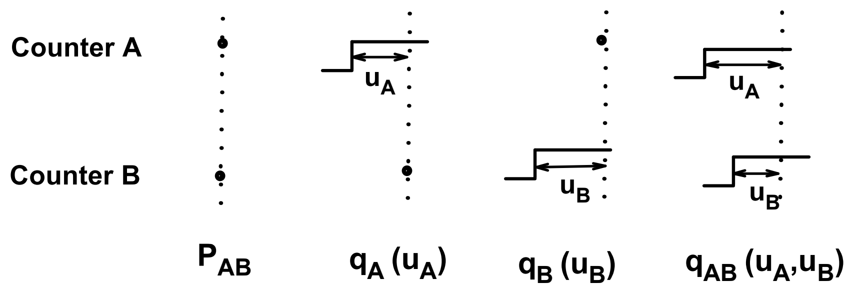
$$P_{AB} = \frac{R_{AB}}{r_{AB}}$$

Thus the factor P_{AB} is taken into accounts to Cox and Isham formulae to correctly determine the true triple coincidence rates r_{ABC} and so it is;

$$r_{ABC} = \frac{R_{ABC} - 2rR_{AB}R_C}{(1 - R_{AB} \cdot t)(1 - R_C \cdot t)X(t, r, r) + R_{ABC}tY(t, r)} \bullet \frac{1}{P_{AB}},$$

Thus K is directly obtained by the definition, $K = \frac{N_T}{N_D}$.

A set of the values $\{K_i\}$ were obtained by applying the computer discrimination



P_{AB} ; Probability for both counters are open

$q_A(u_A)$; Probability for counter A has been blocked for a time u_A($0 < u_A < t_A$)
and counter B is open at that instant

$q_B(u_B)$; Probability for counter B has been blocked for a time u_B($0 < u_B < t_B$)
and counter A is open at that instant

$q_{AB}(u_A,u_B)$; Probability for counter A has been blocked for a time u_A and
counter B for a time u_B

Fig. 4 Four possible states of counter A and counter B.

technique in the region where K_i ranged from 15 % to 94 % in case of ^{14}C and 25 % to 99 % for ^{204}Tl. Figure 5 shows the LSC spectrums of ^{204}Tl obtained with each of counter used at the present work, along with the computer discrimination range set on each spectrum at off-line. Figure 6 and figure 7 show plots of $N_D(K)$ and $N_T(K)$ versus K. As K increases or decreases, both functions $N_D(K)$ and $N_T(K)$ converge to almost same value simultaneously. The function $N_D(K)$ is obtained by means of least squares fitting in the region where it varies linearly. And the data points of $N_D(K)$ were extrapolated to $K=1$ by the linear least-squares method in the region of K ³ 60 % for ^{14}C and K ³ 75 % for ^{204}Tl. Then both the activity of ^{14}C and ^{204}Tl were determined from $N_D(K=1)$, and the results were found to be (2761 ± 21) Bq and (338 ± 3) Bq at reference date of 1 March 11, 2001.

V. Conclusions

The account of applying a TDCR method using the three-dimensional data acquisition

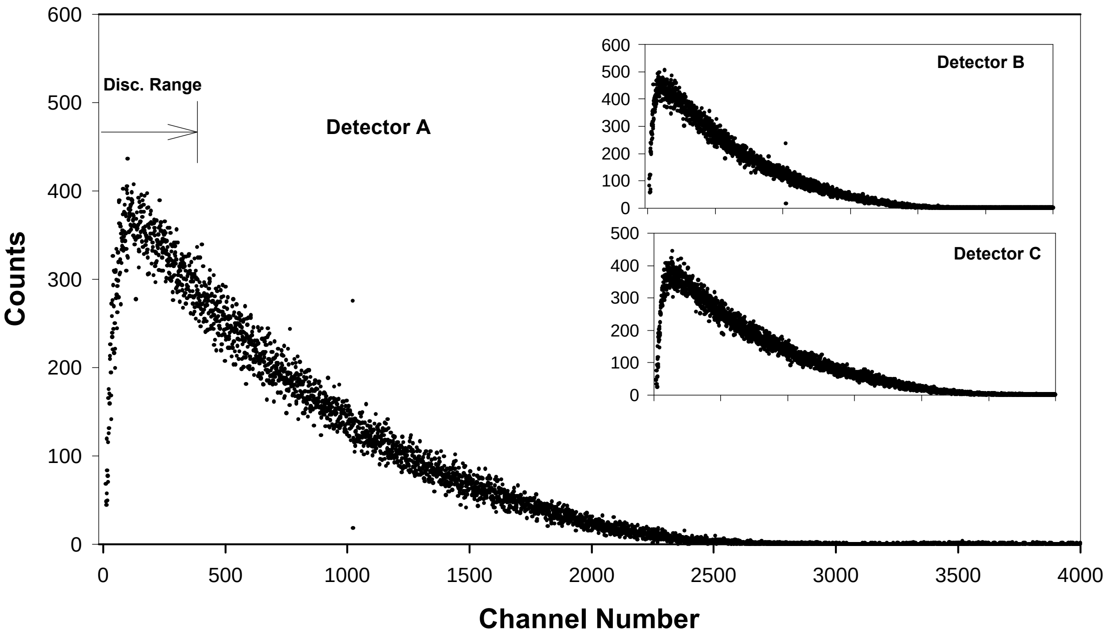


Fig.5 Singles liquid scintillation spectrum of ^{204}Tl obtained by using the 3-PM LSC at the present work. The computer discrimination range is marked on there.

method developed at our laboratory has been fully demonstrated by measuring the activity of ^{14}C and ^{204}Tl. As it is stated at the section I, there are some difficulties encountered when using the earlier methods, for example large fractions of random events and extra materials for efficiency variations. With the TDCR method using three PM tubes as detectors, such difficulties are negligible. In addition, multi-channel time scaling together with computer discrimination enable to obtain the efficiency functions overall the regions. Thus the activity is determined only with a sample, not multiple sample. Perhaps it is still open question whether the triple to double coincidence ratio can be of true absolute detection efficiency in LSC experiment. It requires sufficient data to conform it 100 %. However, we can say that the method is quite suitable for activity measurement of pure beta emitters such as ^{14}C and ^{204}Tl. In additions, since the expected efficiency is sufficiently high, the result can be yielded with good precisions and high accuracy.

In the analysis, since all necessary parameters (pulse heights and clock times) are stored sequentially, the overall TDCR efficiencies can be derived by applying the multi-

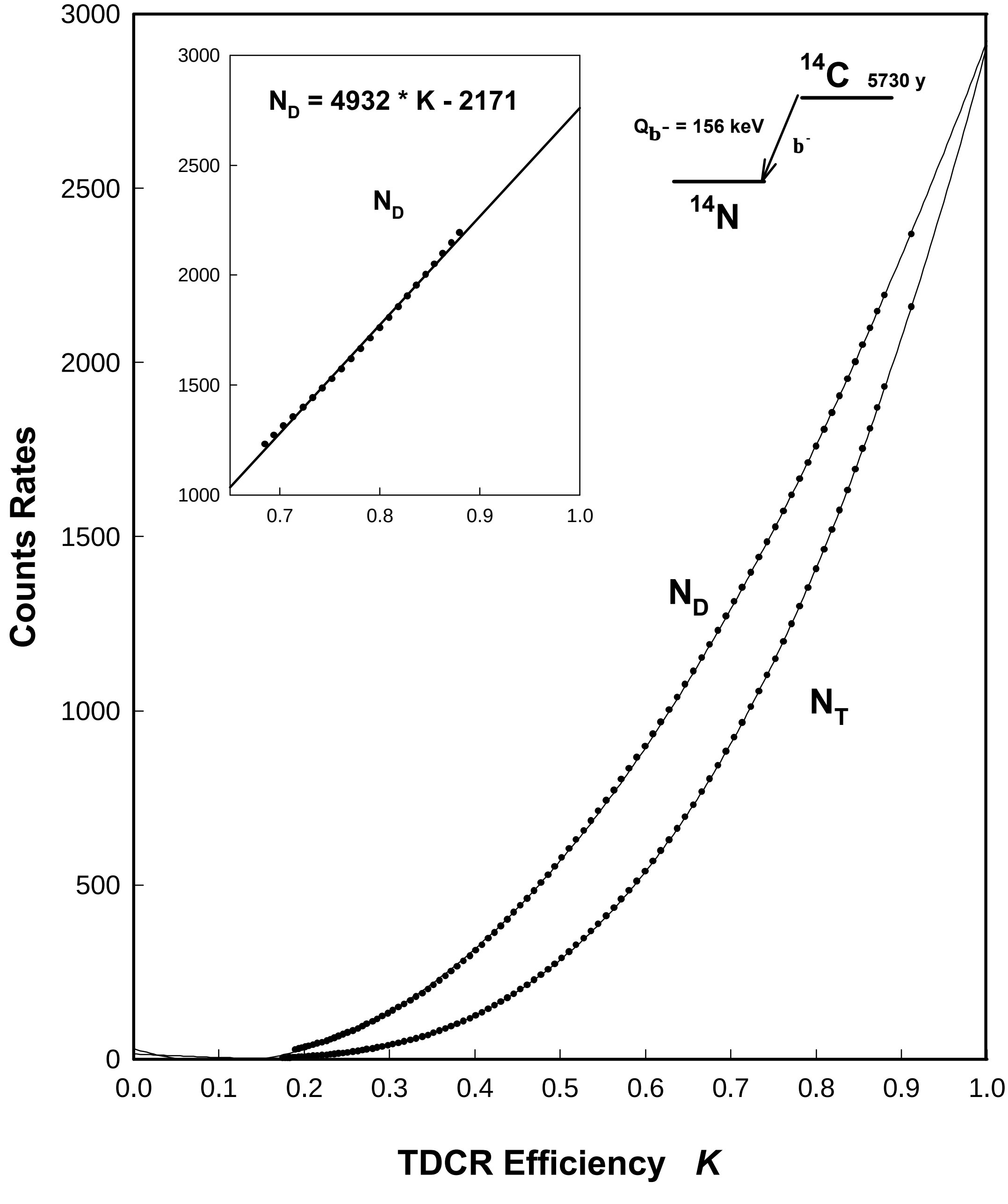


Fig. 6 Plots of $N_D(K)$ and $N_T(K)$ versus K. The data points were extrapolated to K = 1 by the linear least-squares fit in the region of K ³ 60 %.

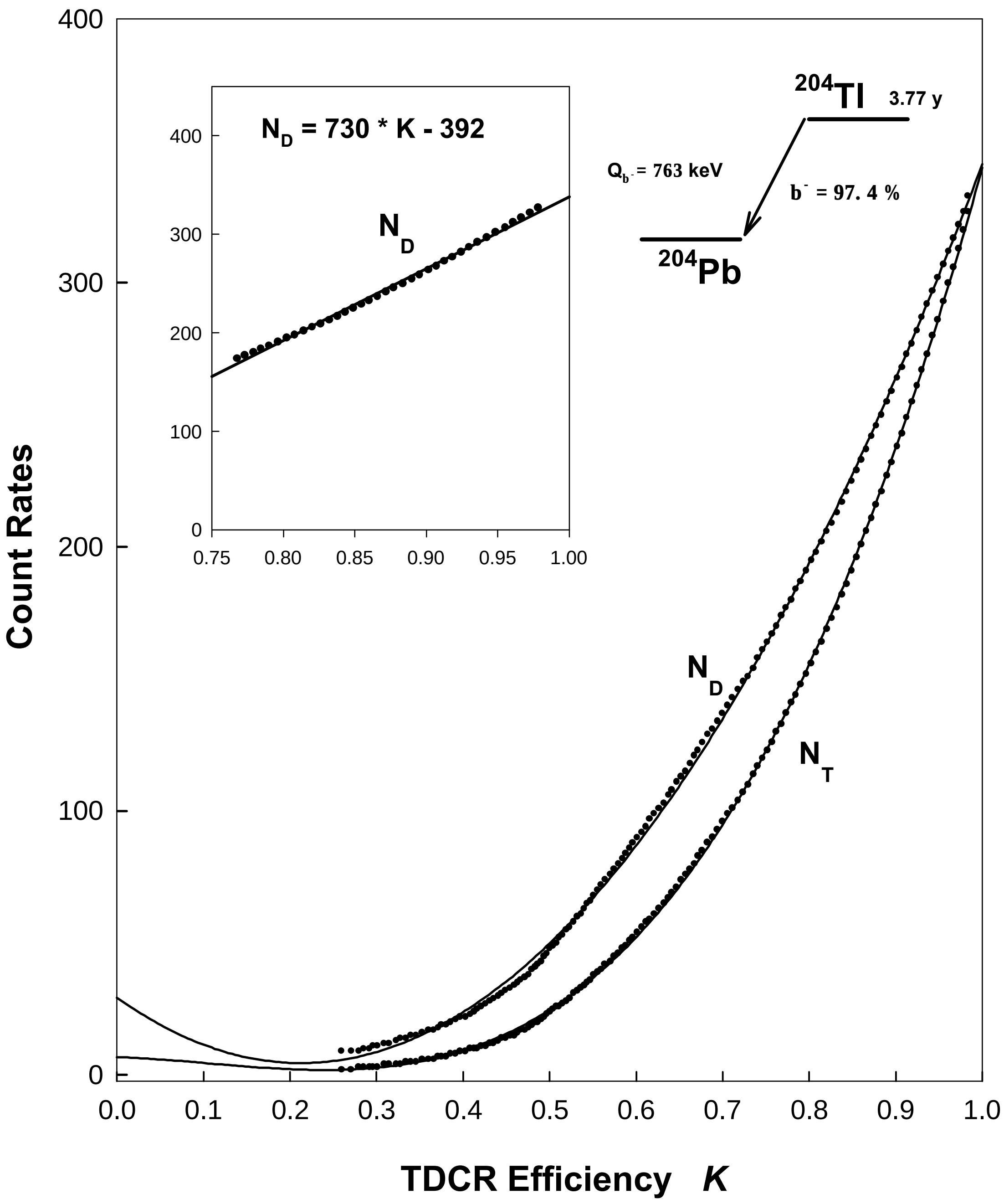


Fig. 7 Plots of $N_D(K)$ and $N_T(K)$ versus K. The data points were extrapolated to K = 1 by the linear least-squares fit in the region of $K \geq$ 75 %.

channel time scaling technique at the same time with the computer discrimination method. The activity of ^{14}C and ^{204}Tl can be determined in a relatively short time with the new 3D-DAQ. Short data acquisition time together with the early noted fact that only a minimal electronics proceed digitization should imply results obtained with the present method is quite free of such systematic errors as those associated with instrumental drift, replication of experimental conditions and source decay.

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