Absolute Activity Measurement of a ^{137}Cs Solution by the Tracer Method

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추적자 방법에 의한 ^{137}Cs-용액의 방사능 절대측정

박태순 · 오필제 · 황선태

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Abstract

Using a solution of ^{134}Cs as a tracer, radioactivity of a ^{137}Cs solution is measured by coincidence and anticoincidence counting methods. Sample sources in this study are fabricated by mixing ^{137}Cs with tracer, and the mixing ratios are 1/3 and 1/5, respectively in coincidence and anticoincidence methods. Specific activities of a ^{137}Cs solution are calculated by means of β-channel low energy threshold level variation and efficiency extrapolation. The results obtained by coincidence and anticoincidence counting methods are 552.78 kBq/g and 554.32kBq/g, respectively on the reference date and the combined uncertainties are 1.60kBq/g and 1.51kBq/g for each method. These two results show a good agreement within the uncertainty range.

초   록

^{134}Cs-용액을 추적자로 사용하여 동시와 역동시 계수방법에 의해 ^{137}Cs-용액의 방사능을 측정하였다. 본 연구에서의 측정선원은 ^{137}Cs과 추적자를 혼합하여 제작하였으며 혼합 비율은 동시와 역동시 계수 방법에서 각각 1/3과 1/5로 하였다. ^{137}Cs-용액의 비 방사능은 β채널 저에너지 문턱 준위 변화와 효율외삽 방법에 의해 산출 되었다. 동시와 역동시 계수 방법에 의해 산출된 결과는 기준일 현재 552.78kBq/g과 554.32kBq/g이었으며 합성오차는 각각 1.60kBq/g과 1.51kBq/g이었다. 두 개의 결과는 오차범위 내에서 잘 일치함을 보여준다.

I. Introduction

The ^{137}Cs nuclide, which has a long half-life and a single γ-ray energy, is widely used as a standard source for detector calibration. It is also important in nuclear reactor engineering and environmental studies.

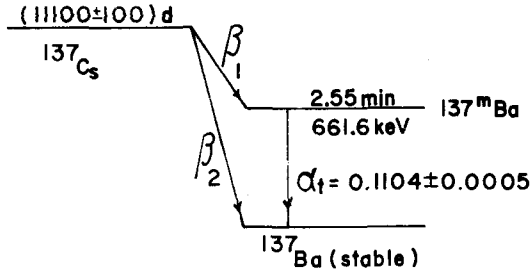Despite of its simple decay scheme, the absolute activity measurement of ^{137}Cs nuclide is not easy.

Fig. 1. Decay Scheme of ^{137}Cs.

As shown in Fig. 1, ^{137}Cs nuclide is transmuted to ^{137m}Ba nuclide by the β_1 decay and ^{137m}Ba nuclide becomes ^{137}Ba stable nuclide by the γ-ray transition in 2.55 minutes.

The activity of ^{137}Cs can not be measured by the conventional β-γ coincidence or anticoincidence methods because the time interval between β and γ events is 2.55 minutes for one disintegration. In order to solve this matter, a tracer nuclide was used in this experiment. The ^{134}Cs nuclide was chosen as a tracer considering its chemical and physical properties. The ^{137}Cs and ^{134}Cs mixed sources were measured by 4π β-γ coincidence and anticoincidence methods. The activity was determined from β-efficiency extrapolation by the low energy threshold level variation for both methods.

II. Principles

A 4π β-γ coincidence counting is one of the most appropriate methods for the absolute activity measurement.[1,2] This method needs only β, γ and coincidence counting rates, not requiring decay scheme parameters, to determine the activity. The basic equations in the coincidence method can be written as

$$N_\beta = N_0 \varepsilon_\beta, \quad N_\gamma = N_0 \varepsilon_\gamma, \quad N_c = N_0 \varepsilon_\beta \varepsilon_\gamma$$
$$N_0 = N_\beta N_\gamma / N_c \qquad (1)$$

where N_0, N_β, N_γ and N_c are disintegration rate of a measuring sample, β, γ and coincidence counting rates. ε_β and ε_γ are β and γ counting efficiencies, respectively. But eq. (1) can apply

only to a very simple decay scheme. In most cases, eq. (1) has to be modified for a complex decay scheme.

$$N_\beta = N_0 \sum a_r \left[\varepsilon_{\beta r} + (1 - \varepsilon_{\beta r}) \left(\frac{\alpha \varepsilon_{ce} + \varepsilon_{\beta \gamma}}{1 + \alpha} \right)_r \right]$$
$$N_\gamma = N_0 \sum a_r \varepsilon_{\gamma r} \qquad (2)$$
$$N_c = N_0 \sum a_r [\varepsilon_{\beta r} \varepsilon_{\gamma r} + (1 - \varepsilon_{\beta r}) \varepsilon_{cr}]$$

where a, ε_{ce}, $\varepsilon_{\beta\gamma}$ and ε_c are β-branching ratio, internal conversion electron counting efficiency, γ-ray sensitivity in β-detector and probability of γ-γ coincidence or compton scattering coincidence, respectively. The subscript r denotes r-th β-branching. For the experimental purpose, the detector used in the coincidence counting has to satisfy the following two conditions.

1 The r-th β-efficiency is defined by the s-th β-efficiency.

2 $\varepsilon_{\beta\gamma}$, ε_{ce}, ε_γ, ε_c are to be constants by experimental conditions and N_β approaches to N_0 as $N_c/N_\gamma \rightarrow 1$.

Eq. (2) can be simplified to,

$$N_\beta = N_0 F(N_c/N_\gamma) \qquad (3)$$

where F is the efficiency function of N_c/N_γ and generally has a polynomial form.

The anticoincidence counting method is similar to the coincidence method except being measured anticoincidence counting rate, N_Y instead of coincidence counting rate, N_c.[3]

It has an advantage that the method can be used to measure the activity of γ-ray delayed nuclide.

Experimental equations for the anticoincidence method can be written as the following forms:

$$N_\beta = N_0 F(N_c/N_\gamma), \qquad N_0 \text{ as } N_c/N_\gamma \rightarrow 1$$
$$= N_0 + f(1 - N_c/N_\gamma), \qquad N_0 \text{ as } N_c/N_\gamma \rightarrow 1$$
$$= N_0 + f(N_Y/N_\gamma), \qquad N_0 \text{ as } N_Y \rightarrow 0$$
$$= N_0 + g(N_Y), \qquad N_0 \text{ as } N_Y \rightarrow 0$$
$$\qquad (4)$$

where F, f, g are polynomial efficiency functions and N_Y denotes anticoincidence counting rate $(N_\gamma - N_c)$. From the results measured by the coincidence and anticoincidence methods, the

activity of ^{137}Cs can be expressed as:[4]

$$\frac{N_{137}D_{137}}{m_{137}}=A_{137}[\varepsilon_{137}+C] \qquad (5)$$

where A_{137}, N_{137}, D_{137}, m_{137} and ε_{137} denote activity concentration on the reference date, β-counting rate, decay correction factor, mass of sample source and β-counting efficiency for ^{137}Cs, respectively. And a constant C is the contribution of converson electrons and unconverted photons.

As the efficiency ε_{137} can not be measured directly, the tracer ^{134}Cs is added quantitatively to produce mixed sources. It is assumed that ε_{137} can be approximated as a function of the inefficiency, $1-\varepsilon_{134}$

$$\varepsilon_{137}=f(1-\varepsilon_{134})=f(1-N_c/N_r)$$
$$=1+C_1[1-N_c/N_r]+C_2[1-N_c/N_r]^2+\cdots$$

Similarly, the activity of ^{134}Cs is

$$N_{134}=\frac{m_{134}A_{134}}{D_{134}}g(1-\varepsilon_{134}) \qquad (6)$$

where $g(1-\varepsilon_{134})=1+d_1[1-N_c/N_r]+d_2[1-N_c/N_r]^2+\cdots$. For a ^{137}Cs and ^{134}Cs mixed source, the counting rate of ^{137}Cs is

$$N_{137}=N_{tot}-N_{134} \qquad (7)$$

where N_{tot} is the total β-counting rate from a mixed source. Substitute eq. (5), (6) to eq. (7)

$$\frac{D_{137}}{m_{137}}\left[N_{tot}-\frac{m_{134}A_{134}}{D_{134}}g(1-N_c/N_r)\right]=A_{137}$$
$$x[f(1-N_c/N_r)+C] \qquad (8)$$

From polynomial extrapolation,

$$A_{137}(1+C)=\frac{D_{137}}{m_{137}}\left[N_{tot}-\frac{m_{134}A_{134}}{D_{134}}\right],$$
$$\text{as } N_c/N_r\rightarrow1 \qquad (9)$$

where N_{tot} and A_{134} can be measured from the coincidence or anticoincidence method, and m_{134} and m_{137} can be measured from weighing by the pycnometer method. Constant C and D_{137}, D_{134} are calculated using the related constants. Therefore, the activity of ^{137}Cs, A_{137} can be obtained directly from the efficiency extrapolation.

III. Sample Source Preparations

The ^{137}Cs solution used in this experiment was

supplied from Physikalisch-Technische Bundesanstalt (PTB) in Federal Republic of Germany and its chemical composition was 0.2 mol HCl per cm^3 with 20μg of CsCl per gram of solution. A tracer nuclide of ^{134}Cs has to satisfy the following three conditions: ① β-γ emitter with β-spectrum not too different from that of ^{137}Cs, ② uniform mixing ^{137}Cs with a tracer, ③ easy standardization. The nuclides which satisfy these conditions are ^{60}Co, ^{82}Br, ^{134}Cs, etc.

In the experiment, ^{134}Cs solution from PTB was chosen as a tracer considering its half-life and chemical properties. Its chemical composition was the same as ^{137}Cs solution.

The activity of ^{134}Cs tracer solution turned out to be 166.8$\pm$0.16kBq/g on the reference date (1-May-1986, 0 h UT) from results of international intercomparison.[5] Impurity check for each nuclide was carried out by the γ-ray spectrometry. No γ-ray emitting impurities exceeding 5×10^{-5} of the main activity could be identified for ^{134}Cs solution. The relative content of ^{134}Cs in the ^{137}Cs solution was about 4×10^{-5} to ^{137}Cs solution.

The source mounting has to be fabricated as thinly as possible to eliminate back scattering and foil absorption.

The source mounting was carried out using collodion thin film supported by aluminum ring.
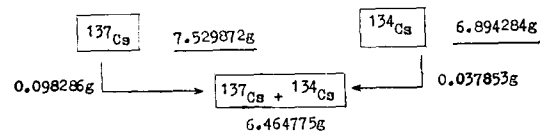
The thin film has chemically stable and suitable mechanical strength. Its thickness is about 13 μg/cm^2.

Also, the thin film used in a 4π proportional counter has to be electrical conduction material to prevent the charging-up effect which cause an electric field distortion.

Collodion thin film was coated with gold(Au) in the vacuum evaporator.

The thickness of a gold-coated thin film was about 30μg/cm^2.

The measuring solution was prepared by mixing ^{137}Cs with ^{134}Cs solution. The mixing

Table 1. Preparation of ^{137}Cs and ^{134}Cs Mixed Sources.

$$\boxed{^{137}\text{Cs}} \xrightarrow{7.529872g} \qquad \boxed{^{134}\text{Cs}} \quad 6.894284g$$

$$0.098286g \xrightarrow{\quad\quad} \boxed{^{137}\text{Cs} + ^{134}\text{Cs}} \xleftarrow{\quad\quad} 0.037853g$$

$$6.464775g$$

m_{obs} (g)	Δm_{obs} (mg)	Δm_{corr} (mg)
6.464775		
1. 6.445785	18.990	19.009
2. 6.426749	19.036	19.055
3. 6.406308	20.441	20.462
4. 6.385786	20.522	20.543
5. 6.369130	16.656	16.673
6. 6.356300	12.830	12.843
7. 6.339070	17.230	17.247

Before sampling: $T=23.5°C$ $RH=61\%$
$P=752.4$mmHg

After sampling : $T=24.1°C$ $RH=58\%$
$P=751.6$mmHg

ratios were 1/3 and 1/5, respectively in coincidence and anticoincidence methods. The mixed solution was dropped on the surface of goldcoated thin film. Its mass on the surface was measured to 10^{-6} g range by the gravimetric method.
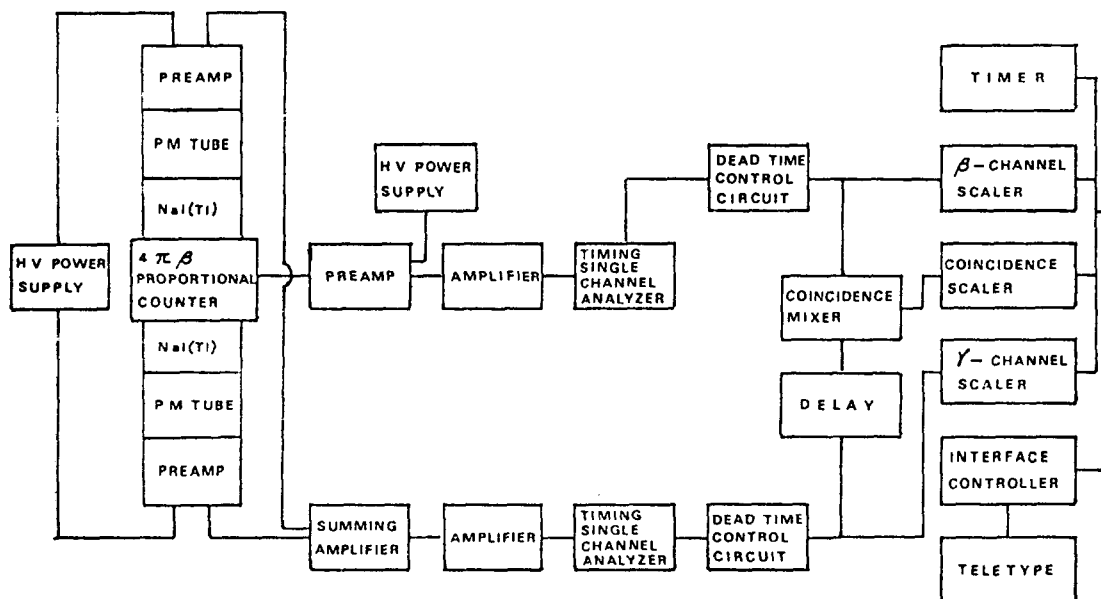
The mass data was obtained from the buoyancy correction for temperature, relative humidity and pressure. The wetting agent was used for uniform deposition of the mixed solution on the surface. The wetting agent in the experiment was Ludox SM-15 diluted with distilled water by a factor of 10^4. The prepared mixed sources were dryed in the fume hood for about one day in the natural atmosphere. Mixed sources which were used for measurements were 7 ones per each mixing ratio.

Table 1 illustrates the dilution method and shows mass data for mixed sources diluted by 1/3.

IV. Measurement

(a) Detectors

β-detector is a 4π pressurized proportional counter using P-10 gas. The 4π proportional counter of pill-box type has sensitive volume of 57.84cm^3. Its wall material is aluminum. This counter can maintain up to 7.5 MPa of the counting gas pressure and is insulated between detector wall and anode wire by teflon. Also,

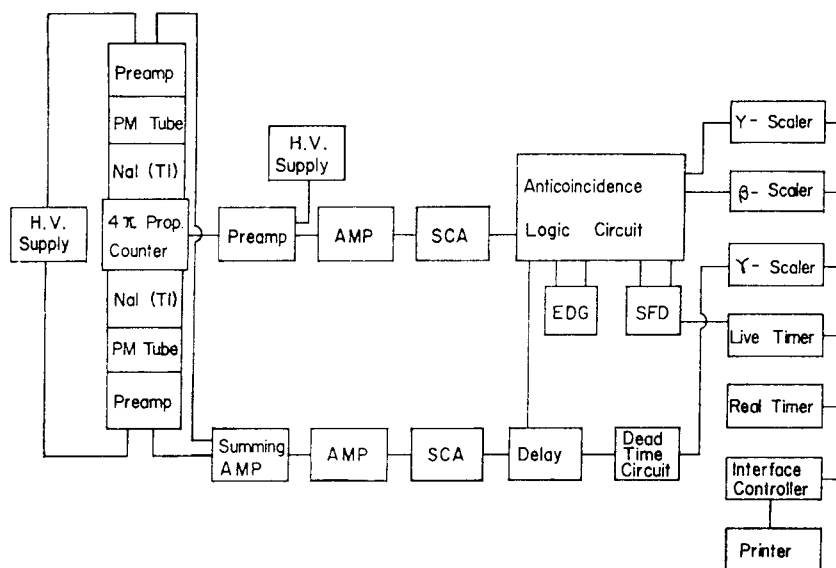Fig. 2. Block Diagram of 4π β-γ Coincidence Counting System.

Fig. 3. Block Diagram of 4π β-γ **Anticoincidenc Counting System.**

each side of the slide which is used to insert a sample source is equipped with a quad-ring to prevent the leakage of counting gas. In this experiment, the pressure of counting gas was determined to be 1.31 MPa considering the insulation of high voltage shunt circuit and capacity of electric components. The high voltage applied at anode wire was determined to be 5.5 kV from the plateau curve.

Counting gas pressure system was fabricated to be controlled automatically within 350 Pa at 5cm³ flow rate by the differential pressure transmitter and the electrically motorized valve.[6]

The γ-rays are measured by two $3'' \times 3''$ NaI (Tl) scintillation detectors which are positioned over and under the 4π proportional counter to enhance γ-ray efficiency. The applied voltages of NaI(Tl) scintillation detectors were determined to be 908 V and 950 V, respectively to adjust the γ-ray energy gain.

(b) Methods

In coincidence counting method, the measurement system consists of β-, γ-and coincidence channels as Fig. 2.

This method is used to measure β and γ

counting rates, and coincidence counting rate to record input pulses of β-, γ-channels within the resolving time.

For the dead time correction, a nonextendable dead time control circuit was inserted between timing single channel analyzer and coincidence mixer for β-and γ-channels, respectively. The dead time was determined to be 3.03μs for β-channel and 3.06μs for γ-channel, respectively considring the dead times of counting components, the resolving time of coincidence channel was determined to be 1.02μs to minimize accidental coincidence and true coincidence loss.

In the anticoincidence counting method, the measurement system is similar to the coincidence counting system except using a live-timed anticoincidence logic circuit instead of a coincidence mixer as shown in Fig. 3.

This method is used to measure β,γ and anticoincidence counting rates, where anticoincidence counting rate, N_Y is equal to $N_\gamma - N_c$. As the live time clock which is contained is anticoincidence logic circuit does not operate during the dead time generated from the extending dead time generator. In the coincidence and anticoin-
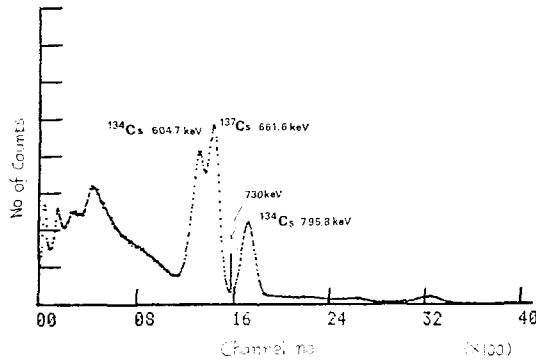
Fig. 4. γ-Ray Spectrum of ^{137}Cs and ^{134}Cs Mixed Source.

cidence methods, the low energy threshold level variation method was used for the efficiency variation in β-channel.

7 through 10 data points were obtained for one sample source.[7,8]

The measuring time for one point was 400s for coincidence and 500s for anticoincidence. The window setting of γ-channel was determined to be 730 keV to 890 keV to exclude the ^{137}Cs γ-ray peak from the γ-ray energy spectrum of ^{137}Cs and ^{134}Cs mixture source. Fig. 4 shows a γ-ray energy spectrum of ^{134}Cs and ^{137}Cs mixed

source.

The ranges of efficiency parameters for the extrapolation method appeared to be 70% to 83% for N_c/N_γ and 70% to 85% for $(N_\gamma-N_c)/N_\gamma$ in the coincidence and anticoincidence methods. Table 2 shows the countng data for mixed sources.

V. Results and Analysis

The counting data were corrected for decay time, and background counting rate in the coincidence and anticoincidence counting methods. The dead time correction was added in the coincidence counting method.

The activities of ^{137}Cs and ^{134}Cs mixture sources were calculated by the extrapolation from β-efficiency at each threshold level. Thus the activity of ^{137}Cs solution can be calculated using the previous eq. (9). That is,

$$A_{137}(1+C)=\frac{D_{137}}{m_{137}}\left[N_{tot}-\frac{m_{134}A_{134}}{D_{134}}\right]$$

as N_c/N_γ or $(N_\gamma-N_Y/N_r)\rightarrow1$

The right side of this equation can be determined

Table 2. Counting Data for ^{137}Cs and ^{134}Cs Mixed Sources in Coincidence and Anticoincidence Methods.

	Coincidence counting	Anticoin. counting
$\circ$ γ-channel window limits (keV)	730~890	730~890
$\circ$ Typical count rates(s^{-1})		
β	5000	6000
γ	135	130
c & anti-c	90~150	20~40
$\circ$ Background rates(s^{-1})		
β	1.5	1.5
γ	3.5	3.5
c & anti-c	0.08	3.5
$\circ$ No. of data points	7~10	7~10
$\circ$ Range of efficiency parameter(%)	70~83	70~85
N_c/N_γ & $\frac{N_\gamma-N_Y}{N_\gamma}$		
$\circ$ Method used for varying efficiency	threshold level variation	threshold level variation
$\circ$ Mean measurement time (s^{-1}) per one point.	500	400
$\circ$ Time of the measurement	21-May-1986 to 24-May-1986	17-June-1986 to 19-June-1986

Table 3. Uncertainty Components of the Final Result (in %)

due to	coin.	anti coin.	How was it obtained?
○ Counting statistics	0.21	0.18	standard error of mean of 5 values
○ Weighing	0.03	0.03	$\Delta m/\bar{m}$
○ Dead times	0.02	—	estimated from measured uncertainty
○ Resolving time	0.02	—	estimated from measured uncertainty
○ Fitting procedure	0.12	0.12	estimated from limits of least square fit
○ Background	0.01	0.01	$\Delta B_r/N_r$
○ Timing	0.002	0.002	crystal oscillator inaccuracy
○ α_t of Ba and branching ratio of ^{137}Cs	0.072	0.072	calculated using stated limits
○ 662 keV photons detected in 4π counter	0.02	0.02	calculated using estimated limit of measurent
○ 662 keV photons detected in γ channel	0.08	0.08	calculated using estimated limit of measurement
○ Half-lives of ^{137}Cs and tracer	0.001	0.001	calculated using stated limits
○ Tracer activity	0.12	0.12	calculated from previous measurement
○ Combined uncertainty	0.290	0.272	Square root of summed squares

by the experimental data. However the constant C is expressed by internal conversion coefficient (α), β-decay probability (b), β-detector efficiency for the internal conversion electrons (ε_{ce}) and γ-ray sensitivity in the β-detector ($\varepsilon_{\beta\gamma}$) as shown in the following equation.

$$C = \frac{b}{1+\alpha}(\alpha\varepsilon_{ce}+\varepsilon_{\beta\gamma}) \tag{10}$$

where

$b=0.9461\pm0.0023,\quad \alpha=0.1104\pm0.0005$

$\varepsilon_{ce}\simeq1,\quad \varepsilon_{\beta\gamma}\simeq0.05\sim0.54\%$ (for proportional counter)

The constant C value used in this experiment was 0.0987.[4]

The specific activity of ^{137}Cs solution was 552.78kBq/g in the coincidence counting method and 554.32kBq/g in the anticoincidence counting method on the reference date (10-May-1986, 0 h UT).

The combined uncertainty was obtained by the square root of summed squares of each component uncertainties.

Table 3 shows components, uncertainty of each component and evaluation methods.

Among the uncertainty components, the uncertainty from counting statistics was the first significant portion and one due to the fitting procedure was the second in the both counting

methods. The final combined uncertainty was 0.29% for the coincidence counting method and 0.27% for the anticoincidence counting method.

VI. Conclusions

Activity of a ^{137}Cs solution was measured by the coincidence and anticoincidence counting methods. The activity obtained by the tracer method was 552.78kBq/g and 554.32kBq/g on the reference date (01-May-1986; 0 h UT), respectively in the both counting methods and the combined uncertainty was 1.60kBq/g and 1.51 kBq/g, respectively. Two results show a good agreement each other as 0.34% of difference within the uncertainty range. From this experiment, an effect due to any change of the mixing ratio of ^{137}Cs to ^{134}Cs solution does not appear in the activity measurements. And it is clarified that the anticoincidence counting data is more accurate than the coincidence counting data in uncertainty.

But in order to obtain the better accuracy, it is necessary to improve the the efficiency of 4π proportional counter up to above 90%.

ations of Lee, Hyun Ku in the sample source preparations as a trainee from Chungnam National University and assistances of Lee, Min Kie in the electronic circuit fabrications as an electronic technician.

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