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■ 論 文 ■

Absolute ^{56}Mn Activity Measurement by 4π β–γ Conincidence Counting Technique

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• Abstract

In order to determine the ^{56}Mn γ-detection efficiency of a $MnSO_4$ bath system, it is essential to do the absolute activity measurement of ^{56}Mn solution. For the fabrication of ^{56}Mn samples, a 13.718 mg of ^{55}Mn metal flake with 99.99% purity was irradiated for 12 minutes at the thermal neutron field of about 10^{13} n/cm^{2}s of flux density. The neutron activated ^{56}Mn metal sample was dissolved in 50 ml of 0.1 N-HCl solution. The ^{56}Mn samples were fabricated by using the dissolved stock solution and the activity of each of them was measured by the 4π β-γ coincidence counting technique. The obtained result was 408.070 kBq/mg with total uncertainty of 0.366% at reference date, 0 h on October 15, 1987.

1. INTRODUCTION

The 4π β-γ coincidence counting technique has been in use over several decades for very accurate measurement of the disintegration rate of a radionuclide. With particular reference to the 4 π β-γ coincidence counting system, the detailed discussion has been given by P.J. Campion [1] and the principles of the coincidence method as well as the nature of the corrections required are now well understood. However, a valid application of the principles to the actual measurement is not simple. In particular, for complex decay of a radionuclide exceptional care is necessary in the selection of β–detector system and experimental technique.

In this study, a 4π pressurized proportional counter(PPC) is used for simplifying the complex decay scheme correction when β–detection efficiencies for the different β–groups of the decay

scheme are linearly inter-related. The 4π PPC is a β–detector with zero intrinsic noise, low sensitivity to γ-rays and a recovery time of the order of 10^{-7}s [2]. In the 4π β-γ coincidence measurements, the absolute disintegration rate of a radionuclide is deduced from the efficiency function by linear extrapolation of β– detection efficiencies on the basis of low energy threshold level variation. In this application, the 4π PPC is operated at 1.02 MPa (150 psi) of the counting gas pressure. At higher pressures, the property of true proportionality may be added. This offers significant advantages [3] in meeting some of the experimental requirements for activity measurements involving complex decay.

2. PRINCIPLES

The characteristic advantage of the coincidence technique lies in the fact that the result is, in principle, independent of the efficiency of either detector. Thus the fundamental problem of 4π β counting, namely the self absorption correction, can be avoided.

In the 4π β-γ coincidence counting technique, the basic equations for a point source of the disintegration N_o can be written by

$$\left.\begin{aligned} N_\beta &= N_o\varepsilon_\beta \\ N_\gamma &= N_o\varepsilon_\gamma \\ N_c &= N_o\varepsilon_\beta\varepsilon_\gamma \end{aligned}\right\} \quad N_o=\frac{N_\beta\cdot N_\gamma}{N_c} \qquad (1)$$

where N_β, N_γ and N_c are the observed counting rates less background and accidental coincidences in the β–, γ– and coincidence channels, respectively, ε_β and ε_γ the overall efficiencies of the β– and γ– detectors. However, eqs.(1) can be applied only to a simple decay scheme. When the decay scheme involves complex β– and γ– branching, the expressions for the individual channel counting rates become correspondingly more elaborate. The required modifications to basic equations have been discussed by A.P. Baerg [4,5]. For the coincidence measurements on a radionuclide with a decay scheme involving n different β–branches, the observed counting rates for the three channels are given by

$$\begin{aligned} N_\beta &= N_o\sum a_r\left(\varepsilon_{\beta r}+(1-\varepsilon_{\beta r})\left(\frac{\alpha\varepsilon_{ce}+\varepsilon_{\beta\gamma}}{1+\alpha}\right)_r\right) \\ N_\gamma &= N_o\sum a_r\frac{\varepsilon_{\gamma r}}{1+\alpha_r} \qquad (2) \\ N_c &= N_o\sum a_r\left(\frac{\varepsilon_{\beta r}+\varepsilon_{\gamma r}}{1+\alpha_r}+(1-\varepsilon_{\beta r})\varepsilon_{cr}\right) \end{aligned}$$

where the summations extend over the n β–branches and thus for the r-th β–branch ;

a_r = fractional β–intensity,
$\varepsilon_{\beta r}$ = β–detection efficiency,
α_r = total internal conversion coefficient with this β–branch,
$(\varepsilon_{ce})_r$ = β–detector efficiency for conversion electrons,
$\varepsilon_{\gamma r}$ = γ–detection efficiency,
$(\varepsilon_{\beta\gamma})_r$ = β–detection efficiency for γ–rays,
ε_{cr} = probability of recording a coincidence when the associated β–particle is not detected.

In the more general case, it is necessary to choose detectors and experimental conditions to provide additional information to solve eqs.(2). Theoretically a set of equations describing experimental conditions which can be closely approximated in practice and which permit a solution for N_o derived from the fact that all β-branches are subjected to the same physical variables responsible for counting losses. Therefore, the efficien-

cies ε_i can be taken as functionally related as follows :

$$\begin{aligned}
\varepsilon_{\beta r} &= 1-g_r(1-\varepsilon_{\beta s}) \to 1 \\
\varepsilon_{ce} &= 1-g_{ce}(1-\varepsilon_{\beta s}) \to 1 \\
\varepsilon_{\beta\gamma} &= \varepsilon^0_{\beta\gamma}-g_{\beta\gamma}(1-\varepsilon_{\beta s}) \to \varepsilon^0_{\beta\gamma} \\
\varepsilon_{cr} &= \varepsilon^0_{cr}-g_{cr}(1-\varepsilon_{\beta s}) \to \varepsilon^0_{cr}
\end{aligned} \qquad (3)$$

as $\varepsilon_{\beta s}$ approaches to 1.

where $\varepsilon_{\beta s}$ is the β–detection efficiency for any one arbitrarily selected β–group, while g_i are themselves to be regarded as single valued functions in $\varepsilon_{\beta s}$. Using the first of eqs. (3) defining the $\varepsilon_{\beta r}$, eqs. (2) reduce to

$$N_\beta = N_0\left[1-G\left(1-\frac{N_c}{N_\gamma}\right)\right] \cdots\cdots (4)$$

where $N_\beta \to N_0$ as $\frac{N_c}{N_\gamma}$ approaches to 1, and

$$G=\frac{\sum a_r[1-\{(\alpha\varepsilon_{ce}+\varepsilon_{\beta r})/(1+\alpha)\}]_r g_r}{\sum a_r[\varepsilon_\gamma/(1+\alpha)]-\varepsilon_c]_r g_r}\sum\frac{a_r\varepsilon_{\gamma r}}{1+\alpha_r}$$

Thus a plot of N_β vs. $(1-\frac{N_c}{N_\gamma})$ yields a straight line with the slope GN_0 and intercept N_0 for $\frac{N_c}{N_\gamma} \to 1$.

If G may be assumed constant when the $\varepsilon_{\beta r}$ were linearly inter-related, then N_0 may be obtained by linear extrapolation of appropriate efficiency function data. Substituting the remaining condition eqs. (3) into eqs. (2) as well, it follows that

$$N_\beta = N_0 F\left(\frac{N_c}{N_\gamma}\right) \cdots\cdots (5)$$

where the efficiency function F is usually defined simply as a polynomial in $\frac{N_c}{N_\gamma}$. As $\frac{N_c}{N_\gamma}$ approaches to 1, $F \to 1$ and thus $N_\beta \to N_0$.

The significant feature of these coincidence equations is that the disintegration rate of a ^{56}Mn measuring source can be obtained from the observed counting rates without requiring accurate information about the decay scheme parmeters by the β–efficiency extrapolation.

3. EXPERIMENTAL

(1) Dead Time and Resolving Time Corrections

Before the coincidence equations are used to deduce the disintegration rate N_0, the observed counting rates were corrected for dead time losses and accidental coincidences. The β–, γ– and coincidence channel count rates were corrected for dead time losses in a simple way by using the simple way by using the equations

$$N'_\beta = \frac{N''_\beta}{1-N''_\beta\tau}\ ;\ N'_\gamma = \frac{N''_\gamma}{1-N''_\gamma\tau}\ ;\ N'_c = \frac{N''_c}{1-N''_c\tau}$$

Where τ is the value of the imposed dead time, 3.02μs. N''_β, N''_γ, and N''_c denote the observed counting rates. Then N'_β, and N'_γ and N'_c are the dead time corrected rates. To obtain the required values of N_β and N_γ, the background count rates were similarily corrected. The corrected ratio of coincidence to γ–channel counting rates was obtained from J. Bryant's expression [6]. The formula used for calculating the data points of efficiency function is given by the equations

$$\frac{N_c}{N_\gamma} = \frac{(N'_c/N'_\gamma) - (b'_c/b'_\gamma)(b''_\gamma/N''_\gamma)(1-N''_\gamma\tau)/(1-b''_\gamma\tau)}{1-(b''_\gamma/N''_\gamma)(1-N''_\gamma/\tau)/(1-b''_\gamma\tau)}$$

$$\approx \frac{(N'_c/N'_\gamma) - (b''_c/N''_\gamma)}{1-(b''_\gamma/N''_\gamma)}$$

where $\frac{N'_c}{N'_\gamma} =$

$$\frac{(N''_c - 2\tau_r N''_\beta N''_\gamma)[1-(N''_\beta+N''_\gamma)\tau/2]}{N''_\gamma(1-N''_\beta\tau)[1-(N''_\beta+N''_\gamma-2N''_c)\tau/2-\tau_r(N''_\beta+N''_\gamma)]}$$

and $\frac{b'_c}{b'_\gamma} =$

$$\frac{(b''_c - 2\tau_r b''_\beta b''_\gamma)[1-(b''_\beta+b''_\gamma)\tau/2]}{b''_\gamma(1-b''_\beta\tau)[1-(b''_\beta+b''_\gamma-2b''_c)\tau/2-\tau_r(b''_\beta+b''_\gamma)]}$$

b''_β, b''_γ and b''_c denote the obsered background count rates. τ_r is the value of the resolving time, 1.085 μs.

The approximated expression,

$\frac{N_c}{N_\gamma} \approx \frac{(N'_c/N'_\gamma) - (b''_c/N''_\gamma)}{1-(b''_\gamma/N''_\gamma)}$, would be normally quite acceptable.

(2) Samples Preparation

The 13.718mg of a ^{55}Mn metal flake with 99.99% purity was irradiated in the thermal neutron field of about 10^{13}n/cm^2·s of flux density for 12 minutes in TRIGA MARK-III research reactor at the Korea Advanced Energy Research Institute in Seoul. This neutron activated ^{56}Mn metal flake was dissolved in 50ml of 0.1 N-HCI solution in order to prepare the ^{56}Mn stock solution at the Korea Standards Research Institute in about 5 hours after the neutron irradiation.

As shown in Fig. 1, the ^{56}Mn samples have to be fabricated as thinly as possible to minimize the back scattering and source absorption effects. The sample preparation was carried out using collodion thin film, about 13μg/cm of thickness, supported by aluminum(Al) ring. The thin film that is chemically stable and mechanically strong must have electrical conductivity to prevent a charging-up effect which causes an electric field distortion. Thus the collodion film was coated with gold (Au) in the vacuum evaporator(VECCO V-300). The thickness of the gold foil is about 30 μg/cm^2.

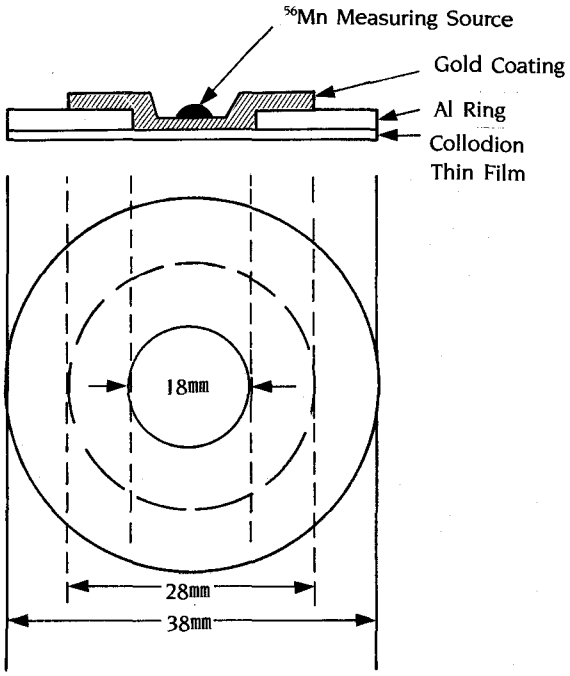

Fig. 1. Schematic diagram of ^{56}Mn sample

The ^{56}Mn stock solution of the order of 20mg was dropped on the surface of gold foil and its mass measured up to the 10^{-6}g range by the gravimetric method. Table 1 illustrates the mass data for ^{56}Mn samples.

The mass data was obtained from the buoyancy correction for temperature, pressure and relative humidity. The wetting agent (Ludox SM-15) diluted with distilled water by a factor of 10^4 was

Table 1. Mass correction of ^{56}Mn measuring sources

Sample No.	m_{obs}(g)	$\triangle m_{obs}$(mg)	$\triangle m_{corr}$(mg)
1	7.894960	29.817	29.853
2	7.875610	19.350	19.373
3	7.855742	19.868	19.892
4	7.835686	20.056	20.080
5	7.814045	21.641	21.667

P=760.2mmHg, T=22.3℃, R.H=58%

used for uniform deposition of the measuring source on the surface of gold foil. The prepared samples were dried under the infrared lamp for an hour.

(3) Detection and Mesurement

As shown in Fig. 2, the decay scheme of ^{56}Mn is relatively complex [7]. A 4π pressurized propo-

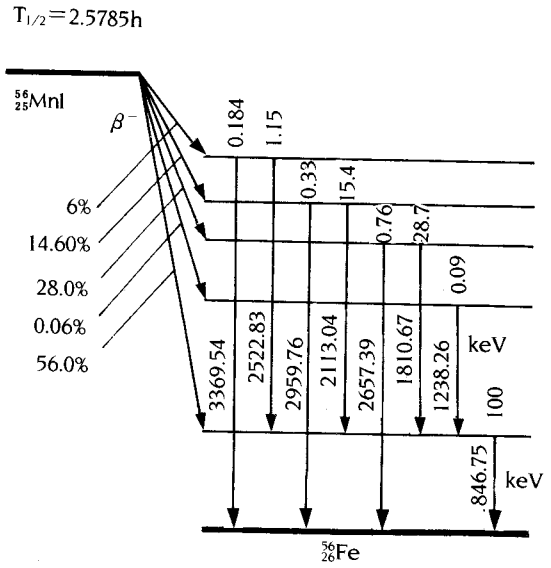

Fig. 2. Decay scheme of ^{56}Mn

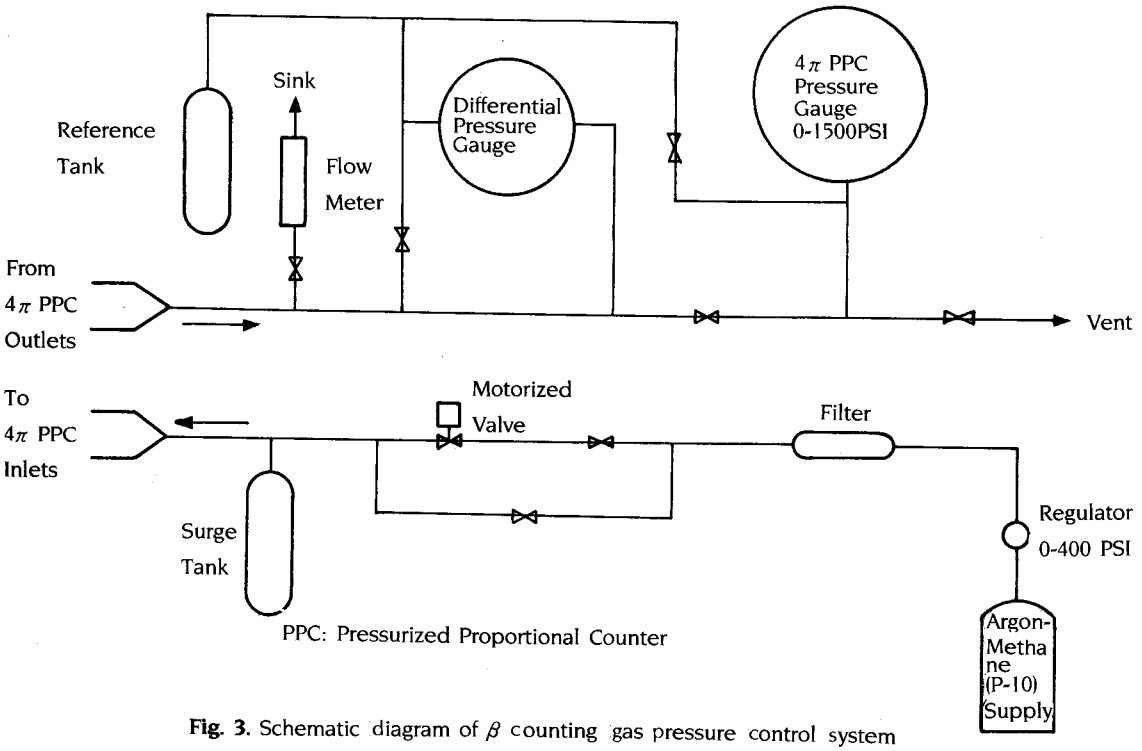

Fig. 3. Schematic diagram of β counting gas pressure control system

rtional counter(PPC) is used for β–detection. The P-10 is selected as a counting gas through the counter. The 4π PPC of Al pill-box type has the sensitive volume of 57.84cm^3. This counter is insulated by the teflon plate between detector wall and anode wire. Each side of the Al-slide that is used for the ^{56}Mn sample mount is equipped with a quad-ring to prevent the counting gas leakage. In this study, the gas pressure was set to be 1.02 MPa (150 psi) considering the insulation of high voltage shunt circuit and capacity of electronic components. The high voltage applied to the anode wire set to be 5500 V selected from the β–plateau curve. As illustrated in Fig. 3, the β counting gas system was fabricated to be controlled automatically by the differential pressure transmitter and the electrically motorized valve [8].

Two NaI(Tl) scintillation detectors, 3″φ ×3″, are usd for γ–detection. The applied voltages to these detectors were set to be 852.6 V for top one and 920 V for bottom one, respectively, to adjust the γ–ray energy gain. Fig. 4 shows the detection and measurement system for Mn activity determination by the 4π β-γ coincidence counting technique. This technique was introduced to measure β–, γ– and coincidence counting rates to record the input pulses of β– and γ– channels within the resolving time. For the dead time correction. a nonextendable dead time control circuit was inserted between timing single channel anal-

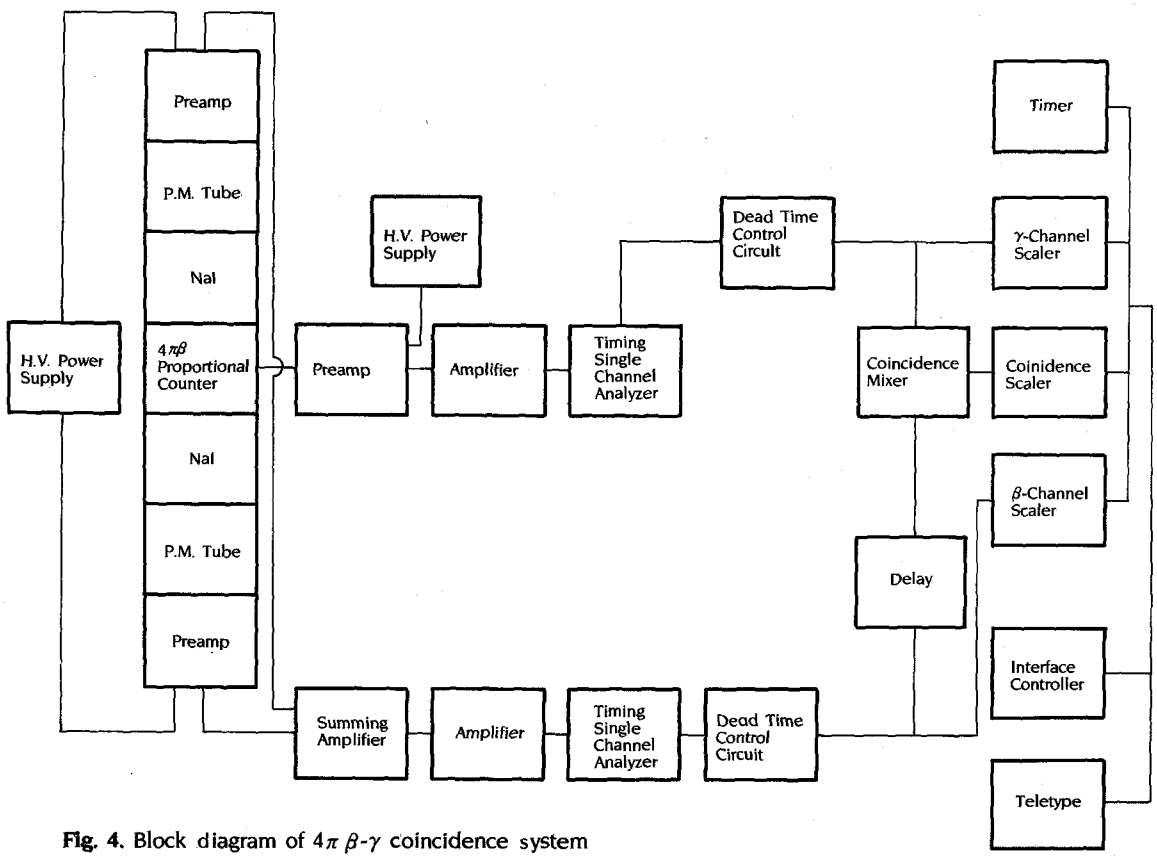

Fig. 4. Block diagram of 4π β-γ coincidence system

yzer and coincidence mixer for β–and γ–channels, respectively. The resolving time of coincidence channel was determined to be 1.085 μs to minimize accidental coincidence and true coincidence loss. In this study, the low energy threshold level variation method was used for the efficiency variation in β–channel. Thus, 11 data points were determined for one measuring sample and the measuring time was 300 s for each data point.

4. RESULTS AND ANALYSIS

The counting data were corrected for decay time, dead time and backgrounds in the coincidence method. The specific activities of ^{56}Mn measuring sources were calculated by the linear extrapolation from the β–efficiency at each threshold level. Thus the activity of a ^{56}Mn solution can be calculated using the eq. (4). Fig. 5. illustrates a graph of a fitted efficiency function and of the residuals. Each point corresponds to the weighted mean A of a series of measurements. The uncertainty bars represent the ranges of efficiency ±1 s (sample standard deviation).

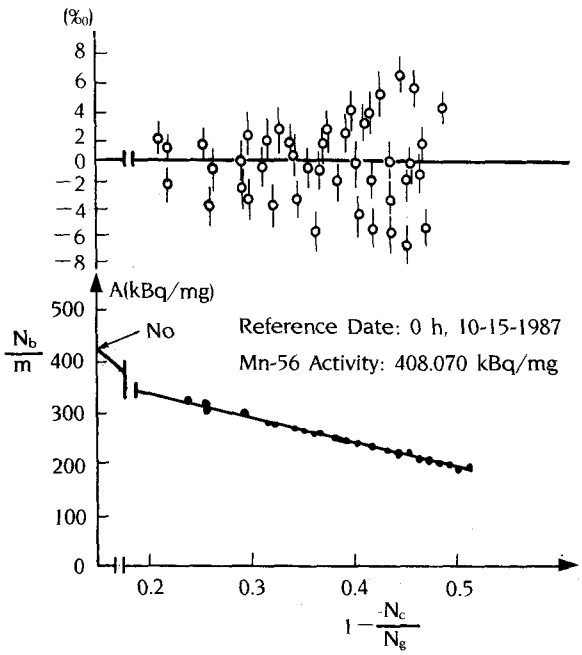

Fig. 5. Fitted efficiency function and residuals

The ranges of efficiency parameters for the extrapolation method appeared to be 52% to 72% for $\frac{N_c}{N_\gamma}$. Table 2 shows the counting data for the ^{56}Mn measuring sources.

The combined uncertainty was obtained by the quadrature propagation method. Table 3 shows the uncertainty components of final result in %.

Among the uncertainty components, the uncertainty from the extrapolation was the first signnificant portion due to the fitting procedure. The final combined uncertainty was 0.336%.

5. CONCLUSIONS

The activity of a ^{56}Mn solution originally prepared using the neutron activated ^{56}Mn metal sample was measured by the coincidence counting technique. The activity obtained from the linear extrapolation(Fig. 5) was 408.070 kBq/mg as the value of specific activity with total uncertainty of 0.366% at reference date, 0 h on October 15, 1987. In this study, it was difficult to introduce the ^{56}Mn samples more than four of them to the experiment since ^{56}Mn nuclide has a half-life of 2.5785h and it takes about three hours to complete the data acquisition necessary for each sample.

From the viewpoint of KSRI experiences associated with the international comparison studies [9, 10] on absolute activity measurements, it

Table 2. Counting data for ^{56}Mn measuring sources

Classification	Coincidence method
1. G-channel setting (keV)	757−922
2. Background count rates (s^{-1})	
B-channel	10.12
G-channel	2.56
coincidence channel	0.007
3. Number of sources measured	4
4. Number of data points	44
5. Mean measurement time(s) for one date point	300
6. Range of $\frac{N_c}{N_\gamma}$(%)	52−79
7. Procedure used to vary $\frac{N_c}{N_\gamma}$	Threshold level variation
8. Slope to intercept ratio	-0.98 ± 0.0016
9. Final result	
(1) Intercept for $\frac{N_c}{N_\gamma}\rightarrow 1$ at reference date	408.070 kBq/㎎
(2) Standard error in the mean	0.149 kBq/㎎ (0.366%)
(3) Number of degrees of freedom	10
10. Time of the measurements (year-moth-day)	87-10-15 to 87-10-16

Table 3. Uncertainty components of final result

Component	Uncertainty (%)	Obtained method
1. Weighing	0.03	$\triangle m/\bar{m}$
2. Dead time	0.02	Estimated from measured uncertainty
3. Resolving time	0.02	Estimated from measured uncertainty
4. Background	0.05	$B\gamma/N\gamma$
5. Extrapolation	0.36	Estimated from limits of least squares
6. Timing	0.005	Crystal oscillator inaccuracy
7. Combined uncertainty	0.366	Quadrature combination of components

is convinced that the obtained result is accurate enough to use as an essential value for determination of the ^{56}Mn γ-detection efficiency of KSRI $MnSO_4$ bath system that is operated to carry out the strength measurements of radioactive neutron sources such as ^{252}Cf and ^{241}Am-Be. In order to obtain the better accuracy of result, however, it is highly recommended that the β-detection efficiency of 4π PPC be raised as high as possibly achievable in the β-γ coincidence counting system.

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$4\pi\beta$-γ 동시계수기술에 의한 ^{56}Mn방사능 절대측정

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• 요 약 •

황산망간 용액조장치의 ^{56}Mn γ 선 검출효율을 결정하는데 ^{56}Mn용액의 방사능을 절대측정하는 것은 필수적이다. ^{56}Mn시료를 제작하기 위하여 99.99%의 순도를 갖는 Mn금속조각 13.718mg되는 시료를 한국에너지연구소 TRIGA MARK-Ⅲ 원자로의 중성자선속이 약 10^{13}n/㎠·s되는 열중성자장에서 12분간 조사시켰다. 중성자 방사화된 ^{56}Mn금속시료를 0.1N-HCl 용액 50ml에 용해시켜서 ^{56}Mn시료를 제작하여 $4\pi\beta$-γ 동시계수기술로 방사능을 측정한 결과 불확도 0.366%를 갖는 값으로서 1987년 10월 15일 0시를 기준하여 408.070kBq/mg을 얻었다.