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(Technical Report)

The Development Study of A Manganese Sulphate Bath System*

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=Abstract=

In order to establish the national standards of neutron measurements, a manganese sulphate($MnSO_4$) bath system was developed under the IAEA technical support. This bath system was made up of a spherical s.s. 316 L bath, of 3.5 mm thick and of 125 cm internal diagmeter, filled with a manganese sulphate solution, a solution circulating system, and a ^{5}Mn γ-ray monitoring system.

The solution pumped from the bath was introduced into a Marinelli beaker-type monitor vessel which was equipped with two seperate detectors, 3.8 cmϕ×3.8 cm NaI (Tl) crystals. The performance of the system were tested using the neutron sources, ^{241}Am-Be and ^{252}Cf, mounted at the center of the bath. From the decay curve analysis of ^{56}Mn activity, neutron emission rate of ^{252}Cf by the comparative method was obtained to be 3.71×10^{7} n/s per 50 μg as of November 15, 1985.

I. Introduction

The manganese sulphate bath continues to be the preferred method for determining neutron source strength. The neutron source strength means the neutron emission rate which is the number of neutrons per second emitted from the source.

In development of the KSRI $MnSO_4$ bath system, the electronic components of ^{56}Mn γ-ray monitoring system were supplied from IAEA on the technical cooperation project (ROK/1/007). The bath material and components of the circulating system of $MnSO_4$ solution were purchased in the domestic market but the $MnSO_4\cdot H_2O$(purity~99%) was imported from Japan.

The performance test of the bath system was done using a 27 mCi ^{252}Cf source and a 100 mCi ^{241}Am-Be source.

The system analysis was carried out by the evaluation of the growth and decay curves of ^{56}Mn activity. The strength of ^{252}Cf was measured by the comparative method regarding the ^{241}Am-Be as a reference source.

II. Principle of the $MnSO_4$ Bath Method

The measurements of neutron emission rate are made by the $MnSO_4$ bath method. Figure 1 shows a schematic diagram of the principle of this method. The fast neutrons emitted from the neutron source positioned at the center of the bath filled with the manganese sulphate solution isotropically enter the bath,

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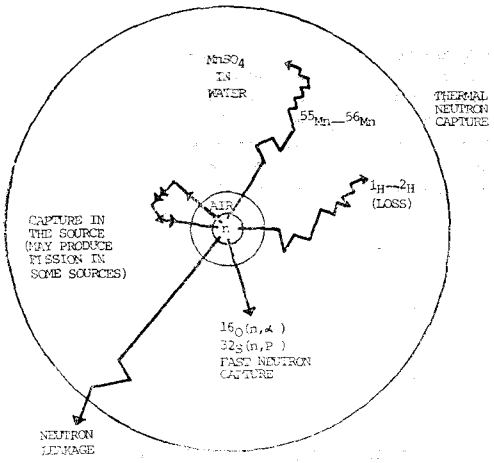

Fig. 1. Schematic Diagram of Principle of $MnSO_4$ Bath Method[1]

are slowed down by a series of collisions with pure water molecules, and finally captured by ^{55}Mn to produce radioactive ^{56}Mn nuclide.

From the measurement of the saturation activity of ^{56}Mn nuclides produced in the $MnSO_4$ solution, the emission rate of a neutron source, Q, can be determined using the following equaton:[2]

$$Q=\frac{A}{f\cdot\varepsilon\cdot(1-L)\cdot(1-S)\cdot(1-O)} \qquad (1)$$

where, A=the saturation ^{56}Mn activity counting rate,

f=the fraction of the neutrons captured by ^{55}Mn in the solution,

ε=the detection efficiency of the $MnSO_4$ system,

L=the fraction of neutrons which escape from the boundary of the $MnSO_4$ bath,

S=the fraction of thermal neutrons captured in the neutron source and source holder,

O=the fraction of fast neutrons undergoing (n, α) and (n, p) reactions in the solution.

The aforementioned method is well established for the absolute measurements of neutron emission rates. In this work, however, the ^{252}Cf source strength was determined by the comparative method using the following equation:[3]

$$Q_u=\frac{(C_n)_u\times(F_{a,l})_u\times Q_r}{(C_n)_r\times(F_{a,l})_r} \qquad (2)$$

where, Q_u=the unknown source strength,

$(C_n)_u$=the net count rate of an unknown source,

$(C_n)_r$=the net count rate of a reference source,

$(F_{a,l})_u$=the correction factor for absorption and leakage for an unknown source,

$(F_{a,l})_r$=the correction factor for absorption and leakage for a reference source,

Q_r=the source strength of a reference source.

According to Spiegel,[4] the corrections for $MnSO_4$ bath method are given as shown in Table 1 for the teflon n-source holder with a thickness of 1 cm. The two major uncertainties of the $MnSO_4$ bath method result from the thermal neutron absorption in ^{1}H and fast neutron absorption in ^{16}O and ^{32}S.[5] Figure 2 shows the schematic diagram of the manganese sulphate bath system used at KSRI.

The spherical bath tank was designed to have the rather large diameter of 125 cm to minimize the neutron leakage from the bath tank. While the neutron activated $MnSO_4$ solution is passing through the marinelli beaker-type monitor vessl, the ^{56}Mn γ activity is measured by the two NaI(T1) scintillation counters. Figure 3 shows a schematics of ^{56}Mn detection vessel.

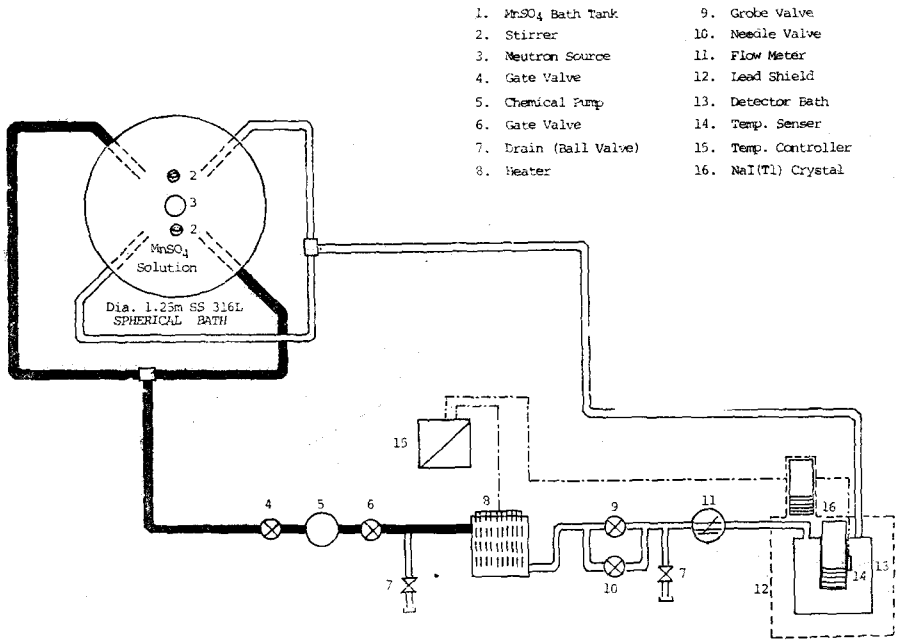

Fig. 2. Schematic Diagram of $MnSO_4$ Bath System

Table 1. Corrections for $MnSO_4$ Bath Method

Neutron Source / Correction Type	^{241}Am-Be(α,n) (±%)	^{252}Cf(f,n) (±%)
O&S fast neutron capture	2.88±0.5	0.63±0.1
Teflon fast neutron capture	0.39±0.1	0.11±0.05
Teflon thermal neutron capture	0.017±0.01	0.022±0.01
Fast leakage	0.23±0.2	0.03±0.1
Thermal leakage	0.028±0.02	0.02±0.01
Source self-absorption	0.0±0.01	0.01±0.01
Total Correction Factor	3.545±0.55	0.822±0.21

III. Experimental Arrangement

1. Neutron Source

The neutron sources of ^{241}Am-Be and ^{252}Cf (50μg) were used in this work. The neutron sources were of the doubly encapsulated

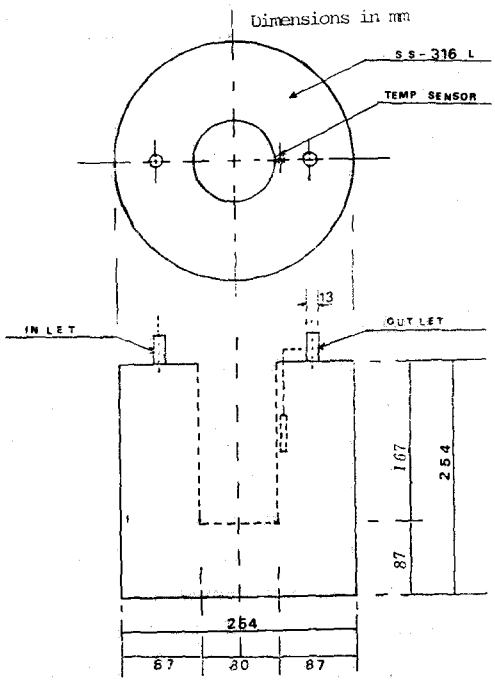

Fig. 3. Schematics of ^{56}Mn Detection Vessel

Table 2. Source Dimensions and Emission Data

Source	Dimensions(mm)			Emission Data		
	Diameter	Height	Thickness	Neutron emission	γ-exposure rate	Neutron dose rate
^{241}Am-Be	17.4	19.4	0.8	2.2×10^{6} n/s per Ci	2.5 mR/h at 1 m per Ci	2.2 mrem/h at 1 m per Ci
^{252}Cf	7.8	10	0.8	2.3×10^{9} n/s per mg	160 mR/h at 1 m per mg	2.3 rem/h at 1 m per mg

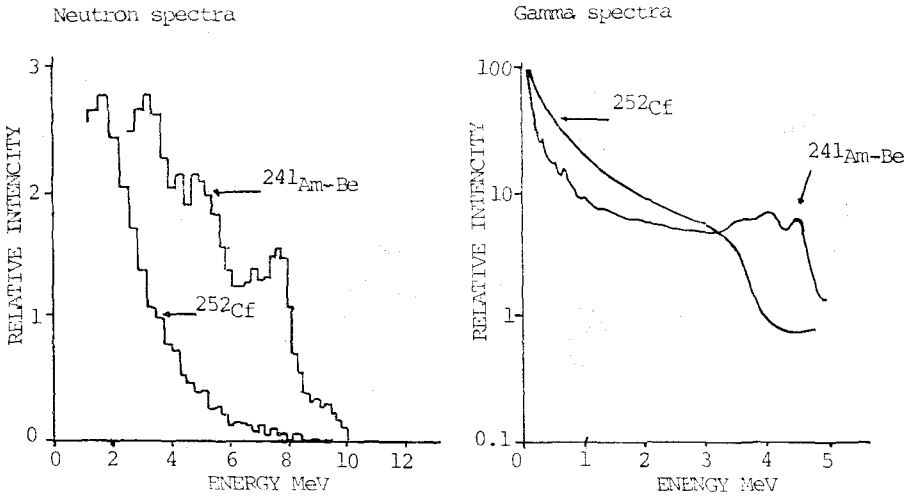

Fig. 4. Energy Spectrum of ^{241}Am-Be and ^{252}Cf[7]

variety in welded stainless steel capsules made and measured at the Radiochemical Center in Amersham UK using a stilbene crystal and pulse shape discrimination. Figure 4 shows the neutron and gamma spectra of the neutron sources. Details of the sources are given in Table 2[6].

2. Manganese Sulphate Bath

Figure 5 shows the $MnSO_4$ bath(Q-Ball). The manganese sulphate solution was totally enclosed. Introduction of the neutron source was accomplished by positioning a n-source holder made of teflon at the center of the bath tank. The bath was filled from the top by pumping the solution from the polyethylene storage tank. A uniform filling was achieved by mixing the solution using the two stirrers mounted at the levels of top and bottom of the bath, respectively. Before transferring the solutions from the storage tank to the bath tank, the full bath system was run first with the distilled water to find out any leakage. The reagent grade $MnSO_4\cdot H_2O$ and distilled water were used to prepare the manganese sulphate solutions. This is the typical grade which was used throughout the series of manganese bath measurements. It was assumed that the $MnSO_4\cdot H_2O$ was consistent in composition, with approximately 1% of impurities and that no strong neutron absorbers exist in the distilled water. In the experiment, the manganese concentration was 277.8 g of $MnSO_4\cdot H_2O$ per one liter of solution. The solutions pumped from the bath tank was circulated throughout the bath system. When the solution was mixed using a PVC rod, a fairly large amount of MnO_2

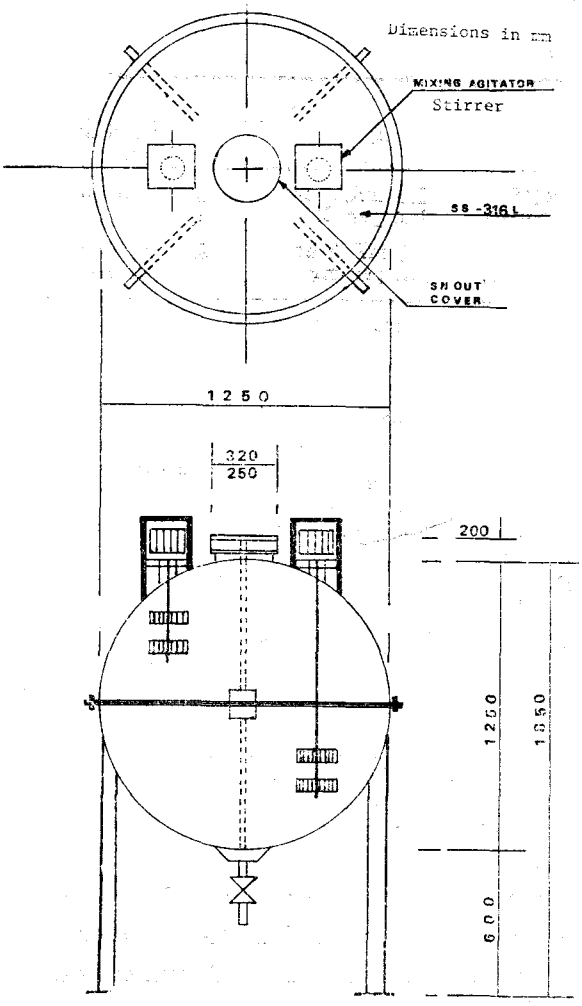

Fig. 5. KSRI $MnSO_4$ Bath(Q-Ball)

precipitate was observed. This precipitate was removed from the solution by filtering before the solutions were used.

3. ^{56}Mn γ-ray Measurement

The neutron source was positioned in a cylindrical teflon beaker of 10 cmϕ×10 cm and of 1 cm thickness located at the center of the bath filled with a $MnSO_4$ solution. The solution was transferred with a 1/2 Hp pump from the bath to a monitor bath equipped with two 3.8 cmϕ×3.8 cm NaI(Tl) detectors for the detection of γ-rays from ^{56}Mn produced by ^{55}Mn(n, γ) ^{56}Mn reaction. The growth and decay data acquisitions of the ^{56}Mn activity were made at a flow rate of about 10 l/min.

Figure 6 shows a block diagram of the ^{56}Mn gamma counting system. Signals were fed from the photomultipliers through preamplifiers, thence to amplifiers. The single channel analyzers(SCA) were used for discrimination. In the experiments, the ^{56}Mn γ-ray monitoring system was calibrated using a ^{137}Cs source(NBS 4200 B-14). The operating voltage of the detectors was set to 670 V.

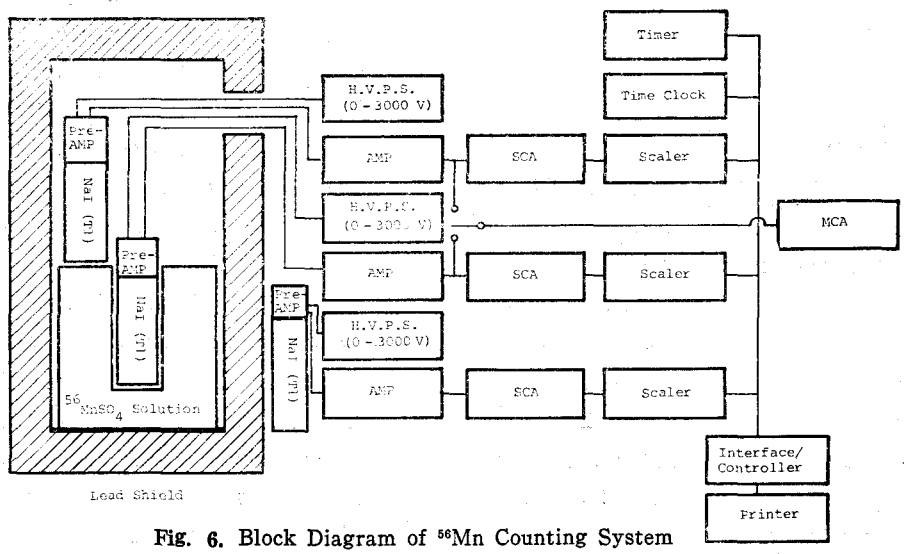

Fig. 6. Block Diagram of ^{56}Mn Counting System

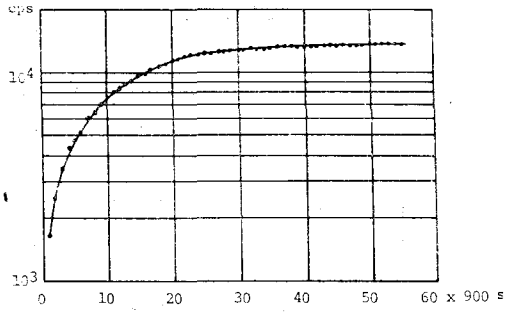

Fig. 7. ^{56}Mn Activity Growth Curve

IV. Experimental Results

The ^{56}Mn half-life measurement was carried out using the neutron source, ^{241}Am-Be (S.N. 4637 NE, Amersham), calibrated by the National Physical Laboratory in Teddington, England. From the acquisition data plotting, the half-life of ^{56}Mn nuclide was determined to be about 2.6 h. This result is close enough to the value, 2.580 h, given in the nuclear data[8]. Figure 7 shows the ^{56}Mn activity growth curve, while Figure 8 shows the ^{56}Mn activity decay curve.

In this work, the ^{241}Am-Be neutron source was regarded as a reference source for the strength measurement of a ^{252}Cf spontaneous fission neutron source. The ^{252}Cf source strength was determined to be 3.71×10^{7} n/s as of November 15, 1985 by the comparative method using Eq. (2). The ^{252}Cf source was purchased from the Amersham International Ltd. without a calibration certificate of measurement. According to the supplier, however, the source strength was 1.15×10^{8}n/s as of June 23, 1981. By the physical decay correction of ^{252}Cf with its half-life of 2.651 years[9], the source strength of 3.60×10^{7} n/s was obtained. This result shows that the difference between the measured and calculated values is about 3%.

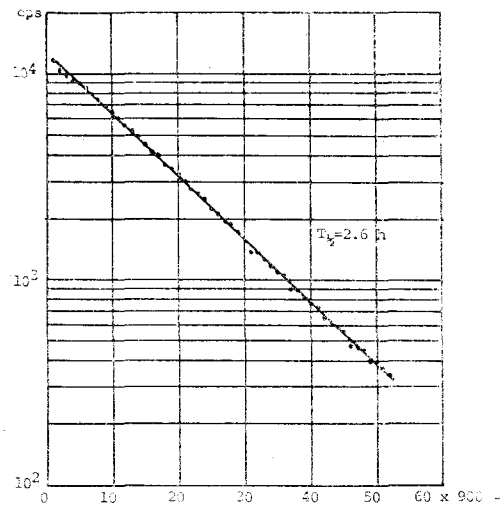

Fig. 8. ^{56}Mn Activity Decay Curve

V. Conclusions

The development study of a manganese sulphate ($MnSO_4$) bath system has included the absolute measurement of the half-life of ^{56}Mn nuclide and the comparative measurement of the source strength of ^{252}Cf neutron source for the systematics of the $MnSO_4$ bath method.

The combined information from the experimental results clarified that the $MnSO_4$ bath system was well designed and successfully constructed. In this regard, the KSRI $MnSO_4$ bath (Q-Ball) system can be regarded as a pilot system for establishment of the neutron measurement standards in Korea.

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$MnSO_4$용액조 장치 개발 연구

황선태 · 이경주 · 최길웅 · 김원식

한국표준연구소 방사선표준연구실

=요 약=

중성자 측정의 국가표준을 확립하기 위하여 국제원자력기구(IAEA)로 부터 기술지원을 받아서 $MnSO_4$용액조장치를 개발하였다. 본 용액조장치는 구형의 s.s. 316 L $MnSO_4$용액조(두께 3.5 mm, 내경 125 cm), 용액의 순환계 및 ^{56}Mn γ-선 검출계등으로 구성되어 있다.

용액조 본체로부터 퍼내어진 용액은 2개의 3.8 cmϕ×3.8 cm NaI(Tl) 검출기가 설치된 Marinelli 비이커형의 모니터 용기로 진입되어 순환된다. 본 장치의 성능검사는 ^{241}Am-Be 과 ^{252}Cf 중성자 선원을 사용하여 수행하였다.

^{56}Mn 방사능의 붕괴 곡선을 분석한 결과, ^{252}Cf 의 중성자 방출율은 1985년 11월 15일 현재 3.71×10^7 n/s per 50 μg 으로 얻어졌다.