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Radioactive Neutron Source Calibration at the Korea Standards Research Institute

Sun-Tae Hwang, Kil- Oung Choi

Radiation Standards Laboratory
Korea Standards Research Institute(K-SRI)

=Abstract=

The manganous sulfate bath method for neutron source calibrations at the K-SRI is described together with the measurement of neutron emission rate of a source and the corrections applied for capture by competing nuclei of neutrons, and thermal neutron leakage, neutron absorption in the source itself. The commercially available neutron sources(Am-Be, ^{252}Cf) for the calibration checks of neutron radiation instruments in the MeV range are considered in this paper.

Introduction

This paper introduces a manganous sulfate ($MnSO_4$) bath system which in under installation in the Radiation Standards Laboratory at the K-SRI in partial support of International Atomic Energy Agency and describes the $MnSO_4$ bath method to calibrate the emission rate of radioactive neutron sources.

Neutron sources which are used for the calibration of neutron radiation instruments are selected on the basis of their size, stability, long half-life, energy spectrum and emission rate. The emission rate is used to calculate the fluence rate of the neutron field at the position of the neutron radiation instrument.

Very significant improvements in neutron source calibrations were achieved in 1960. At present, the $MnSO_4$ bath method gives the best accuracies by using better cross section data and modern knowledge of the neutron spectrum. It is the purpose of this paper to describe that the $MnSO_4$ bath operation is most easily checked by an^{241}Am-Be or a^{252} Cf neutron source.

Manganous Sulfate Bath Method

At the beginning stage, the manganous sulfate bath method will be used to check up on the neutron emission rate of the known sources in the Radiation Standards Laboratory at the K-SRI. The principle of this method is indicated in Figure 1. Neutrons emitted from the source enter the bath, are slowed down by a series of collisions, and finally captured by a ^{55}Mn nucleus to become ^{56}Mn. The bath consists of a 1.25 m diameter sphere filled

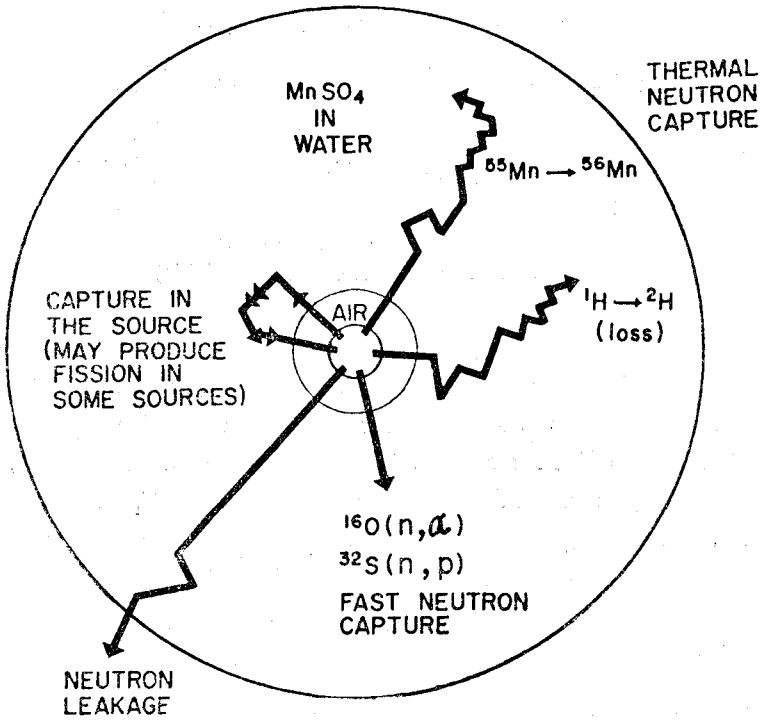

Fig. 1. Schematic Diagram of the Principle of $MnSO_4$ Bath Method

with an aqueous solution of manganous sulfate. The source is placed in a thin-walled, air-filled, teflon beaker at the center of the sphere where it is calibrated under the condition of a nearly saturated solution of $MnSO_4 \cdot H_2O$ in pure water.

Due to the large size of the bath, it can be assumed that very few neutrons escape from the $MnSO_4$ System. Most neutrons from the source are captured by any of the following processes:[1] (a) fast capture in the fluorine (^{19}F) of the beaker, (b) fast capture in ^{16}O or ^{32}S, (c) slow neutron capture in ^{55}Mn or hydrogen, or (d) slow neutron capture in the beaker or the source itself. However, the majority of neutrons are moderated in the bath and than captured primarily in the ^{55}Mn and hydrogen. The slow neutron capture in ^{55}Mn produces ^{56}Mn which decays to ^{56}Fe with a half-life of 155 minutes. The induced ^{56}Mn activity in the bath solution is monitored as a measure of neutron emission rate of the source. It is the gamma activity of the ^{56}Mn which is measured at the NaI(Tl) crystals.

A circulating manganous sulfate bath system as shown in Figure 2 is a type used at the U.S. NBS and another system as shown in Figure 3 is a type used at the LMRI in France. Among those types, the bath system of LMRI-type is under installation at the K-SRI. In the bath system, radioactive $MnSO_4$ solution is passed continuously through a lead shielded

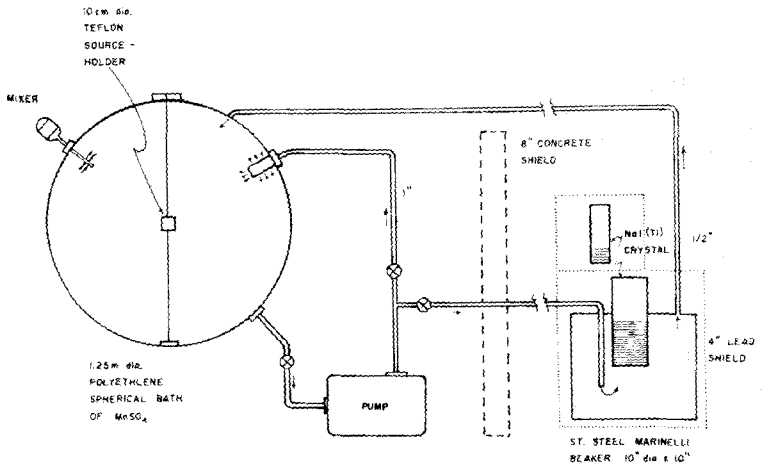

Fig. 2. Schematic Representation of NBS-type Monganese Bath System.

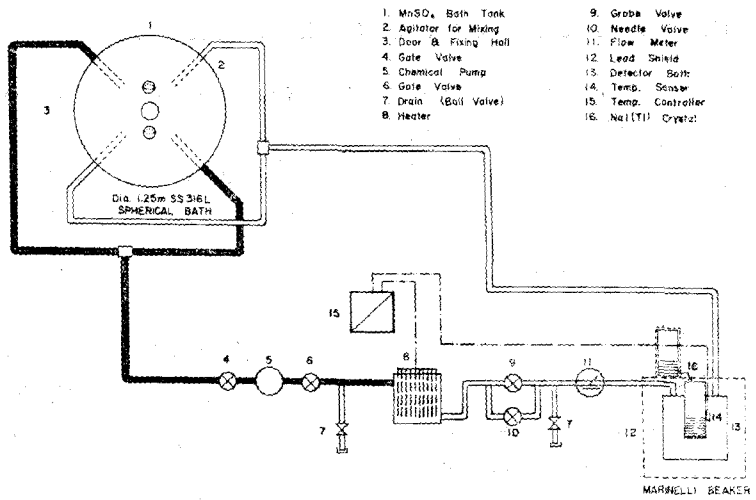

Fig. 3. Schematic Representation of LMRI-type Manganese Bath System.

Marinelli beaker monitored by gamma-ray detectors which have high stability and efficiency. The circulating system pumps thoroughly mix the $MnSO_4$ bath solution by maintaining a flow rate of about 10 L/min. At a flow rate of about 5 L/min, the solution flows to the remotely located gamma-ray detection system where the bath counting can be continued and statistics are accumulated as long as desired.

The NaI(Tl) crystal, located at the center of the Marinelli beaker, is used for neutron sources with emission rates between 10^5 n/s and 10^7 n/s. The second crystal, located above the Marinelli beaker, views a small fraction of the radioactive bath solution through the lead shield and thus is used for neutron sources with emission rates between 10^7 n/s and 10^9 n/s. The reason why the second crystal is needed for more intense neutron sources is

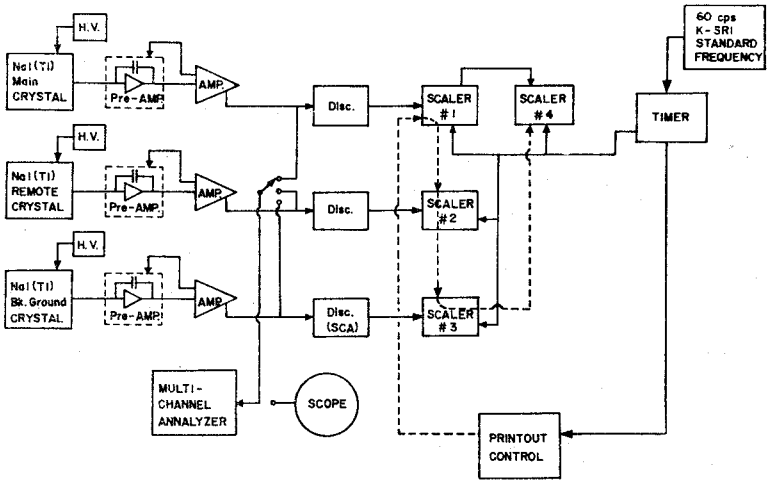

Fig. 4. Detailed Electronic Setup for Neutron Source Calibration

because the first crystal counting system would be paralyzed by the resulting ^{56}Mn activity.

Calibration of Neutron Emission Rates

A block diagram of $MnSO_4$ bath counting system used for neutron source calibration is shown in Figure 4. The necessary counting data for determination of neutron emission rate are acquired by this system. The dimensions of main, remote, and background crystals are $1.5''\phi \times 1.5''$, $1.0''\phi \times 1.0''$, and $1.5''\phi \times 1.5''$, respectively.

The calibration of the emission rate, Q, of a radioactive neutron source involves absolute measurement of ^{56}Mn activity by the $4\pi\beta$-γ coincidence counting system and the determination of detection efficiency of the bath-detector system.

For the determination of source strength, the neutron emission rate of a sonrce is derived from the following equation:[2)]

$$Q = \frac{A}{f\epsilon(1-S)(1-O)(1-L)} \quad \cdots\cdots(1)$$

where, A = The saturation ^{56}Mn activity counting rate measured by the bath-detector system,

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

ϵ = The detection efficiency of the system,

S = The fraction of thermal neutrons recaptured by the source,

O = The fraction of fast neutrons undergoing (n,α) and (n,p) reactions in ^{16}O and ^{32}S in the bath solution,

L = The fraction of neutrons escaping from the bath boundary.

The two major uncertainties of the $MnSO_4$ bath system are thermal neutron absorption in ^{1}H and fast neutron absorption in ^{16}O and ^{31}S.

Table I. Corrections for the $MnSO_4$ Bath Calibration[6]

Neutron Source / Correction Type	^{241}Am-Be(α,η) (%)	^{252}Cf (%)
O_2 & 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
Total Error	±0.75	±0.41

The inverse ^{55}Mn capture fraction, 1/f, in the absence of impurities is given by:[3]

$$\frac{1}{f}=1+\frac{\sigma s}{\sigma_{Mn}(1+G\bar{r}s)_{Mn}}+\frac{N_H}{N_{Mn}}\times\frac{\sigma_H}{\sigma_{Mn}(1-G\bar{r}s)_{Mn}} \quad \cdots\cdots(2)$$

where, σ_X=The thermal neutron capture cross section for element X,

N_X=The atom concentration of element X in the bath solution,

G=The resonance selfshielding factor,

$\bar{r}$=The Westcott[4] epithermal flux parameter averaged over the bath system,

s=The Westcott parameter representing the resonance activation integral.

Oxygen(^{16}O) is omitted from this expression because the thermal neution capture cross section of oxygen can usually be neglected. Since manganese (^{55}Mn) is not a perfect $\frac{1}{v}$ detector, the additional neutron capture in the resonance region has to be taken into account by the factor $(1+G\bar{r}s)$.[5]

The corrections for a calibration of ^{241}Am-Be and ^{252}Cf are listed in Table I. Escape from the 1.25m diameter bath is quite small and essentially non-existent for low energy neutron sources. The error in the correction depends upon the size of the correction. The correction is minimized by locating the neutron source in an air cavity with thin teflon walls to reduce fast neutrons to the thermal neutron flux.

The total errors listed in Table I. result from the linear addition of the root mean square sum of systematic errors and the random error of ±0.2%.

In the Center for Radiation Research at the NBS, the calibration of neutron emission rate for an unknown source has been done as follows:[7]

$$Q_u=\frac{(C_n)_u\times(F_{a,l})_u\times Q_{NBS-I}\times R_{mtr}}{(C_n)_{NBS-I}\times(F_{a,l})_{NBS-I}} \quad \cdots\cdots(3)$$

where, Q_u=The unknown source strength,

$(C_n)_u$=The net count rate of unknown source in the $MnSO_4$ bath,

$(C_n)_{NBS-I}$=The net count rate of NBS-I in the bath on the calibration date,

$(F_{a,l})_u$=The correction factor for absorption and leakage for unkonown source,

$(F_{a,l})_{NBS-I}$=The correction factor for absorption and leakage for NBS-I,

R_{mtr} = The main-to-remote count rate ratio,

Q_{NCS-I} = The source strength of NBS-I on the calibration date.

The NBS-I standard Ra-Be(γ,η) source consists of a beryllium sphere, 4cm in diameter of which a 1 curie of radium in the form of $RaBr_2$ enclosed in platinum-iridium capsule of 0.2mm wall thickness is placed. As only gamma rays can enter the beryllium. the neutron emission rate is quite constant, unlike a Ra-Be(α,η) source. The factor, R_{mtr}, is only necessary when the remote crystal has been used to obtain the count rate of the unknown source in the bath.

Conclusions

Once a neutron source calibration is done by the manganous sulfate bath method, the source can be considered as a certified neutron source. The neutron sources commonly used for instrument calibration checks are ^{241}Am-Be, ^{226}Ra-Be, and ^{239}Pu-Be. Their average neutron energies are approximately 3.9 MeV, 2.8 MeV and 3.4 MeV, respectively. Only the Am-Be sources have a single well known half-life, 433 years. Another popularly used neutron source is spontaneou sly fissionable ^{252}Cf. Its average neutron energy is 2.3 MeV. The spectrum of ^{252}Cf was well studied at the NBS.[9] However, it has an inconveniently short half-life of about 2.64 years.

The Radiation Standards Laboratory at the K-SRI has two ^{241}Am-Be sources and one ^{252}Cf source. The Laboratory is expecting another ^{252}Cf source in 1986 from IAEA. When the installation of the manganous sulfate bath system is completed, the bath system calibration should be first tried to measure the neutron emission rates of those neutron sources. The performance test of the bath system is also necessary for possible improvement in the accuracy of neutron instrument checks.

In view of the country's fast developing nuclear power program and in support of its nuclear safety program for the personnel protection against neutron radiation, the $MnSO_4$ bath system shall contribute to the establishment of neutron measurments standars. The bath system will be also used for the calibration of neutron beams for neutron research and neutron therapy in the near future.

REFERENCES

1) V. Spiegel, "Neutron Source Calibrations at NBS for Calibration Checks of Neutron Radiation Instruments," *NBS SP* **456**, 87(1976).

2) E.J. Axton, "Accuracies and Corrections in Neutron Bath Techniques," *NBS SP* **493**, 237(1977).

3) E.J. Axton and A.G. Bardell, "Re-evaluation of Neutron Emission Rates Measured at the National Physical Laboratory," *Metrologia* **18**, 97(1982).

4) C.H. Westcott, W.H. Walker, and T.K. Alexander, *Proc. Second International Conference on the Peaceful Uses of Atomic Energy*, Geneva, United Nations(1958).

5) K.W. Geiger, "Recent Improvements in the Absolute Calibration of Neutron Sources," *Metrologia* **4**, 8(1968).

6) V. Spiegel, "Procedure for $MnSO_4$ Bath Calibr-

ations of Intense Neutron Sources," Personal Work Log at the NBS (1981).
7) E.D. McGarry, "Neutron Source Strength Calibration," Personal Work Log at the NBS(1981).
8) J.A. Grundl and C.M. Eisenhauer, "Fission Spectrum Neutrons for Cross Section Validation and Neutron Flux Transfer," Conf. on Nuclear Cross Sections and Technology, Washington, D.C., 1, 250(1975).

K-SRI에서의 방사성 중성자 선원교정

한국표준연구소, 방사선표준연구실

황 선 태·최 길 웅

초 록

임의 중성자 선원의 중성자 방출율 측정과 경합핵종에 의한 중성자 포획, 열중성자 누출 및 선원자체의 중성자 흡수에 적용되는 보정을 포함하여 한국표준연구소에서의 중성자 선원교정을 위한 $MnSO_4$ 용액조 방법을 기술한다. 본 보고서에서는 에너지가 MeV 영역에서 사용되는 중성자 방사선 기기의 교정검사를 위하여 상용화되어 있는 중성자 선원(Am-Be, ^{252}Cf)을 고려하였다.