

## Determination of Neutron Absorption Fraction Factor in Manganese Sulfate Bath System

Kyung-Ju Lee, Kil-Oung Choi, Sun-Tae Hwang

Korea Standards Research Institute

Kun Jai Lee

Korea Advanced Institute of Science and Technology

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### 황산망간 용액조 장치의 중성자 흡수분율 보정인자 결정

이경주 · 최길웅 · 황선태

한국표준연구소

이건재

한국과학기술원

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#### Abstract

The correction factor of neutron fraction absorbed by  $^{55}\text{Mn}$  in the  $\text{MnSO}_4$  bath was determined for the absolute measurement of neutron emission rate by using the solution circulation-type manganese sulfate bath system. For the determination of this correction factor,  $1/f$ , the atomic number density and the effective neutron capture cross section data of Mn, S and impurity elements in the  $\text{MnSO}_4$  solution were determined. For the atomic number density determination, the  $\text{MnSO}_4$  solution concentration was determined by using the volumetric EDTA titration and gravimetric method. The impurity contents were analyzed by using the ICP method. For the calculation of effective neutron capture cross sections, a FORTRAN computer program EASCAL was developed in this study, in which Westcott's parameters and Axton's empirical relations are used.

#### 요 약

용액 순환형 황산망간 용액조 장치를 이용한 중성자 방출류의 절대측정을 위해서 황산망간 용액이 채워진 용액조 안에서  $^{55}\text{Mn}$ 에 의해 흡수되는 중성자 분율에 대한 보정인자를 결정하였다. 이 보정인자,  $1/f$ 은 망간, 유황 및 용액에 함유된 불순물 원소들의 원자수 밀도와 유효중성자 포획단면적을 이용하여 결정되었다. 각 원소들의 원자수 밀도의 결정을 위한 용액의 농도는 EDTA 적정에 의한 용량법과 중량법에 의해 분석하였고 불순물 함량은 ICP 방법을 이용하였다. 한편, 유효 중성자 포획 단면적은 자체 작성한 FORTRAN Program EASCAL, Westcott의 매개변수 및 Axton의 실험식을 사용하여 계산하였다.

## 1. Introduction

The manganese sulfate bath method has been widely used for the neutron emission rate measurement of a radioactive neutron source since 1960.

In this method a neutron source is placed at the center of the spherical bath, and the fast neutrons emitted from the source are moderated, captured and/or scattered, by the hydrogen, sulphur, manganese and others which are impurities in the manganese sulfate solution in the bath.

The captured neutron by  $^{56}\text{Mn}$  produces the radioactive  $^{56}\text{Mn}$ , i.e.,  $^{56}\text{MnSO}_4$  solution, which is continuously circulated through the detection system, where thus one can count the  $^{56}\text{Mn}$   $\gamma$ -activity which corresponds to the number of neutrons affecting the production of  $^{56}\text{Mn}$  nuclides and emitted from the neutron source.

The neutron emission rate of a neutron source,  $Q$ , is given by the following equation.

$$Q = A/\epsilon \cdot f(1-L)(1-O)(1-S) \quad (1)$$

Where  $\epsilon$  is the system efficiency,  $1/f$  is the inverse neutron absorption fraction,  $L$  is the correction factor of neutrons leaked from the bath boundary,  $O$  is the correction factor of the fraction of fast neutrons captured by sulphur and oxygen and  $S$  is the correction factor of the thermal neutrons captured by the source itself and the source holder.

The inverse neutron absorption fraction,  $1/f$ , in the manganese sulphate bath is given by

$$\begin{aligned} 1/f &= \frac{N_{\text{Mn}}\sigma_{\text{Mn}}(1+G\bar{r}s)_{\text{Mn}} + N_{\text{S}}\sigma_{\text{S}}(g+\bar{r}s)_{\text{S}} + N_{\text{H}}\sigma_{\text{H}}(g+\bar{r}s)_{\text{H}} + N_0\sigma_0 + \sum_i N_i\sigma_i(g+\bar{r}s)_i}{N_{\text{Mn}}\sigma_{\text{Mn}}(1+G\bar{r}s)_{\text{Mn}}} \\ &= 1 + \frac{\sigma_{\text{S}}(g+\bar{r}s)_{\text{S}}}{\sigma_{\text{Mn}}(1+G\bar{r}s)_{\text{Mn}}} + \frac{N_{\text{H}}}{N_{\text{Mn}}} \cdot \frac{\sigma_{\text{H}}}{\sigma_{\text{Mn}}(1+G\bar{r}s)_{\text{Mn}}} + \frac{\sum_i \sigma_i(g+\bar{r}s)_i}{N_{\text{Mn}}\sigma_{\text{Mn}}(1+G\bar{r}s)_{\text{Mn}}} \end{aligned}$$

Where  $N_{\text{H}}$ ,  $N_{\text{Mn}}$ , are the number density of H, Mn and impurities, respectively and  $\sigma_{\text{S}}(g+\bar{r}s)_{\text{S}}$ ,  $\sigma_i(g+\bar{r}s)_i$ ,  $\sigma_{\text{Mn}}(1+G\bar{r}s)_{\text{Mn}}$  are the effective cross section of S, impurities and Mn, respectively. The  $g$ ,  $\bar{r}$ ,  $s$  and  $G$  will be explained in the next section. The  $N_0 \cdot \sigma_0(g+\bar{r}s)_0$

term is omitted because the absorption cross section of Oxygen,  $\sigma_0$ , is negligible compared to  $\sigma_{\text{Mn}}$ .

In this work, the chemical analysis and theoretical calculations were carried out for the determination of the atomic number densities and the effective neutron capture cross sections of the each elements for the determination of absorbed neutron fraction factor.

## 2. Principle of the Effective Cross Section Determination

For the determination of neutron absorption fraction factor in the manganese sulfate bath, it is required that the effective neutron capture cross section be considered because the neutrons emitted from a neutron source have the wide range of energies which contain the thermal and the epithermal neutrons.

According to the Westcott's convention [1] for effective capture cross section, the neutron reaction rate,  $R$ , is given by

$$R = n v_0 \cdot \hat{\sigma} \quad (3)$$

Where  $n$  is the neutron density including both thermal and epithermal neutrons,  $v_0$  is the neutron velocity, 2200 m/s and  $\hat{\sigma}$  is the effective capture cross section.

In this convention,  $n \cdot v_0$  has a role of flux,  $\phi$ , and  $\hat{\sigma}$  is defined by following equation.

$$\hat{\sigma} = \sigma_0 \cdot (g + rs) \quad (4)$$

where  $\sigma_0$  is the cross section for neutrons of 2200 m/s and  $g$  and  $rs$  are special values as a function of the

temperature  $T$  depending on the departure of the  $1/v$  form. If the neutron cross section follows only  $1/v$  form, the values of  $g$  and  $s$  are 1 and 0, respectively.

These values are derived by the neutron velocity distribution and neutron flux density. The detailed

derivation processes of them are omitted in this paper.

The neutron reaction rate,  $R$ , is also given by

$$R = n \cdot v_0 \cdot \hat{\sigma} = \int_0^\infty (v) \cdot \sigma(v) \cdot v \, dv \dots \dots \dots (5)$$

$$= n(1-p) \int_0^\infty \frac{4}{\sqrt{\pi}} \frac{v^3}{v_T^3} e^{-(v/v_T)^2} \cdot \sigma(v) \, dv + np \cdot v \cdot \frac{1}{\sqrt{\pi}} \int_0^\infty \frac{1}{v} \sigma(v) \, dv$$

In this equation, if the pure Maxwellian distribution is only considered i.e., if  $P=0$ ,  $R$  is given by

$$R = n \cdot v_0 \cdot \hat{\sigma} = n \cdot \int_0^\infty \frac{4}{\sqrt{\pi}} \frac{v^3}{v_T^3} e^{-(v/v_T)^2} \cdot \sigma(v) \, dv \dots \dots \dots (6)$$

Where effective cross section  $\hat{\sigma}$  is equal to  $\hat{\sigma}_m$  which is the effective cross section of the Maxwellian distribution neutrons. Now the  $g$ -value is defined as the following equation.

$$g = \hat{\sigma}_m / \sigma_0 = \frac{1}{v \cdot \sigma_0} \int_0^\infty \frac{4}{\sqrt{\pi}} \frac{v^3}{v_T^3} e^{-(v/v_T)^2} \cdot \sigma(v) \, dv \dots \dots \dots (7)$$

In the mean time, if it has not the pure Maxwellian distribution, equation(6) will become

$$\hat{\sigma} = g \cdot \sigma_0 + \frac{P \cdot \sqrt{\mu} \cdot v_T}{2 v_0} \int_0^\infty \left[ \sigma(v) \cdot \frac{v_0 \cdot \sigma_m}{v} \right] \frac{\Delta \cdot dE}{E} \dots \dots \dots (8)$$

[  $\Delta=1$  for  $E > \mu kT$ ,  $\Delta=0$  for  $E < \mu kT$  ]

At this stage, we can define the spectral index,  $r$ , which is given by

$$r = P \cdot \frac{1}{\pi \cdot \mu / 4} \dots \dots \dots (9)$$

and the excess resonance absorption integral,  $s$ , which accounts for the contribution of the epithermal neutron flux distribution to the thermal neutron flux distribution is given by

$$s = \frac{2}{\sqrt{\pi}} \cdot \sqrt{\frac{T}{T_0}} \frac{\Sigma'}{\sigma_0} \dots \dots \dots (10)$$

Where  $\Sigma'$  is the integral term of Eq(8).

In order to determine the effective capture cross section of  $^{55}\text{Mn}$  in the manganese sulfate bath K.W. Geiger [2] suggested that the capture cross section of  $^{55}\text{Mn}$  increases as much as  $\sigma_{0\text{Mn}} \cdot (1 + G\bar{s})_{\text{Mn}}$  where  $G$  is the resonance neutron self-shielding factor which

can be calculated by using the flux measurement from the activated thin Mn foil. But this  $G$ -value is more difficult to evaluate in the manganese sulfate bath because it has a large moderation media where strong scattering resonances are present. E.J.Axton et al., [3] calculated the  $G$ -value in the manganese sulfate bath by using the removal cross section of manganese,  $\sigma_{r\text{Mn}}$ .

They assumed that the resonance neutrons were monoenergetic with a removal cross section,  $\sigma_r$  and suggested that  $G$ , for the manganese sulfate solution, is given by

$$G = \frac{\sum_k N_k \sigma_{sk}}{\sum_k N_k \sigma_{sk} + N_{\text{Mn}} \cdot \sigma_{r\text{Mn}}} \dots \dots \dots (11)$$

Where  $N_k$  is the atomic number density of nuclide  $k$  and  $\sigma_{sk}$  is the scattering cross section of nuclide  $k$ . The averaged epithermal flux parameter,  $\bar{r}$ , was defined by the ratio of epithermal flux density,  $\phi_{\text{epi}}$ , to Maxwellian flux density  $\phi_{\text{max}}$  and is given by

$$\bar{r} = \frac{\phi_{\text{epi}}}{\phi_{\text{max}}} = \frac{\sqrt{\pi}}{2} \frac{\sum_k N_k \sigma_{0k}}{\sum_k N_k \sigma_{sk} \cdot \xi_k}$$

Where  $\sigma_{0k}$  is the capture cross section of nuclide  $k$  and  $\xi_k$  is the averaged logarithmic decrement of nuclide  $k$  in the bath and is given by [4]

$$\xi = 1 - \frac{(A-1)^2}{2A} \ln \left( \frac{A+1}{A-1} \right)$$

Where  $A$  is the mass number of the scattering nuclide.

### 3. Experimental Results

#### 1) Chemical Analysis of the $\text{MnSO}_4$ Solution Concentration

It is well known that the volumetric method by EDTA (Ethylene Diamine Tetracetic Acid) titration and gravimetric method are used for the concentration analysis of  $\text{MnSO}_4$  solution introduced in the manganese bath. In this work, 8 samples of  $\text{MnSO}_4$  solution were analyzed by using these two methods, whose detail procedures are referred to J.R.Smith's report [5]. The results of the concentration analysis for 8 solutions are shown in Table 1.

**Table 1.  $\text{MnSO}_4$  Solution Concentration Data.**Unit : g of  $\text{MnSO}_4 \cdot \text{H}_2\text{O}$  / kg of Sol'n

No.	CV[Conc. by EDTA]	$\pm\%$	CG[Conc. by Grav]	$\pm\%$	%*
1	262.5464	0.8004	271.1594	0.1963	3.2121
2	215.5414	0.7242	223.6434	0.4813	3.6227
3	182.3694	3.8879	189.7337	0.4201	3.8814
4	126.5195	0.6342	131.1546	0.8336	3.5341
5	104.3047	1.0434	110.0724	0.2972	5.2399
6	96.4608	0.3369	99.4206	0.1353	2.9770
7	71.2710	0.7808	72.3658	0.2901	1.5129
8	41.9511	1.1466	44.6714	0.1757	6.0896

$$\times \% = (\text{CG} - \text{CV}) / \text{CG} \times 100$$

According to the results in Table 1., the concentration by EDTA titration, CV, are less than the concentrations by gravimetric method, CG. It accounts for the existence of the impurities which were analyzed by ICP method and will be mentioned following section 2.

## 2) Impurity Analysis of $\text{MnSO}_4 \cdot \text{H}_2\text{O}$

The impurities in the  $\text{MnSO}_4$  solution absorb the neutrons emitted from a neutron source and moderated by the solution in the manganese bath. To calculate the fraction of the neutrons captured by impurities, the impurity contents must be determined.

Table 2. shows the analytical result of impurity contents in the  $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ . These results were obtained by ICP method [6]. The error range of the impurities is  $\pm 5\%$  typically.

**Table 2 : Impurities in the  $\text{MnSO}_4 \cdot \text{H}_2\text{O}$** 

Impurity element	Content(ppm)
Co	61
Cd	9
Ni	72
Cu	13
Al	14
Zn	2201
Fe	20
Cr	4
Ca	1383
Mg	3404
K	2650
Na	150
Total	9981

## 3) Calculations of $N_H/N_{Mn}$ Effective Cross Sections and 1/f Factors.

In order to calculate the correction factor of the manganese capture fraction of neutrons, 1/f, the atomic ratios of the number density of hydrogen atoms to the number density of manganese atoms were firstly calculated by using the concentration data of  $\text{MnSO}_4$  solution and the atomic number density data which were obtained by impurity analysis. The results of  $N_H/N_{Mn}$  ratio calculation are summarized in Table 3.

In This Work, The  $N_H/N_{Mn}$  ratios in the third column were used for the determination of 1/f factor. the effective cross sections of manganese,  $(1 + G\bar{r}s)_{Mn}$ , sulfur,  $(1 + \bar{r}s)_s$ , and impurities,  $\sum_i N_i \cdot \sigma_i(g + r_s)_i$ , were calculated by using the results and equations of the previous sections and the developed program EASCAL.

The calculation results of effective cross section for the  $\text{MnSO}_4$  solutions are shown in Table 4., where the excess resonance absorption integral of sulfur ( $S_s = 0.025$ ) referred to the report of Axton et al. [7]

For the evaluation of the effective cross section of manganese in the  $\text{MnSO}_4$  solution used in this work, the obtained values of  $(G\bar{r}s)_{Mn}$  were compared with the results of De Juren [8], Wunderer [9], Louwrier [10], and Axton [3]. The comparison results are shown in Fig. 1.

For the calculation of inverse neutron absorption fraction factors and their errors, the EASCAL program was used also. Table 5. shows the 1/f factors and their errors depended on the solution concentration,  $N_H/N_{Mn}$  ratio values.

**Table 3 :  $N_H/N_{Mn}$  Ratios of  $MnSO_4$  Solutions.**

Sample No.	$N_H/N_{Mn}$ Ratio			Calculation Method :
	( I )	( II )	( III )	
1	47.0851	45.0342	46.5294	( I ) $N_H/N_{Mn} = \frac{1000 \times CV}{CV} \times F^*$
2	61.0091	58.1916	60.3790	
3	75.1554	71.5877	74.4785	
4	115.7313	11.0489	115.1172	( II ) $N_H/N_{Mn} = \frac{1000 \cdot CG}{CG} \times F^*$
5	143.9500	135.5288	143.0230	
6	157.0187	151.8451	156.5043	
7	218.4396	209.9382	217.8110	( III ) $N_H/N_{Mn} = \frac{1000 \cdot CG}{CV} \times F^*$
8	382.8243	368.3590	382.1947	

\*  $F = 16.7631335$ 

$$F = \frac{\text{g of H}_2\text{O} \times \frac{2.0158(\text{H}_2)}{18.0152(\text{H}_2\text{O})} \times \frac{2 \text{ Mole}}{2.0158(\text{H}_2)} \times \frac{6.023 \times 10^{23}}{1 \text{ Mole}}}{\text{g of MnSO}_4 \times \frac{54.9380(\text{Mn})}{150.9956(\text{MnSO}_4)} \times \frac{1 \text{ Mole}}{54.9380(\text{Mn})} \times \frac{6.023 \times 10^{23}}{1 \text{ Mole}}}$$

**Table 4. Effective Cross Section Data**

No.	$(1 + G\bar{r}s)_{Mn}(b)$	$(1 + \bar{r}s)_s(b)$	$\sum_i N_i \sigma_i (1 + \bar{r}s)_i(b)$
1	1.014158	1.000681	198.61360
2	1.013333	1.000608	154.41220
3	1.012757	1.000561	126.71570
4	1.011782	1.000488	83.02465
5	1.011437	1.000464	68.21178
6	1.011264	1.000453	60.68726
7	1.010852	1.000433	44.13784
8	1.010366	1.000396	25.30478

**Table 5. Calculated 1/f Values**

$N_H/N_{Mn}$	1/f factors	% errors
46.5294	$2.192697 \pm 0.03235034$	1.475
60.3790	$2.535464 \pm 0.03931766$	1.551
72.4785	$2.884489 \pm 0.08443096$	2.927
115.1172	$3.890740 \pm 0.06801727$	1.748
143.0230	$4.581690 \pm 0.0880878$	1.923
156.5043	$4.915681 \pm 0.08772113$	1.785
217.8110	$6.433942 \pm 0.12550100$	1.951
382.1947	$10.505210 \pm 0.2298067$	2.188

#### 4. Conclusions

From the present work the conclusions can be made as follows:

- 1) The  $1/f$  factors determined have accuracy of  $\pm 3\%$
- 2) The  $(Grs)_{Mn}$  values are comparable with the Axton's results, but it is about 5% of discrepancy between ours and Axton's. This difference is due to the existence of impurities in the solution used. That is, our impurity contents are more than Axton's impurity contents.
- 3) It is required that the impurity analysis of solution be more precise and the elements having high neutron capture cross section, Cd, B and rare earth elements, have to be analyzed.

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