

ABSOLUTE TECHNIQUE FOR NEUTRON SOURCE CALIBRATION BY RADIATION INDUCED ACTIVITY

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Abstract

The neutron yield from a Radium Beryllium neutron source has been determined experimentally by the induced Mn-56 activity. The neutron source was placed in the center of a tank filled with aqueous manganese sulphate (MnSO₄) solution. Irradiation time usually lasted about 16-18 hours in order to secure saturation. The average induced Mn-56 activity within the MnSO₄ bath was then measured by the use of NaI scintillation detector. This detector was placed in a sealed aluminum jacket at the center of the tank. This detector was connected with the necessary electronic counting system and was precalibrated against a 4 $\pi\beta\text{-}\gamma$ coincidence counting system. The efficiency of the NaI counting system as a function of MnSO₄ solution density is investigated as well as the proper dimension of the used tank for the sake of calibration purposes. The neutron leakage within the MnSO₄ baths was also investigated for different dimensions of tanks. The experimental errors involved in the counting system were also considered. The numerical value of neutron yield from the used radium beryllium neutron source was given with its corresponding statistical errors as $(1.10 \pm 0.065) \times 10^6$ neutron per second.

Key Words: *Neutron source, calibration, manganese sulphate, induced activity, neutron leakage.*

INTRODUCTION

Numerous techniques are involved for evaluating neutron yield from neutron sources [1-3], from these methods:

- (1) Sources immersed in a large moderating volume containing a solution of manganese salt and the neutron flux out put was determined from the Mn-56 induced activity [4-7].
- (2) Sources immersed in a large moderator and the neutron density are integrated by BF₃ counters [8, 9].
- (3) Determination of the neutron yield from neutron sources by induced activity of some threshold detectors [10-12].
- (4) Sources are placed in the center of a moderator as water or graphite with thermal neutron detector located at a large distance from the source [13-15].

The object of this experimental work is to:

- (1) Determine the optimum conditions for calibration of neutron sources and to find out the efficiency of measuring system.
- (2) Test experimentally the feasibility of using MnSO₄ baths of various sizes for the purpose of calibration of neutron sources.
- (3) Determine the absolute neutron emission rate from radium beryllium (Ra – Be) neutron source.

EXPERIMENTAL

Variety of aluminum tanks of different sizes are used for this calibration. These are six cylindrical tanks painted from inside and outside to prevent corrosion. The height of each tank is equal to its diameter. Their dimensions (six tanks) are 30, 35, 40, 45, 50 & 60 cm (for the height and diameter.). These tanks were filled with MnSO₄ solution. The concentration of the solution was determined with high accuracy. The counting system consists of NaI (Tl) detector connected with ultra-scaler. To calibrate this system (aqueous MnSO₄ solution in the tank + counting scaler), an auxiliary experiment is carried out. Small quantity of solid MnSO₄ powder is irradiated in the reactor. Its absolute activity is measured using 4 π β - γ coincident counting technique. The active solid MnSO₄ powder is then dissolved in the solution of the inactive MnSO₄ path and stirred thoroughly to ensure homogeneity. Then the count rate is determined by the NaI (Tl) detector which is placed at the center of the tank (taking into consideration the decay time). This calibration is done for each tank. Thus the efficiency of the counting system could be well defined using the earlier data of the coincident technique.

The neutron source to be calibrated is then placed at the center of the tank containing the inactive MnSO₄ solution (after complete decay). The time of irradiation till saturation is about 16-18 hours. The source is removed and the solution is well stirred. The resulting activity was measured using the dip scintillation counting assembly. Thus the exact neutron emission rate of the neutron source can be derived after making the necessary experimental corrections. For precise measurements the experimental steps were repeated several times to

ensure its validity. Since the neutron yield depends upon Mn concentration, so the density of MnSO₄ solution is determined before and after each irradiation.

THEORETICAL CONSIDERATIONS

For calibration of neutron sources, the neutron source is placed in the middle of a cylindrical tank containing an aqueous solution of MnSO₄. The neutron irradiation is continued till complete saturation and the absolute activity of Mn-56 (gamma emitter) is determined. For production of Mn-56, it is known that the macroscopic neutron scattering cross-section for Mn in an aqueous solution is much larger than that due to absorption cross-section (case of fast neutron region). This means that most neutrons are slowed down to thermal energies (Mn capture cross-section is 13.2 barns). Also for H $\sigma_c = 0.3$ b, S $\sigma_c = 0.05$ b, and O $\sigma_c = 0.0002$ b. The emission rate (Q) of neutrons from the neutron source is calculated from the equation [16],

$$Q = R_{Mn} + R_H + R_S + R_O + R_L + R_{cont}. \quad (1)$$

Where:

R = rate of neutron capture in Mn, H, S, O, leakage and in container respectively.

R_O and R_{cont} can be neglected because of its small value.

S_O, this equation can be simplified to read

$$Q = R_{Mn} + R_H + R_S + R_L \quad (2)$$

The rate of emission of neutrons Q (n / sec) by some numerical analysis can read [17]

$$Q = \left(1 + \frac{1}{1.012} \frac{\sigma_S}{\sigma_{Mn}} + \frac{N_H \sigma_H}{N_{Mn} \sigma_{Mn}} \right) Q_{Mn} + R_L \quad (3)$$

The factor 1.012 takes into account the absorption of epithermal neutrons.

By substitution the numerical values for the cross-section in equation (3) becomes:

$$Q = (1.038 + 0.0257 X) Q_{Mn} + R_L \quad (4)$$

Where X = the ratio of concentration of H and Mn atoms in the bath (*i.e.* $\frac{N_H}{N_{Mn}}$)

$$Q_{Mn} = \frac{R_o}{\varepsilon}$$

Q_{Mn} = the activity of Mn – 56 in (dps)

R_o = the count rate at zero time after irradiation.

ε = the efficiency of the counting system.

So, equation (4) can be written as

$$Q = (1.038 + 0.0257 X) \frac{R_o}{\varepsilon} + R_L \quad (5)$$

To evaluate the neutron yield (ϕ) from the neutron source, the following formula can be applied [18]

$$Q = \frac{mN\rho N\phi}{A} (1 - e^{-\lambda t_1}) e^{-\lambda t_2} \quad (6)$$

m = mass of Mn in MnSO₄

N = Avogadro's number.

ρ = natural abundance of Mn 55 = 100%.

σ = Absorption cross-section of Mn.

ϕ = neutron yield (n / sec).

A = atomic weight of Mn-55

t₁ = time of irradiation.

t₂ = time of decay.

By substituting for the values of N, σ and A in the last equation, thus it will be [19]

$$Q = 0.0475 m \phi (1 - e^{-\lambda t_1}) e^{-\lambda t_2} \quad (7)$$

The relation between MnSO₄ density and its concentration can be estimated by empirical formula [20]

$$C = 14.2857 (140 d - 98.554)^{1/2} - 91.805 \quad (8)$$

Where: C = concentration of MnSO₄ solution.

D = its specific density at 25 °C.

RESULTS AND DISCUSSION

The absolute measurements of Mn-56 activity were counted by two standard techniques. These are 4 π β - γ coincidence and compared its results with 4 π β proportional counter. The assessment of the percentage error involved in the calibration of the two systems can be derived from the relation.

$$\% \text{ error} = \left(\frac{\text{coinc} - 4\pi\beta}{\text{coinc}} \right) \times 100$$

Five irradiated MnSO₄ solid samples have been measured using the two standard measuring techniques. The calibration result of the two systems can be summarized in table (1).

Table (1)

exp. no.	4 л	Coinc	Error	(%) error
1	2.778×10^3	3.032×10^3	$- 0.254 \times 10^3$	2.5 %
2	7.119×10^3	7.246×10^3	$- 0.137 \times 10^3$	1.9 %
3	3.794×10^3	4.153×10^3	$- 0.359 \times 10^3$	2.9 %
4	25.50×10^3	23.62×10^3	$+ 2.95 \times 10^3$	1.3 %
5	6.248×10^3	6.197×10^3	$+ 0.051 \times 10^3$	0.82 %

It can be seen from this table that for five different experiments, the absolute count rates derived by the β - γ coincidence system does not deviate by more than ($\pm 3\%$) from the count rate derived by the standard $4\text{л}\beta$ counter. This satisfactory result give the reason to proceed using the β , γ coincidence technique for absolute counting measurements. It is worth while to mention that by using the coincidence system, it could be avoid self absorption of activated MnSO_4 samples.

It should be mentioned that six cylindrical tanks of different radii (R) ranging from 15 to 30 cm are used. The characteristics of these tanks can be tabulated as in table (2)

Table (2)

Tank no.	Radius R (cm)	Height H (cm)	Vol. (cm)	Density (g/cm^3)	Percentage conc. C (%)
1	15	30	21195	1.0960	13.7656
2	17.5	35	33657	1.0954	13.6600
3	20	40	50240	1.0948	13.5799
4	22.5	45	71533	1.0935	13.3864
5	25	50	98125	1.0990	14.1942
6	30	60	169560	1.0620	9.0371

To find out the efficiency of the counting system (1.5'' x 1.5'' NaI crystal + photomultiplier + fast scaler) against the coincidence system the following experiment was performed. A solid MnSO_4 sample (0.3g) was irradiated in the reactor (2MW) for 13 minutes in the thermal coulumn at a distance 140 cm from the reactor core. The thermal neutron flux at this point was $5.1 \times 10^9 \text{ n}/\text{cm}^2/\text{sec}$. The obtained Mn-56 activity was about 15 micro curies. The irradiated sample was absolutely measured using the standard $4\text{л}\beta$ - γ coincidence counting system (taking into account the decay time). Then this irradiated sample was dissolved in the solution of the first MnSO_4 tank and was measured by the counting system (NaI detector) and corrected to zero time. On this basis the efficiency (ϵ) of this counting system was derived from the equation:

$$\varepsilon = \frac{R_o}{N_o} \times 100$$

where: ε = counting system efficiency

R_o = count rate from the NaI (TI) detector

N_o = count rate from the coincidence system

The absolute error of the efficiency was derived from the relation:

$$\Delta \varepsilon = \frac{R_o}{N_o} \sqrt{\left(\frac{\Delta R_o}{R_o}\right)^2 + \left(\frac{\Delta N_o}{N_o}\right)^2}$$

This experiment has been repeated for the six different $MnSO_4$ tanks. The result can be shown in table (3).

Table (3)

Tank no.	Radius R (cm)	Efficiency (ε) %
1	15	36.30 \pm 0.029
2	17.5	29.61 \pm 0.017
3	20	24.44 \pm 0.019
4	22.5	20.48 \pm 0.016
5	25	16.60 \pm 0.014
6	30	12.42 \pm 0.015

The variation of detector efficiency with variation of $MnSO_4$ tank size (in terms of its radius R) are Provided in Fig. (1)

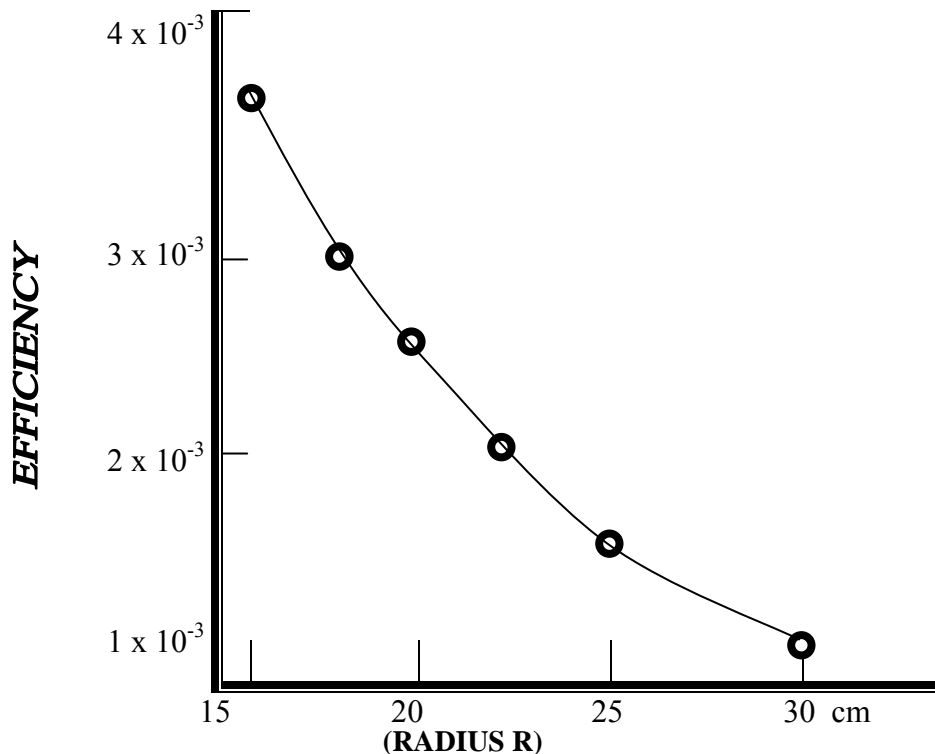


Fig. (1) Variation of the counting system efficiency (ϵ)
Against tank radius (R)

From this figure it could be concluded that the efficiency of the counting system decreases exponentially with the increase of tank radius.

To find the neutron leakage from MnSO_4 bath, a standard $R_a - B_e$ neutron source was located at the center of the bath. This source was left irradiating the MnSO_4 solution for a good time enough to secure complete saturation (16-18 hours). The neutron source was then removed and the MnSO_4 solution was stirred to guarantee homogeneous distribution of the radioactive Mn-56. The count rate of radioactivity from Mn-56 was determined by the pre-calibrated counting system (i.e of known absolute efficiency). This counting was repeated several times with intervals of 15 minutes and for a period equivalent to $3 T_{1/2}$ (about 8 hours). In each step the net count rate was derived and was corrected to zero time. The activity of Mn-56 was evaluated. So, the number of neutron absorption was estimated. By the knowledge of neutron yield from the standard neutron source, the percentage of neutron leakage can be estimated. The fraction of neutron leakage as well as neutron absorption in MnSO_4 tanks of different sizes can be shown in Table (4)

Table (4)

Tank no.	Radius R (cm)	Percentage absorption (%) R_{abs}	Percentage leakage (%) R_L
1	15	66.5	33.5
2	17.5	73.8	26.2
3	20	79.9	20.1
4	22.5	84.2	15.2
5	25	87.7	12.3
6	30	92.5	7.5

These data can be illustrated in figure (2). It could be concluded that the relationship between the percentage neutron leakage and the radius of $MnSO_4$ tank seems to be exponential with negative slope i.e neutron leakage decrease with increase of tank radius.

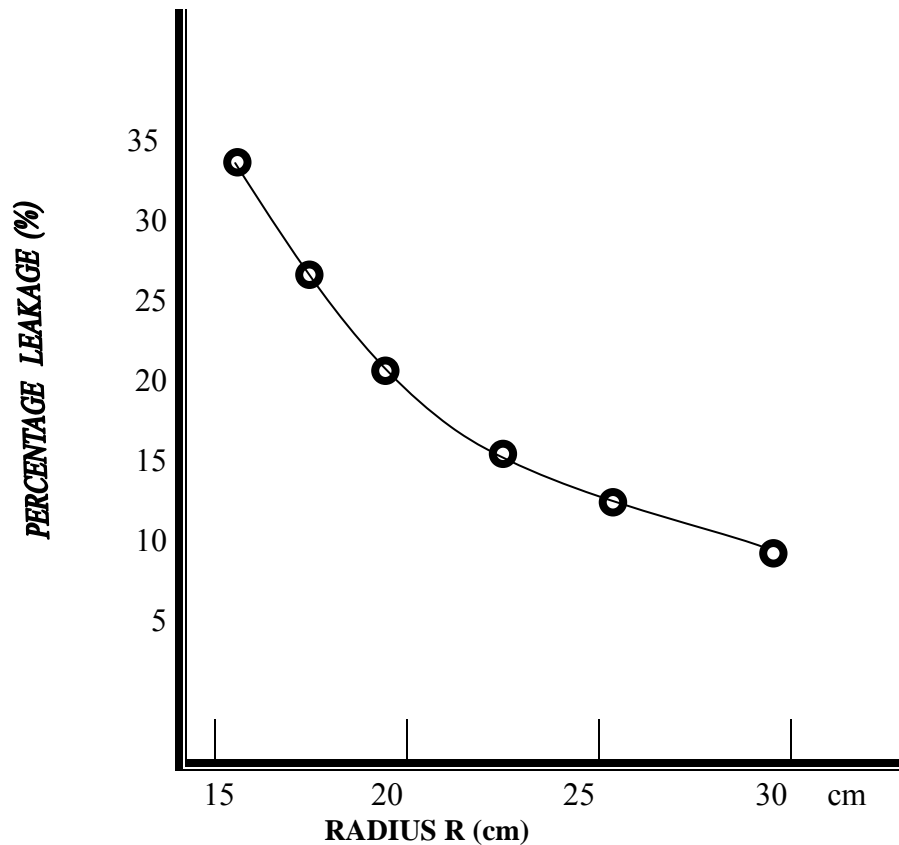


Fig. (2) Relation between percentage of neutron leakage against tank radius (R)

The density of MnSO_4 solution is one of the most important factor that affect the fraction of neutron leakage. This factor was investigated by the use of MnSO_4 tank of diameter equals height = 30 cm with different concentrations. The results are summarized in table (5)

Table (5)

Density (g/cm³)	Conc. (%)	Q_{abs} (%)	Q_L (%)
1.0792	11.4799	63.224	36.776
1.0876	12.6227	64.575	35.425
1.0961	13.6941	66.008	33.992
1.1045	14.8370	67.350	32.650
1.1129	15.9085	68.736	31.264
1.1264	17.1228	70.020	29.980
1.1298	18.3370	71.325	28.675
1.1382	19.3367	72.450	27.550
1.1467	20.3370	73.575	26.425

From this table it could be concluded that the amount of neutron leakage from the MnSO_4 solution during neutron source calibration was decreased by increase of solution density. The obtained data can be illustrated as shown in figure (3).

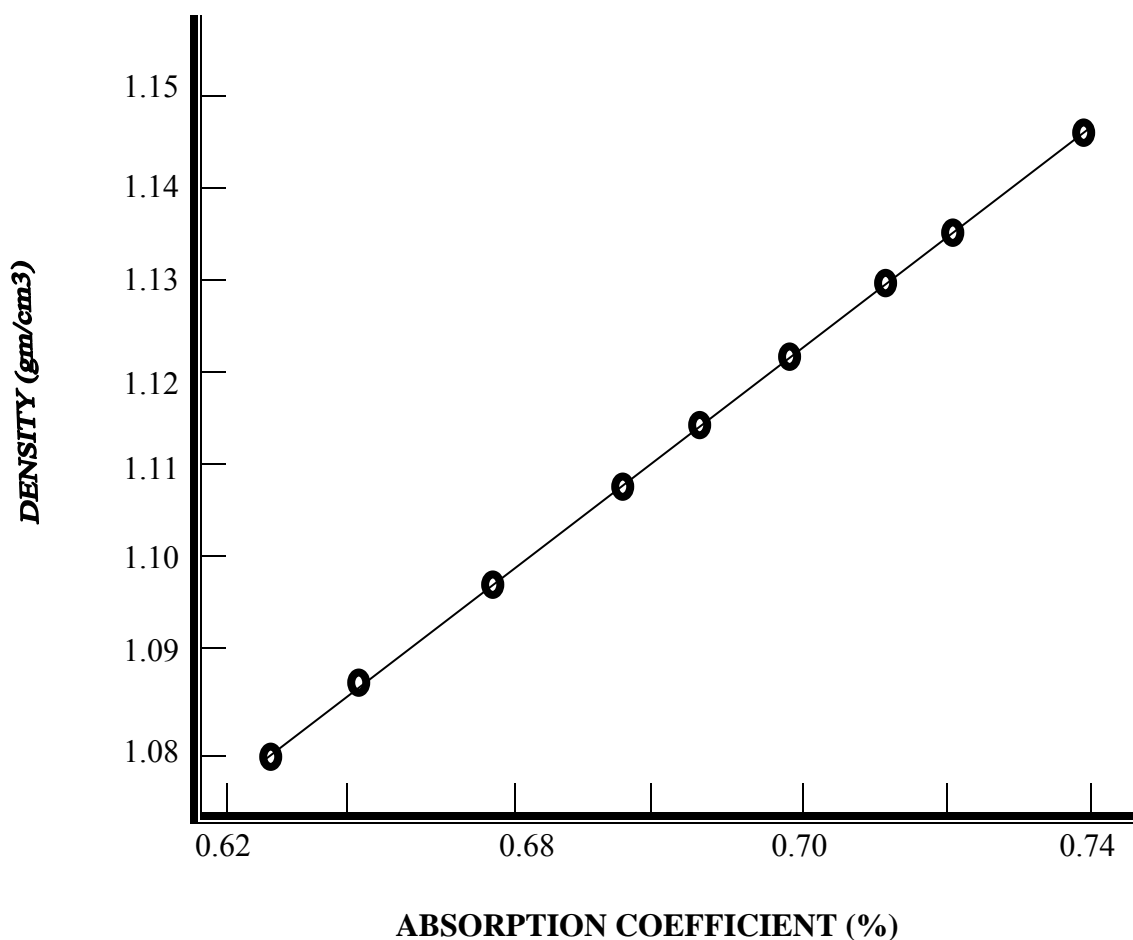


Fig. (3) Relation between neutron absorption in $MnSO_4$ Solution and its density.

It could be concluded from this figure that a linear relationship exists between the neutron absorption and the density of $MnSO_4$ solution with positive slope. This means that neutron absorption increases with increasing $MnSO_4$ concentration. For this reason in case of weak sources, it is recommended to use more concentrated solution in order to minimize the fraction of neutron leakage. Also when using a counter of poor efficiency it is advisable to use high concentration.

It may be of great interest to find out the importance of the size of the tank containing $MnSO_4$ solution. For this reason tanks of different sizes were used. The solution in each tank was irradiated separately by the neutron source which was placed at the center of the tank till complete saturation. The solution was then stirred to ensure homogeneity of Mn-56 activity. The detector was then placed instead of the source. The rate of counting of active manganese was taken every half an hour. This work has been repeated for the six tanks. The results could be illustrated in figure (4).

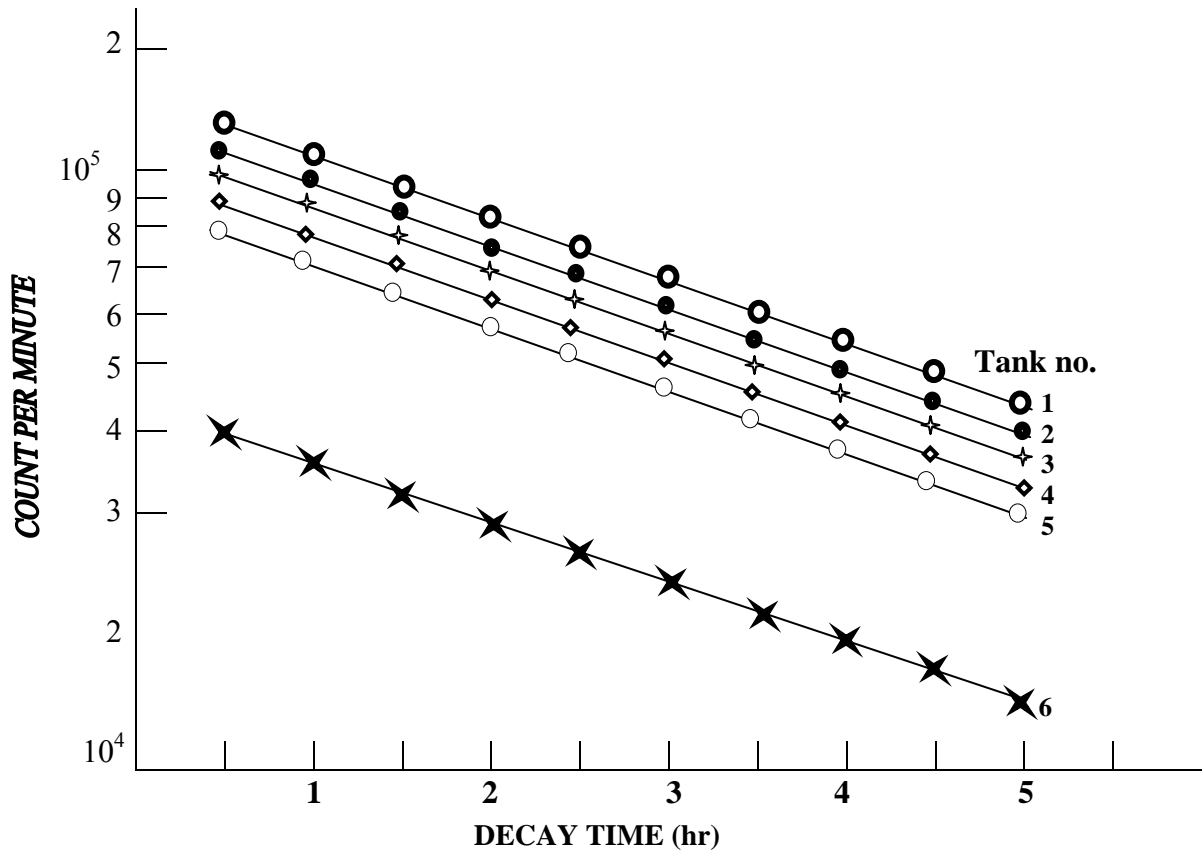


Fig. (4) Relation between the count rate of the detector and decay time for the different tanks of different sizes.

It is obvious from figure (5) that the count rate is high in case of small sizes than that of big sizes. This is in spite of high neutron leakage in small tanks than that in big tanks. For this reason it is recommended to use small tank for the calibration especially if the detector efficiency is small

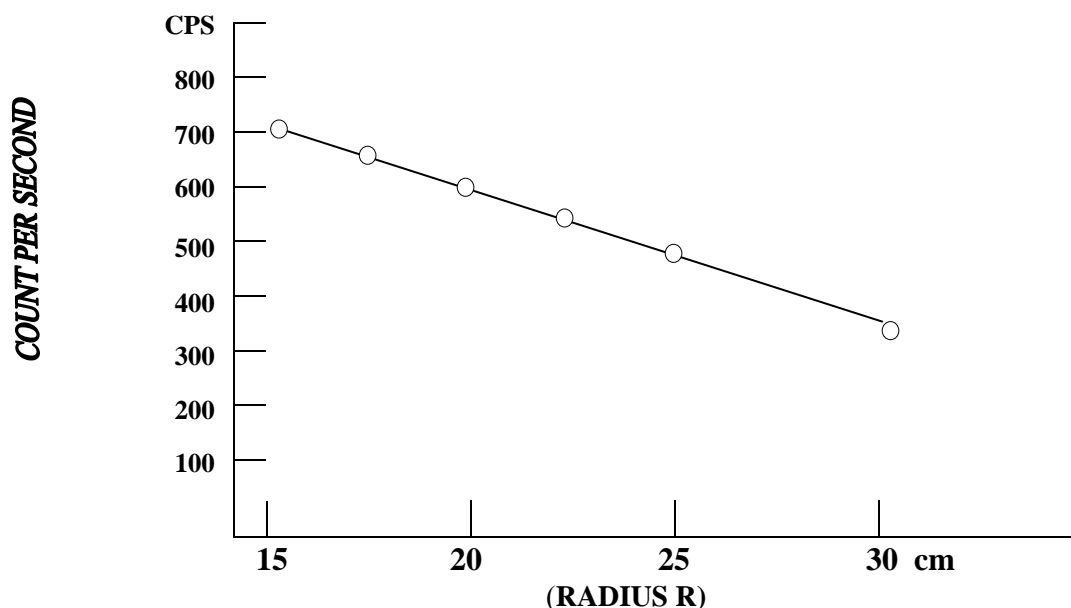


Fig. (5) Relation between the tank radius and the counting Rate corrected to zero time.

Calculation of absolute neutron yield from radium beryllium neutron source was carried out using MnSO_4 solution in six tanks of different sizes. This depends on the previous experimental findings such as: neutron leakage, detector efficiency and effect of MnSO_4 concentration. These results can be summarized in table (6).

Table (6)

Tank no.	Radius R (cm)	Absolute neutron Yield Q (n/sec)	Absolute error ΔQ (n/sec)
1	15	1.0460×10^6	0.0615×10^6
2	17.5	1.0948×10^6	0.0585×10^6
3	20	1.0915×10^6	0.0694×10^6
4	22.5	1.1332×10^6	0.0710×10^6
5	25	1.1160×10^6	0.0645×10^6
6	30	1.1299×10^6	0.0684×10^6
	Average	1.1016×10^6	0.0655×10^6

From this table it could be concluded that the numerical values of the neutron yield did not change significantly with the size of the manganese sulphate bath. Even the experimental errors are more or less the same. This data can be illustrated in figure (6).

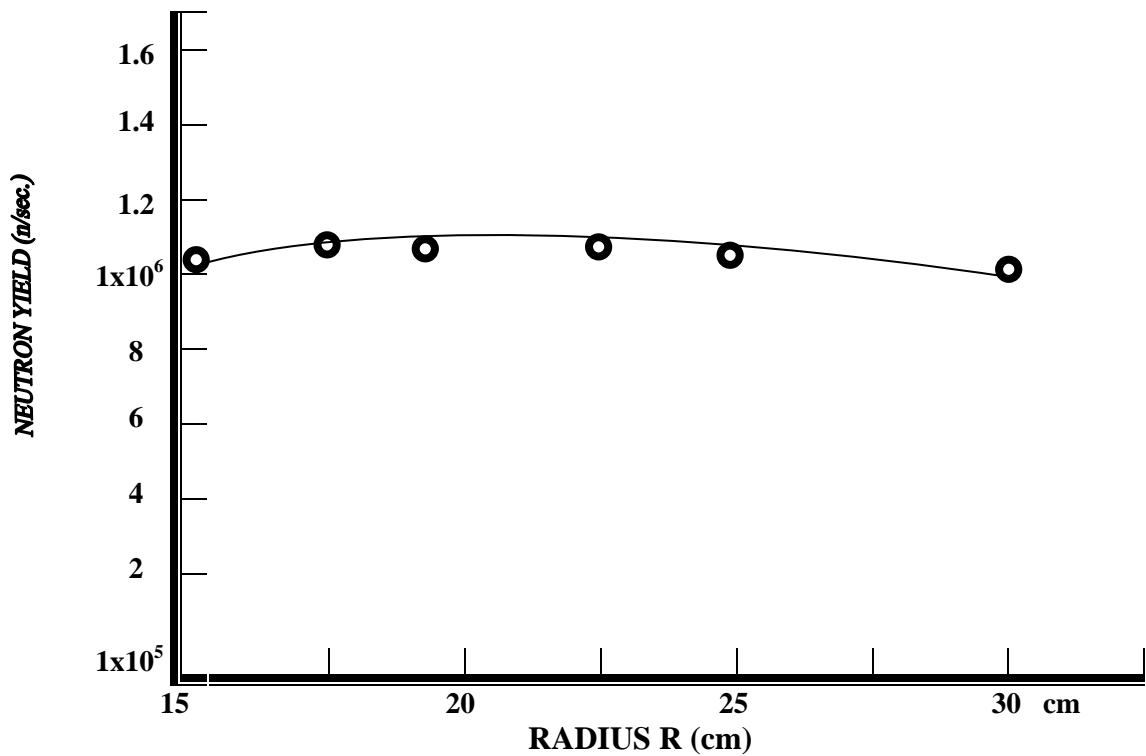


Fig. (6) Relation between the tank radius and the activity of the neutron sources.

It is shown from this figure that the neutron emission of the source are equal regardless the container dimension so, the average neutron yield from the used ($R_a - B_c$) neutron source using the suggested $MnSO_4$ baths gives rise activity equal to $(1.10 \pm 0.065) \times 10^6$ neutron per second.

CONCLUSION

Calibration of radium beryllium neutron source with manganese sulphate baths could lead to the following conclusions:

- (1) The efficiency of NaI (Tl) counting system was found to decrease exponentially by increasing $MnSO_4$ bath dimensions.
- (2) The neutron leakage within $MnSO_4$ tanks was found to decrease exponentially by increasing tank dimensions.
- (3) The neutron leakage was also found to decrease linearly by increasing $MnSO_4$ density.
- (4) The nominal losses of neutron yield from the neutron source can be attributed to:
 - (a) Fast neutron capture in sulphur and Oxygen in $MnSO_4$ solution.
 - (b) Thermal neutron absorption in the neutron source itself as well as its container.
 - (c) Fast neutron resonance capture in Mn.

- (5) Manganese sulphate baths of relatively small dimensions are generally recommended to be used for calibration purposes. This is because of:
- Relatively less consumed MnSO_4 salt necessary for preparation of saturated aqueous solution.
 - Ease of mobility with counting system.
 - Relatively higher counting rate.
 - Relatively higher efficiency with the counting system.
- (6) The neutron yield from the used ($R_a - B_e$) neutron source is equal to $(1.10 + 0.065) \times 10^6$ neutron per second. This result does not vary significantly from the international known neutron source of similar type.
- (7) This technique of calibration can be used for calibration of neutron sources of different neutron spectrum.

REFERENCES

- Anderson, H.L., Fermi E and Sziland L, Phys. Rev. Vol. 94, P 284 (1969).
- Johnson L.C., et al. Rev. Sci. Instrument, vol. 66, P 894 (1995).
- Nishitani T, et al. Rev. Sci. Instrument, vol. 63, P. 5270 (1992).
- Saha N.K. and Rangan L.K., Indidn Journal of Phys. Vol. 27, P. 18 (1953).
- Larsson K.E., Arkiv for Fysik., vol. 7, no. 4, P. 323 (1954).
- Tanimura Y. et al. Radiation Protection Dosimetry, vol. 110, P. 85 (2004).
- Baba M. et al. Nuclear Instrument and Methods, vol. A 367, P. 115 (1996).
- Larson K.E., J Nucl. Energy vol. 6, P. 322 (1958).
- Geiger K.W. and White R.B.- Canadian Journal of Physic vol. 37, P. 256, (1959).
- Zanki G. et al.- Nuclear Instruments and Methods. Vol. 185. P. 321 (1981).
- Yoshizawa M. et al. Journal of Nuclear Science and Technology, supplement 2, 1240 (2002).
- Andreev O.L. and Yaritzyna I.A. Atomnaya Energia, vol. 16, no. 3, P. 255 (1964).
- Murphey W.M. – Nuclear Instruments and method, vol. 37, P. 13 (1965).
- Mc Garry E.E. et al., National Bureau of Standard (US). Publ. 250, P. 18 (1988).
- Lamaze G.P. et al. Nat. Bur. Stand. (US) Publ. 250, P. 13 (1988).
- Izurni N. et al. Rev. Sci. Inst. Vol. 70, P. 1221 (1999).
- Nelson M.B. et al. Rev. Scientific Instrument vol. P. 1221 (1999).
- Thomas D.J. et al., Proc. Int. Conf. on Radiation Dosimetry and Safety-Taiwan, P. 285 – 289 (March 1997).
- Adams J.M.– Transactions of the American Nuclear Society, vol. 82, P. 106 (2000).
- Gurfinkel Y. and Amiel S. – Nucleonics, vol. 23, no. 3, P. 67 (1965).