DETERMINATION OF CORRECTION FACTORS RELATED TO THE MANGANESE SULPHATE BATH TECHNIQUE

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1. Introduction

Neutron sources have an irreplaceable role not only in the field of nuclear science but also in medical and industrial applications. Due to radioactive decay of isotopes used in the neutron sources the actual neutron source emission rate is the prime information for all consequent experiments. Nowadays, many technics of neutron source rate determination are available, but the manganese sulphate bath technique is the principal method. The principle of the emission rate measurement using the manganese sulphate bath technique is quite simple. The measured quantity represents the total neutron emission into 4π sr. The emitted neutrons are captured by the manganese in the solution to produce ^{56}Mn activity, which is subsequently detected to determine the source emission rate. However, not all emitted neutrons are captured by the manganese and the magnitude of these losses must be determined by either calculation or measurement. The decay of ^{56}Mn has the half-life of 2.5785 ± 0.0002 hours. Thus, from the measurements of the number of ^{56}Mn nuclei, the neutron emission rate can be calculated from the following formula:

$$Q = \frac{A_m M}{f(1-\delta)},$$ (1)

where Q is the neutron emission rate under 4π sr in neutrons per second, A_m is the ^{56}Mn mass activity of the bath at saturation in Becquerels per kilogram, M is the total mass of solution in kilograms, f is the probability of neutron radiative capture by the manganese of the bath divided by the probability of neutron radiative capture by all nuclei present in the solution (i.e., manganese, hydrogen, sulphur, oxygen and impurities). $\delta = O + S + L$ is the correction factor, where O are the (n,α) and (n,p) reaction probabilities with sulphur and (n,α) for oxygen, S is the recapture probability by the source itself and L is the fraction of the neutrons escaping from the solution tank [1-3].

The correction for thermal neutron capture represented by the factor f, excluded manganese nuclei, is independent of the neutron energy spectrum of the source, the solution concentration and the geometry of the system. This greatly simplifies its determination. Otherwise $1 - O - S - L$ correction would include three components, all of which would dependent on the source energy spectrum, solution concentration and the apparatus geometry. Monte Carlo (MC) simulations are a relatively easy way to calculate these corrective factors since this method is particularly suited to solving complex three-dimensional problems, which could be difficult or even impossible to solve analytically. As the amount of activated manganese depends on the system geometry and composition, each specific case has to be modelled and simulated. The presence of oxygen (in both water and MnSO$_4$) causes significant absorption of neutrons above 4 and Sulphur absorbs neutrons above 2 MeV. The correction considered for neutron absorption in the source and source holder is to account for the reduction of the manganese activity due to the loss of thermalized neutrons absorbed in

the neutron source itself. The last correction which has to be taken into account are neutrons escaping from the bath. In principle, the larger bath the less that escape. However, making the bath to large has compensating disadvantages. In particular, the specific activity of the solution decreases with increasing radius [4].

2. Experiment Description

The manganese bath technique involves placing the neutron source at the centre of a spherical tank, typically composed from aluminum or plexiglass material, of concentrated aqueous solution of pure or monohydrate manganese sulphate ($MnSO_4$). The tank shall be sufficiently large, to ensure almost complete absorption of emitted neutrons. Experimental systems around the world consist of spherical baths with various diameters. Available are results for baths with 50, 98 and 125 cm diameter. Reasonably, the new spherical bath should lie between those values, therefore we decided to design our manganese bath with diameter of 80 cm. Due to low mass activity philosophy, which decreases the requirements to shield the bath, only 5.9 kg of $MnSO_4.H_2O$ will be used for aqueous solution preparation during the first experiments. Then we reached the atomic ratio of N_H/N_{Mn} about 836, which corresponds to a solution density of 1.013 g/cm^3 [2, 5]. After reaching the saturation state, the radioactive solution was circulated at a flow rate of ~ 1 liter/minute through a Marinelli-beaker-type cylindrical container (~23 liters) equipped with 76x76 mm cylindrical NaI(Tl) detector (Amptek GAMMA-RAD) placed in the central hole. The full-energy peak calibration of the NaI(Tl) detector is based on the spectral analyses of manganese sulphate solution by a well-defined High Purity Germanium (HPGe) detector. Photopeak efficiency is than defined as a ratio of specific activity determined by HPGe detector to count rate determined by NaI(Tl). The primary energies of the gammas emitted from [56]Mn are 846.8 keV, 1810.8 keV and 2113.1 keV with 0.989, 0.269 and 14.2 PDF respectively but the spectrum can exhibit a backscattering peak, due to lead shielding placed around cylindrical beaker. The principal scheme of the experimental circuit is shown in Fig. 1.
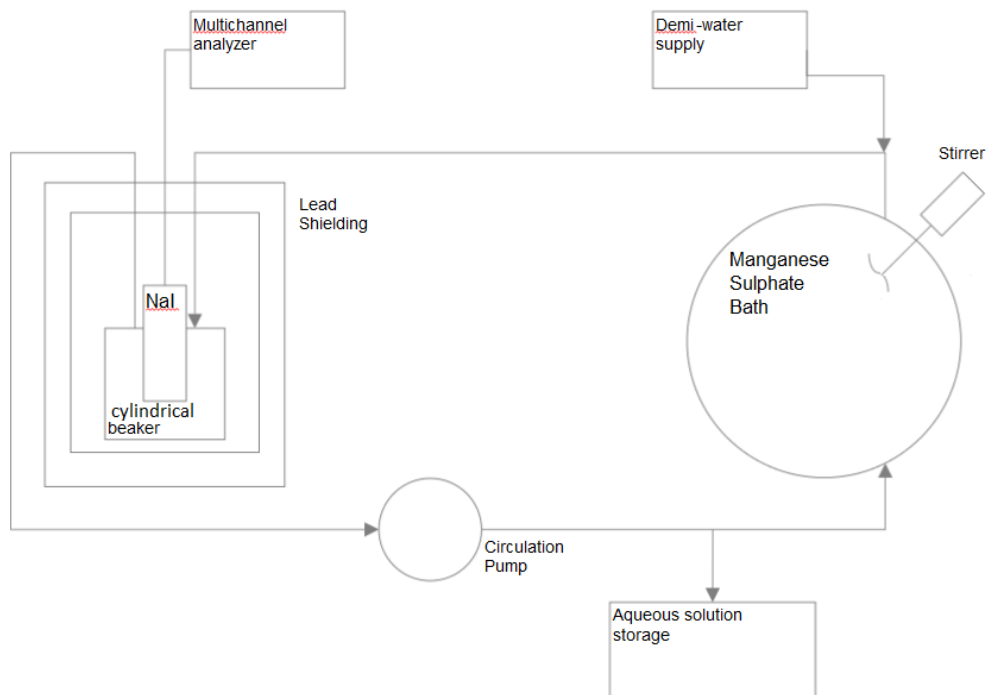

Fig.1: *Principal scheme of the Manganese Sulphate Bath system.*

In this work the SCALE code system and the Monaco code [5] was used to calculate the corrective factors for the Manganese bath and theoretical photopeak efficiency of the NaI(Tl) detector placed in non-standard cylindrical beaker. Monaco is a fixed-source Monte Carlo shielding code that calculates neutron and photon fluxes and response functions for specific geometry regions, point detectors, and mesh tallies. In addition, Monaco has variance reduction capabilities, such as source biasing and weight windows, which can be automated via the MAVRIC sequence (Monaco with Automated Variance Reduction using Importance Calculations). MAVRIC performs radiation transport on problems that are too challenging for standard, unbiased Monte Carlo methods. The intention of the sequence is to calculate the fluxes and the reaction rates with low uncertainties in reasonable times even for deep penetration problems.

The Pu-Be source was simulated as volumetric neutron emitter with strength of 1E5 n/s and neutron spectrum was taken from [6] and is shown in Fig 2. The source was situated to the centre of spherical geometry composed from two layers, manganese solution and plexiglass, to create the model of manganese sulphate bath. Neutron transport and reaction rates on Oxygen, Manganese, Sulphur and Hydrogen nuclei were calculated for this geometry to determine the correction factors and activity of the solution.
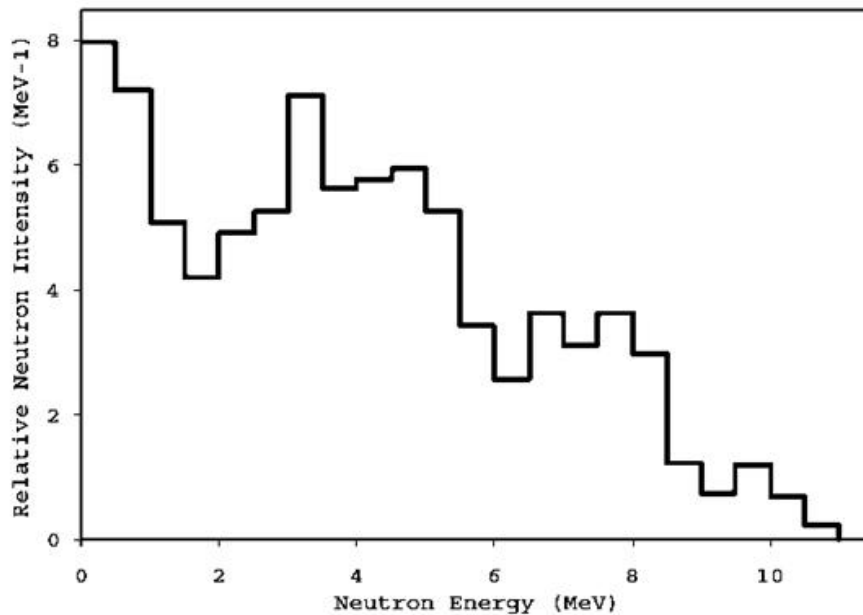

Fig.2: *Pu-Be neutron source spectra [6].*

Based on the calculated specific activity, the theoretical photopeak efficiency was calculated for the cylindrical beaker. The body of the beaker consist mainly from a simple cylinder with conical narrowing to the neck of beaker. The material of the beaker and the detector tube is made of aluminum. Model of the manganese sulphate bath and the cylindrical beaker is presented in Fig. 3. Due to lack of necessary information about the construction of the NaI(Tl) detector only crystal and simple aluminum coating was considered in the beaker model. In the efficiency calculation the volumetric gamma ray source for material of the solution was defined with discrete line spectra and emission probabilities for three primary energies [7]. The total gamma flux, the total gamma interaction and the photoelectric interactions were calculated within the crystal volume.
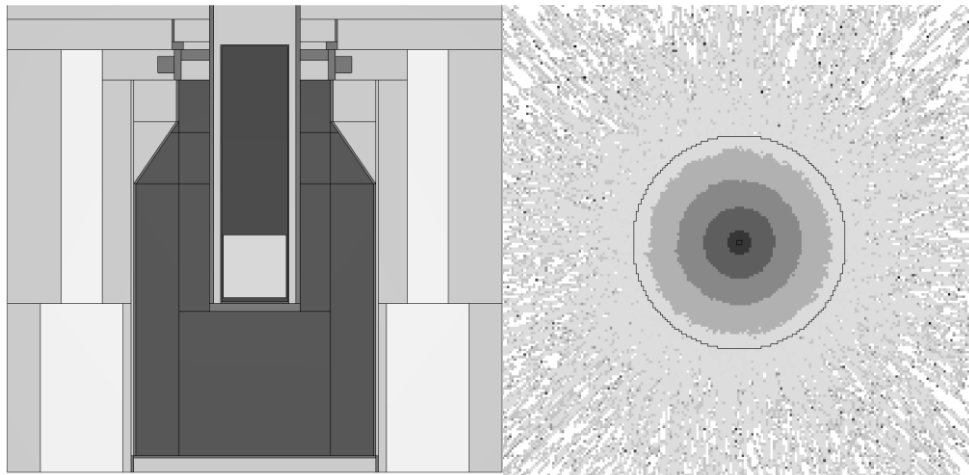
Fig.3: *Geometry model of the beaker (left) and neutron field in the bath (right).*

3. Discussion and Results

The correction factors presented in Eq.1 characterize all processes occurring in the bath. The neutrons emitted from the source are slowed down on Hydrogen nuclei and can be absorbed by Oxygen, Sulphur and also by Hydrogen. Afterwards thermal neutrons can be captured by all nuclei in the bath. The neutrons emitted with highest energies, which are thermalized close to the plexiglass shell or are not thermalized enough, may escape from the bath. On the other hand, neutrons scattered back to the source can be absorbed in its volume. These processes were simulated in MAVRIC calculation and the results in the form of neutron interaction probability vs. content of manganese in the bath are shown in Fig.4. The absorption on the Oxygen and the Leakage mainly depend on the spectra of the source and the geometry of the bath, therefore no dependence on the manganese content can be seen in Fig.4. The main neutron interaction, leading to neutron disappearance, in the presented range of manganese content, is the absorption on Hydrogen nuclei. This parasitic absorption was slightly compensated by absorption on Manganese in case of high content. This can be seen in Fig.4, as the decrease of the Hydrogen absorption fraction and the increase the Manganese absorption fraction. Since the Manganese absorption fraction is normalized to the neutron source strength, it can be considered as a correction factor representing the whole denominator of Eq.1.
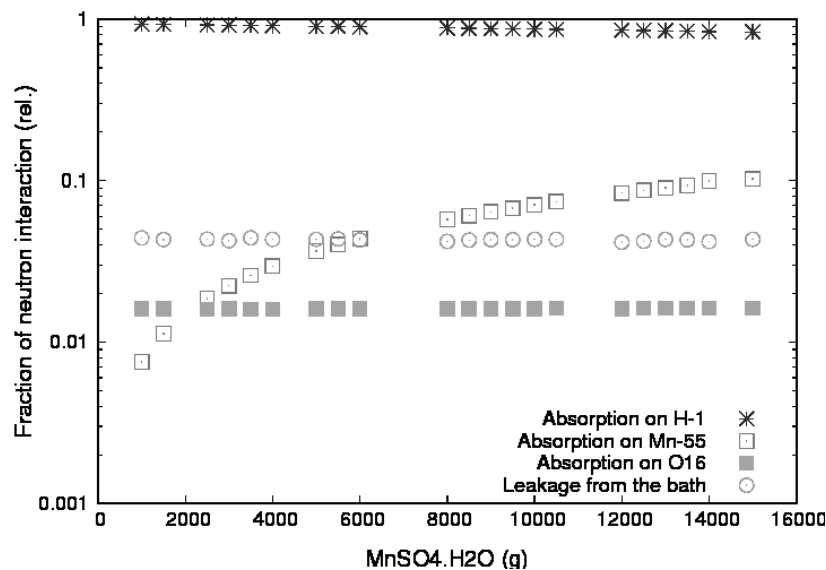

Fig.4: *Decomposition of the neutron interactions in the manganese bath.*

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Four independent experimental values of specific activities were determined. The first three were based on the measurement on HPGe detector. Due to construction of manganese solution circuit for continual measurement of activity, the circulation time is not negligible and the decay of the activated manganese should be considered. Therefore, the sample was taken from the circuit in front of and behind the cylindrical beaker. The third sample was taken under condition, when the circuit was turned off and the stirrer was used just for homogenization of the solution. The last value of specific activity was determined by the NaI(Tl) detector, based on the proposed experiment described in previous part. The results are presented in Tab.1. The lowest value was determined for the NaI(Tl) detector but the presented specific activity was achieved without efficiency calibration. The differences between activities determined by the HPGe detector were also observable. When we compared the values from the continual circulation the difference was around 6 %. Without appropriate number of measurements, to have better statistic, we cannot prove the influence of the decay during the circulation. The repetition of this experiment is necessary. More important difference was obtained between the mean value of specific activity with continual circulation and static activation. The first consideration predicted that the static activation should produce lower specific activity as the continual circulation, but the results presented in Tab.1 show 8% increase compared to the continual circulation. This difference will be used to determine the correction for continual mixing and decay within circulation. The ratio between the mean value of the specific activity of continual circulation and the value measured by NaI(Tl) detector defines the efficiency calibration of cylindrical beaker geometry. The Obtained value 1.42E-05 is comparable with the efficiency calibration used for the HPGe detector and the Marinelli geometry. The efficiency calibration constants were also calculated by the MAVRIC utility, but the value 8.68E-04 was overestimated compared to measurements. There could be a several sources of discrepancy, but probably the main source is that the calculated efficiency did not include the efficiency of the electronics and the photomultiplier behind the crystal, which were not as well considered in the model.

Tab. 1. *Specific activity of the manganese solution.*

Measurement option	Specific activity (Bq/cm^3)	
	Value	Unc.
HPGe before beaker	22.566	±0.284
HPGe behind beaker	23.911	±0.285
HPGe static activation	25.244	±0.298
NaI(Tl)	3.296E-4	±9.023E-6

4. Conclusion

This paper describes the fundamentals of neutron source emission rate measurement using manganese sulphate bath. This technique is known from the 60's but with the development of comprehensive computational tools it still seems to be the best available option. Determination of the neutron source emission rate with uncertainty around 1% is a challenging issue, especially for the laboratory without previous experience with this type of measurement. Several correction coefficients and calibration constants necessary for this experiment using experimental techniques or modern computational methods are presented in this paper. Although the consistency between calculated and experimental results is not sufficient, but with the increasing the number of carried out measurements and with application of statistic methods, better compliance could be found.

References:
[1]   Roberts, N. J.: MCNP Calculations of Correction Factors for Radionuclide Neutron Source Emission Rate Measurements using the Manganese Bath, Centre for Ionising Radiation Metrology, Teddington, (2001).
[2]   Ogheard, F., Chartier, J. L., Cassette, P.: Monte-Carlo simulation of the new LNHB manganese bath facility. *Applied Radiation and Isotopes*, **70**, 4 (2012).
[3]   Park, H., et al.: Absolute Measurements of the Neutron Emission Rate with a Manganese Sulphate Bath System. *Journal of the Korean Physical Society*, **47**, 4 (2005).
[4]   Roberts, N.J., Jones, L.N.: Recent development in radionuclide neutron source emission rate measurements at the NPL. *Applied Radiation and Isotopes*, Volume 68
[5]   D. E. Peplow, Monte, Monte Carlo Shielding Analysis Capabilities with MAVRIC, Special issue on the SCALE Nuclear Analysis Code System, Radiation Protection, Volume 174 (2011)
[6]   A. Elwyn: The Neutron Spectrum form a Pu-Be source, *The Neutron Physics Note 59*, (1986)
[7]   A. L. Nichols: Table de Radionucleides (Mn-56_tables), BNM – LNHB/CEA, France (2004)