MEASUREMENT OF THE NEUTRON EMISSION RATE WITH 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. Although nuclear reactors are the most prolific sources that produce neutrons with different ranges of energy, their applications are limited due to their high cost and non-transportability. 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. The manganese sulphate bath technique is a principal method for determination of the neutron source emission rate. This paper describes the fundamentals of this technique anda new experimental instrumentation developed to enable safe and rapid handling of neutron source during its transportation to manganese bath from storage container.

2. Emission rate measurement principle

The manganese sulphate bath technique is the principal method for the absolute determination of the neutron emission rate of radionuclide neutron sources. The main measurement principle is just special application of activation analysis and can be easily understood. A spherical bath is filled with a concentrated aqueous solution of highly pure $MnSO_4.H_2O$. The neutron source is placed at the center of the bath. The neutrons emitted from the source are then slowed down mainly by the elastic interactions on hydrogen nuclei. Next, thermalized neutrons are captured by the manganese nuclei (see Eq.1) and by nuclei of the other solution components. The thermal neutroncapture cross-section of oxygen nuclei is negligibly small and neutron capture by hydrogen and sulphur produces stable isotopes. Neutron capture by manganese nuclei produces radioactive ^{56}Mn nuclei which activity is then detected to determine the absolute neutron source emission rate into 4π sr.

$$^{55}Mn + n \rightarrow {}^{56}Mn \rightarrow {}^{56}Fe + \gamma + \beta^- \quad (1)$$

Mentioned above, the specific portion of thermalized neutrons is also captured by the nuclei of the solution components. If the spherical bath vessel has finite dimensions, some of the neutrons leak from the vessel, therefore both of the effects need to be assessed by the measurement or calculation of correction factors. The details and the individual evaluation of all correction factors can be found in various papers[1-3].

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 by the following formula:

$$Q = \frac{A_m M}{f(1-\delta)} \quad (2)$$

Where Q is the neutron emission rate under 4π sr in units of neutrons per second, $A_m$is the ^{56}Mn mass activity of the bath at saturation in units of Becquerels per kilogram, M is the total mass of solution in kilogram, $f$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. The correction factorδconsists of the (n,α) and (n,p) neutron reaction probabilities with sulphur, the (n,α) reaction with oxygen, the recapture probability by the source itself and by the fraction of the neutrons escaping from the solution vessel [1-3].

3. Experiment description

The manganese bath constructed at Institute of Nuclear and Physical Engineering and shown in Fig.1-a) is aspherical Plexiglas vessel composed of concentrated aqueous solution of monohydrate manganese sulphate ($MnSO_4.H_2O$) with hydrogen to manganese atom ratio N_H/N_{Mn} reaching the values between 30 and 340. The vessel should be sufficiently large to ensure almost complete absorption of emitted neutrons. Experimental systems around the world consist of spherical baths with various diameters (e.g. with diameter 50, 98 and 125 cm). Reasonably the new spherical bath should lie between those values, therefore we have decided to design our manganese bath vessel with diameter of 80 cm. Due to low mass activity philosophy which decreases the requirements to shield the bath, only 5 kg of $MnSO_4$ will be used for preparation of aqueous solution, leading to N_H/N_{Mn} atomic ratio 341 which corresponds to a solution density of 1.0459 g/cm^3 [2, 4].

a) *b)*

Fig.1:*Manganese sulphate solution vessel and Pu-Be storage container.*

Institute of Nuclear and Physical Engineering has four radionuclide neutron sources, two Pu-Be,one Ra-Be and one Am-Be. The Pu-Be source has been chosen for the first measurements, where the expected neutron emission rate reaches 1.0x10^5 n/s with the mean energy of emitted photons equal to 4.4 MeV [5]. This Pu-Be source produces neutrons as a secondary process, where the Pu isotope undergoes α-decay; this α-particle is absorbed by a light element (Be), which then decays by neutron emission.

$$^4He + {}^9Be \rightarrow {}^{12}C + n + \gamma(4.4\ MeV) \quad (3)$$

The Pu-Be isotopic neutron source is permanently stored in the storage container shown in Fig.1-b). In order to safely move this isotopic source from the transportation

container to manganese bath, a new experimental instrumentation including transportation container handled by crane was developed.

4. Shielding design for Pu-Be neutron source

Adverse effects of neutrons on the human body and some sensitive instruments are a driving force for the development of shielding design enabling the safe and rapid transportation of neutron source from transportation container to sulphate spherical vessel. The basic principle of neutron sources shielding design is well known. If a moderator with suitable thickness thermalizes fast neutrons as first layer, then a thin layer of a proper absorber can attenuate them sufficiently. In addition, since most of the materials generate gamma rays by absorbing neutrons, a layer of gamma ray absorbing material should be used in the system. The design of the new neutron source transportation container is based on these general principles and materials available at our institute.

In this work the SCALE code system and the Monaco code [6] is used to design source transportation container and to calculate expected effective dose map around shielded Pu-Be isotopic source. 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 fluxes and dose rates with low uncertainties in reasonable times even for deep penetration problems.

The source was simulated as neutron emitter with strength of 1E5 n/s and photon emitter of 5 943 photons/s with energy of 4.4 MeV. The source neutron spectrum was taken from [7] and is shown in Fig 2.

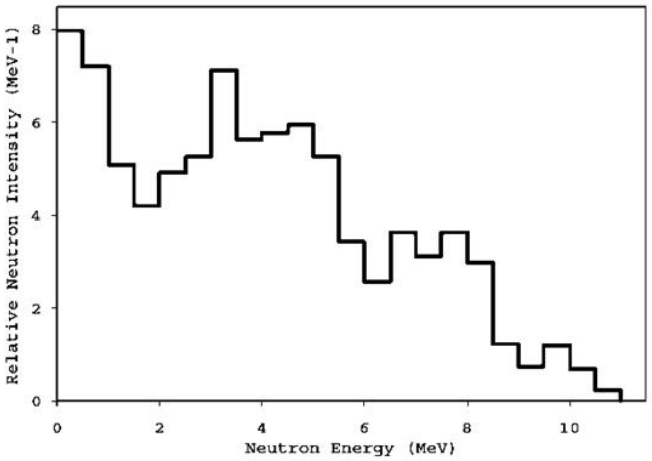
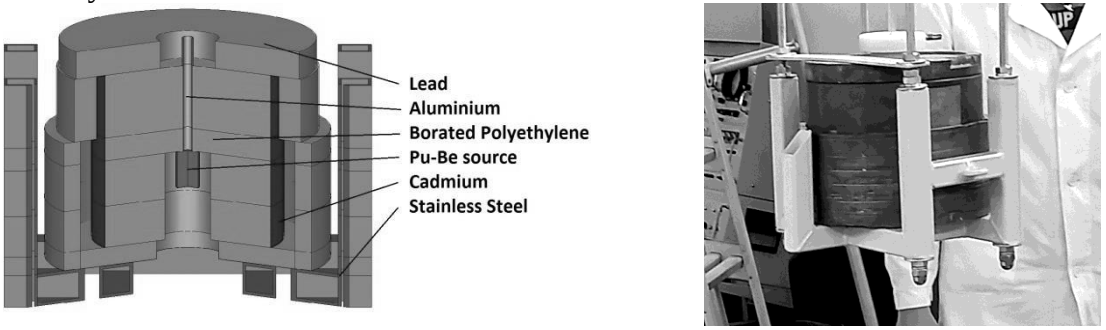


Fig.2:*Pu-Be neutron source spectra.*

Sufficient histories (700 batches per 50 000 neutrons) were run in all simulations in order to keep relative errors of computations below 10%. All of the statistical tests for estimated answers were passed. All of the calculations were carried out using 27 neutron and 19 group photon libraries based on ENDF/B-VII.0 [8] evaluated nuclear data.

Compact transportation container geometry in SCALE system is shown in Fig.3-a) and for comparison the real construction can be seen in Fig. 3-b). The basic configuration consists of a cylindrical Pu-Be source in the center of multilayer cylindrical shield made of moderator, neutron absorber and photon attenuation material. Source cylindrical geometry is modelled with 2.5 cm diameter and is 4 cm tall. Among many materials used as neutron

moderators, borated polyethylene (CH_2+ 5%$_{wt}$B) was selected since it is mainly composed of H and C and is cheap, accessible with reasonable mechanical properties and suitable as neutron moderator. As a thermal neutron filter we used two Cd 0.5 mm thick plates. As the gamma ray shield lead rings and plates were chosen due to their availability at our institute. The total mass of container is little above 100 kg and by using steel slings can be easily handled by crane.

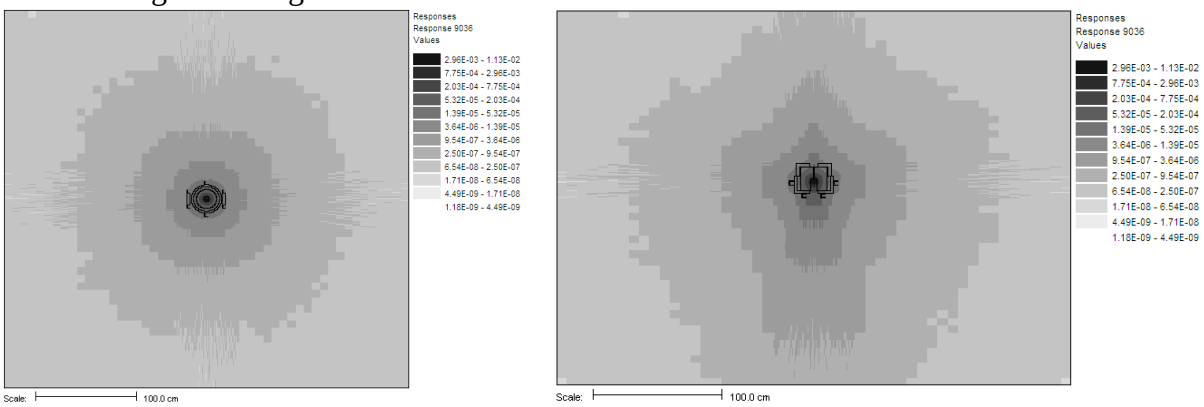


a)SCALE model *b)real construction*

Fig.3: *Transportation container geometry.*

5. Shielding design results

The two responses defined by ICRU-57(flux to dose conversion factors for neutrons in Table A.41; photons in Table A.17) were calculated by the MAVRIC sequence in the SCALE system. The first calculated response with ID 9036 stands for the effective neutron dose conversion factors in units of (Sv/h)/(neutrons/cm^2/s) and the second one with ID 9510 represents the effective photon dose conversion factors in units (Sv/h)/(photons/cm^2/s). The box 400x400x300 cm filled by air placed around the transportation container was used for definition of mesh grid geometry where horizontal axes consist of 50 bins and vertical axisare divided also to 50 bins. Through the geometry of transportation container we placed tighter calculation mesh with 0.5 cm steps in all axes. The results of MAVRIC calculations are shown in Fig. 4 and Fig 5.

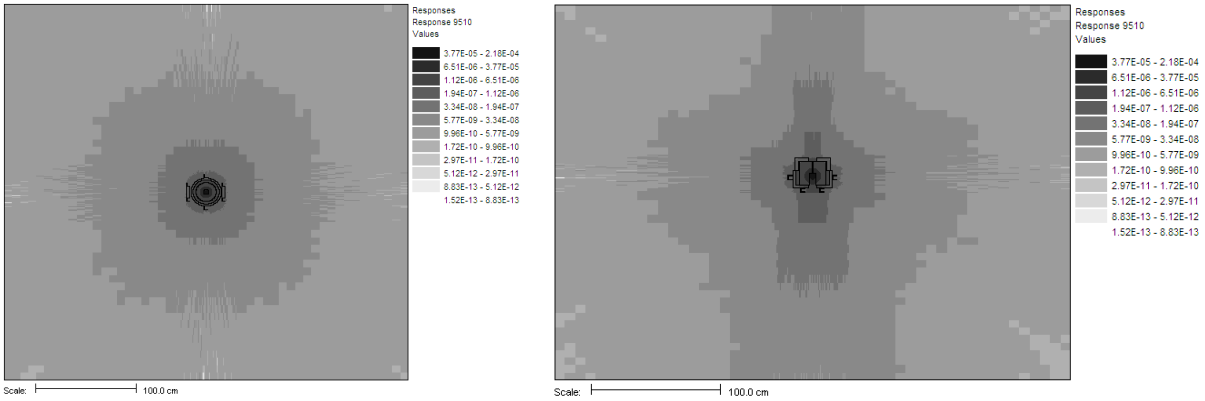


a) x-y plane *b) x-z plane*

Fig.4:*Mesh tally results – neutron effective dose.*

Fig.3 and Fig.4 show that the transportation container construction is symmetrical around z-axis and as expected, the effective neutron dose decreases with the distance from neutron source. The effective neutron dose on the surface of transportation container reaches almost 30 μSv/h, but in the distance of 50 cm the value decreases to 1.8 μSv/h. In the distance of 1 m from the Pu-Be source the calculated effective dose falls below 550 nSv/h.

The dose map in x-z plane shown in Fig.4-b) clearly proofs the presence of cavity in the bottom part of transportation container. There is no shielding material under the cavity; therefore the effective neutron doses are quite high even in farther distances. Namely the effective neutron dose in position 10 cm under the source reaches 16mSv/h, in 50 cm is equal to 4μSv/hcm and in 1m distance from source falls to 1.6 μSv/h.



a) x-y plane *b) x-z plane*

Fig.5: *Mesh tally results – photon effective dose.*

The data presented in Fig.5 belong to the effective photon dose near the source position. Photons around the source mainly originate from neutron interactions with boron and cadmium nuclides and are accompanied by photons directly emitted by the source.The calculated effective photon dose on the container surfacein radial direction reaches50μSv/h. In the distance of 50 cm the calculated gamma dose is approximately equal to 238 nSv/h and in the distance of 1m from the source the dose falls below the natural background. These values are symmetrical around the construction of transportation container.

It can be seen that transportation container provides effective barrier against ionizing radiation, however all radiation principles must be respected during the real work. From the results it can be seen that the best protection measure is to keep safe distance from the neutron source. This simple rule can easily decrease the potential personal exposure. To fulfill this rule the basic manipulation and stabilization of transportation container is performed by the simple handlers as it is demonstrated in Fig.6.

Fig.6:*The radiation decreases in proportion to the inverse to the distance squared.*

6. 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 the 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. To minimise the risk associated with exposure during moving of neutron source from the storage container to the manganese bath, the special transportation container was designed and constructed. The shielding abilities of this new container handled by crane were investigated for Pu-Be isotopic source. It was shown that the new container provides effective barrier against the gamma rays and neutrons emitted by the source. It should be noted that exposure can be also minimised by the physical distance from the source of radiation and by the effectively planned measurement process leading to minimal personnel exposure time. All of the principles mentioned above are actively used in development and improvement of whole measurement methodology at Slovak University of Technology in Bratislava. We believe that our further results may contribute to the world wide database of neutron emission rate measurements.

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