A MONTE CARLO-AIDED DESIGN OF A MODULAR ^{241}Am-Be NEUTRON IRRADIATOR

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Monte Carlo studies aimed at designing a modular ^{241}Am-Be neutron irradiator for testing neutron detectors and personal dosimeters and processing large volume samples are reported in this study.
The evaluation of the shapes and thicknesses of the moderator and shielding materials was carried out by a MCNP5 Monte Carlo simulation code. The reliability of the simulation was experimentally verified by the activation of gold foils and TLD dosimeter measurements in an irradiation cell placed at the center of the test configuration.

Key words: neutron irradiator, ^{241}Am-Be source, Monte Carlo method

INTRODUCTION

Neutron irradiation facilities are used worldwide to investigate material properties or to test and calibrate neutron detectors and environmental or personal dosimeters. An irradiator based on neutron sources can be more suitable for these purposes because of its stable neutron flux over time and the fact that, by means of a proper choice of sources, a specified neutron energy spectrum can be selected. These advantages may balance the drawbacks of an intrinsically low neutron flux value, even if it is many orders of magnitude lower than that of a nuclear reactor or a particle accelerator.

Many irradiators are based on radioisotope neutron sources (^{252}Cf, ^{241}Am-Be, ^{226}Ra-Be, or other sources) placed in fixed positions. Over time, their neutron flux characteristics are established. However, in some applications it may be very interesting to vary irradiation conditions and set the prevalence of thermal, epithermal or fast neutron flux components, *e. g.* in the calibration of dosimeters or neutron detectors. Irradiation facilities based on ^{241}Am-Be sources with these characteristics whose positions may be fixed or modified within a paraffin container so as to irradiate samples with a maximum diameter of 80 mm have already been proposed [1-3].

In this study, a Monte Carlo (MC) simulation was used to design a new modular facility based on four ^{241}Am-Be sources capable of both irradiating large volume samples and, also, modulating neutron flux components. MC simulation is a valuable tool for evaluating the shapes and optimal thicknesses of materials and for avoiding tedious calculations required to achieve design goals, such as obtaining neutron reference fields according to the ISO8529 [4]. Moreover, it takes into account both the physical parameters of the moderator and shielding materials (cross-sections, induced radioactivity and other), as well as the neutron source characteristics.

The experimental validation of the model was performed by comparison of the simulated and measured neutron flux values for a test configuration realized by using the said four sources of ^{241}Am-Be placed at fixed locations and by using water as a moderator and biological shielding.

The final setup of the irradiator can be used to study the response of detectors and dosimeters in various fields – such as neutron measurements around a reactor or a cyclotron, in neutron therapy, as well as for testing materials, electronic components and other devices.

MONTE CARLO MODELING OF A NEUTRON IRRADIATOR

Neutron transport simulation with Monte Carlo codes is used to verify the worth of various materials and for optimizing thicknesses. We used the MCNP (Monte Carlo N-Particle transport) code, version 5 [5], supplied by NEA and widely adopted to simulate the

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transport of neutrons and photon interactions, *e. g.* in [6-12]. It was implemented into the Windows operating system, to be used in personal computers with different computing capabilities (including that of the Intel Core Quad CPU).

Regarding the materials, we have considered water, graphite, polyethylene, polyethylene grains (of about 60% density of the compact polyethylene). Other possible moderators were excluded because of their high cost.

The neutron flux was estimated using tally F4 which calculates the average flux over a cell (particle per cm^2). The absorbed neutron dose rate was obtained through the tally F6 (MeV/g) which considers the average neutron energy deposition over cell volume, *i. e.* the energy deposited per unit of the mass of the material.

For the said MC simulation, the neutron energy range was split into following intervals: a group of thermal neutrons, *i. e.* all neutrons with an energy below the cadmium cutoff energy (0.5 eV), two epithermal neutron groups (between 0.5 eV and 0.1 MeV, 0.1 MeV, and 0.3 MeV) and three fast neutron groups (of 0.3 MeV to 1 MeV, from 1 MeV to 5 MeV, and above 5 MeV). The energy distribution from a ^{241}Am-Be neutron source was given by ISO 8529 [4], with the energy spectrum ranging from 0.025 eV to 12 MeV and an average energy of about 4.4 MeV. This was considered to be more helpful than the energy provided by the IAEA [13], as far as a more detailed description of the spectrum is in question. The energy groups presented were chosen to assess the effective dose outside the irradiator, according to the factors given in the ICRP Recommendation No. 103 [14].

The simulation analysis allowed us to optimize thicknesses and to study a number of combinations of materials while taking into account the introduction of air gaps between them. In particular, it has been shown that, from a physical point of view, the best moderator is graphite which allows a higher thermal neutron flux value at a thickness ranging from 4 to 6 cm. This is also confirmed by comparing the values of the moderating ratio $Q = \xi\Sigma_s/\Sigma_a$, a measure of the capability of the moderator to spread without absorbing a great number of neutrons, with ξ, the average logarithmic energy loss, Σ_a and Σ_s absorption and scattering cross-section (m^{-1}). A good moderator should have a value of Q as large as possible. The value of 192 for graphite is higher than the 71 and 64 values for water and high-density polyethylene, respectively [15]. As for graphite and grains of polyethylene, the maximum value of the thermal neutron flux is obtained with a slightly higher thickness (4-6 cm), while with high-density polyethylene and water the maximum thermal neutron flux component prevalence is obtained at a depth of about 3 cm. Nevertheless, for water and high-density polyethylene, the flux value is lower because of their absorption properties.

Since the largest uncertainty in MC modeling is associated with the lack of information on the structure of available ^{241}Am-Be neutron sources, except for a few data supplied by the manufacturer, a simplified test configuration (TC) to verify the validity of modeling was realized.

The TC consists of a PVC container and a plexiglas structure placed inside it. The sources were placed inside plexiglas tubes held by three square plexiglas plates (sides: 14 cm), while a plexiglas pipe placed at the center of the structure was used as an irradiation cell. ^{241}Am-Be neutron sources (each of 111 GBq activity, neutron emission: 7 10^6 s^{-1}) were realistically modeled as a cylinder (43 mm in length and 20 mm in diameter), coated with a metallic alloy. Am-Be neutron sources emit no penetrating gamma rays (~60 keV is the dominant gamma emission, strength 2.25 10^7 s^{-1} per GBq), except for the 4.438 MeV gamma rays resulting from the de-excitation of ^{12}C, the higher energy of the gamma spectrum, emitted with a strength of 0.57 gamma per neutron [16].

The remaining space inside the container was filled with water which acted both as a moderator and as a biological shielding. This type of configuration is characterized by a prevalence of the thermal neutron flux component. The schematic of the Monte Carlo model is given in fig. 1.

Our MCNP5 validation was carried out by irradiating thermoluminescent dosimeters (TLD600 and TLD700, in pairs) and 2 sets of 5 gold foils (diameter 1.3 cm, weighing approximately 0.6 g each), housed in a plexiglas sample holder, bare and covered with cadmium. The irradiation of TLD dosimeters lasted about 1 hour, 24 hours in the case of bare gold foils, and about 4 days in the case of cadmium covered foils. The gold samples were analyzed by gamma-ray spectrometry, using the system based on an ORTEC HPGe detector with a 60% relative efficiency described in [17]. The efficiency of the detection systems was determined at a distance of 25 cm from the point source of ^{152}Eu (with reference to 411 keV gamma emissions). Gamma ray spectra detected on the irradiated bare and cadmium-covered gold foils are reported in fig. 2.

The values of thermal and epithermal-fast neutron flux components in a TC irradiation cell were determined by applying the well-known *cadmium-difference* method. At the end of irradiation time (EOI), the activity (A_0) of a gold foil can be calculated by the relation

$$A_0 \quad \frac{C\lambda}{\varepsilon I(1 \quad e^{\lambda T_r})} e^{\lambda T_w} \qquad (1)$$

where, with reference to the 411 keV gamma emission of ^{198}Au, C is the photopeak area measured in real counting time T_r, ε – the full-energy peak efficiency, I – the gamma-ray emission probability, λ – the decay constant of ^{198}Au, T_w – the decay time between EOI and the start of counting. A_0 – the activities related to bare foils and cadmium-covered ones should be considered equal to

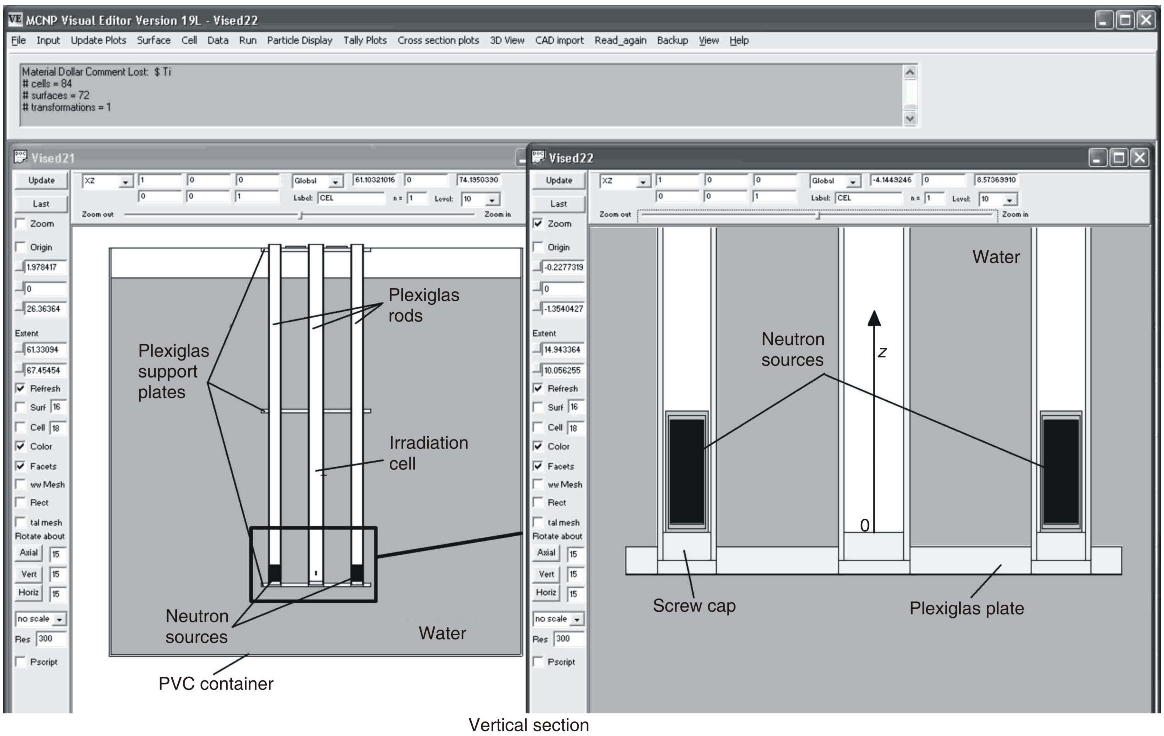

Figure 1. MCNP5 model of the test configuration with four sources placed inside plexiglas rods immersed in water

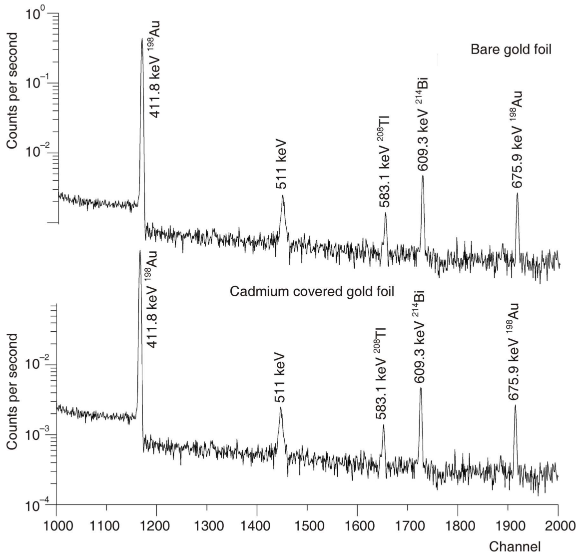

Figure 2. Gamma-ray spectra of irradiated bare and cadmium covered gold foils

$$A_{0(\text{bare})} \quad N_{\text{A}} \frac{m_{\text{bare}}}{M} \frac{\sqrt{\pi}}{2} \sigma_c^{\circ} \Phi_t \frac{I_\gamma \Phi_{\text{e}}}{15.2} (1 \quad \text{e}^{\lambda T_{\text{irr}}}) \tag{2}$$

$$A_{0(\text{Cadmium covered})} \quad N_{\text{A}} \frac{m_{\text{covered}}}{M} \frac{I_\gamma \Phi_{\text{e}}}{15.2} (1 \quad \text{e}^{\lambda T_{\text{irr}}}) \tag{3}$$

where, N_A is the Avogadro's constant, M – the atomic weight, m_{bare} and $m_{covered}$ are the masses [g] of the foils, σ_c° is the radioactive capture cross-section of gold at 2200 ms^{-1}, Φ_t and Φ_e are the thermal and epithermal flux, I_γ [b*] is the capture resonance integral of gold, T_{irr} – irradiation time, and 15.2 the averaging coefficient in the epithermal-fast neutron energy region (0.5-2.5 10^6 eV), assuming that the neutron flux shows a $1/E_n$ energy dependence.

The comparison between the neutron flux values derived from the simulations and the experimental ones is shown in fig. 3, with respect to relevant posi-

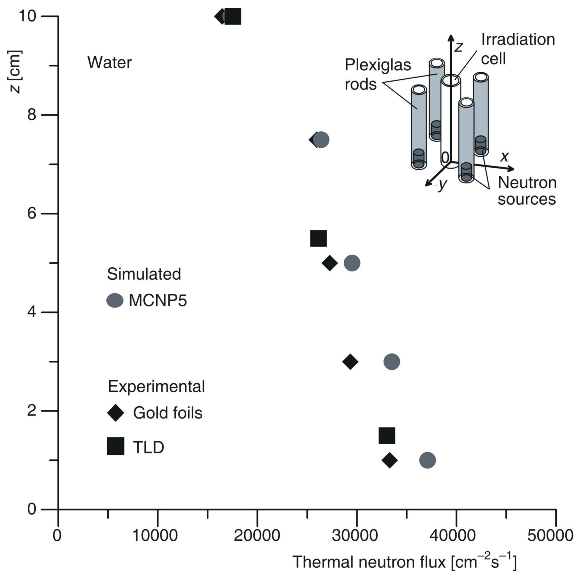

Figure 3. Thermal neutron fluxes with respect to position along the axis of irradiation cell of the test configuration schematically represented in the insert; MCNP5 simulation was carried out with four 111 GBq ^{241}Am-Be sources

*1 b = 10^{-28} m^2

tions along the axis of the irradiation cell. The neutron flux at these positions where the foils or the TLD were placed was modeled to take into consideration the flux perturbation due to their presence.

Measurements of the thermal neutron dose by means of gold foils and TLD dosimeters confirm the usefulness of the MC source-simulation model. Our study indicates that the largest discrepancy between the simulated and experimental values for water is lower than 15%. Thus, taking into account possible differences between the model and irradiation geometry due to the nature of the metal source containers, the thicknesses of the plexiglas source holder (3 mm) and the support structures of the pipes themselves, are to be considered acceptable.

FINAL DESIGN AND FIRST DOSE MEASUREMENT TESTS

A design process by means of a MCNP5 simulation was started taking into account the validated neutron source model, already mentioned modularity needs and certain requirements. The separation between the irradiation and source-recovery positions was specified as a priority in the design of our final configuration. In this manner, the safety requirements for the operators to securely make the necessary changes in the irradiation cavity with sources recovered and shielded in a suitable container are met. A further requirement is the possibility of obtaining a neutron flux characterized by thermal or fast neutron prevalence, simply by the easy positioning of components. The neutron flux within the irradiation cell should be as uniform as possible, so that a constant neutron flux density in the sample volume or the dosimeter to be irradiated can be assumed, apart from the sample self-attenuation that must also be taken into account.

Each of the sources must be easy to handle, without complex procedures for their recovery, while the size of the irradiation cavity should be sufficient enough to allow the placement of large samples/testing of individual dosimeters placed on anthropomorphic phantoms. From a radiation protection point of view, the constraint is that the maximum effective dose value outside the irradiator must not exceed the population dose limit value or be time-limited, whether the sources are exposed or recovered in a shielded box.

The configuration that best meets the mentioned requirements consists of an irradiation box and a separate source-recovery box allowing for the safe preparation of the required arrangement. MC simulation was used for the evaluation of the basic features of the configuration examined (thicknesses, fluxes, doses … and so on). For this purpose, several tests performed with the MCNP5 code were carried out (more than 100 runs, the minimum number of stories examined per each test: 1 10^8, total processing time not less than 1200 h, taking into account the time needed for particular personal computers). The schematic diagram of the final configuration of the modular neutron irradiator (MNI) is shown in fig. 4. The final draft is a result of a compromise between the best features designed by MCNP5 and what is required for the realization of the set goals. A high-density polyethylene was chosen as a moderator because the processing of parts, detachable and reconfigurable, is easier without the intervention of complex structures for their practical realization. Furthermore, it meets the constraints of a minimum overall size of the apparatus.

The irradiation cell of the MNI is obtained at the center of a polyethylene cube (sides: 80 cm) designed so that it is possible to change its geometrical configuration to generate either the fast or thermal neutron flux prevalence. Indeed, a large portion of the polyethylene moderator, including the cap, can be removed to irradiate large volume samples or test detectors on different phantoms or other supports. The dimensions of the cavity range between a minimum of 10 cm in diameter and a height of 15 cm, up to a hollow cube with sides of 40 cm (fast flux prevalence configuration). The sources are housed in pairs of two, properly spaced, handled by suitable source-container stainless steel rods and can be exposed for the time necessary to obtain a given value of the dose.

Irradiation time value can be set on a timer and, once it expires, electric motors will automatically provide the recovery of the sources.

The flexibility of the inner parts of the MNI allows for a selection of the source setup, so as to generate symmetric or asymmetric neutron fields by a combination of polyethylene bricks and one or two source rods. As a biological shielding, concrete bricks surrounding the sides of both irradiation and recovery containers, were used. As demonstrated by the simulation, the density of the concrete must be assumed to be equivalent to 2.3 g/cm^3, as a variation of density from 2.3 g/cm^3 to 1.6 g/cm^3 leads to an increase in the outside dose of more than 80%.

Neutron fluxes (thermal or fast) are almost uniform inside the cavity of irradiation. The behavior of predicted neutron fluxes (normalized to the number of neutrons emitted), evaluated by the MCNP5 simulation at certain representative points of the MNI irradiation cell, are given in figs. 5 and 6. Total and thermal neutron flux evaluations, 1.0 10^5 cm^{-2}s^{-1} and 5.25 10^4 cm^{-2}s^{-1}, respectively, are still many orders of magnitude lower than those pertaining to a nuclear reactor.

Our simulation results show that the average value of the effective dose rate outside the irradiation box is 8.9 μSv/h for the fast flux prevalence configuration and 1.2 μSv/h for the thermal flux prevalence. The neutron contribution to the dose is prevalent in the fast

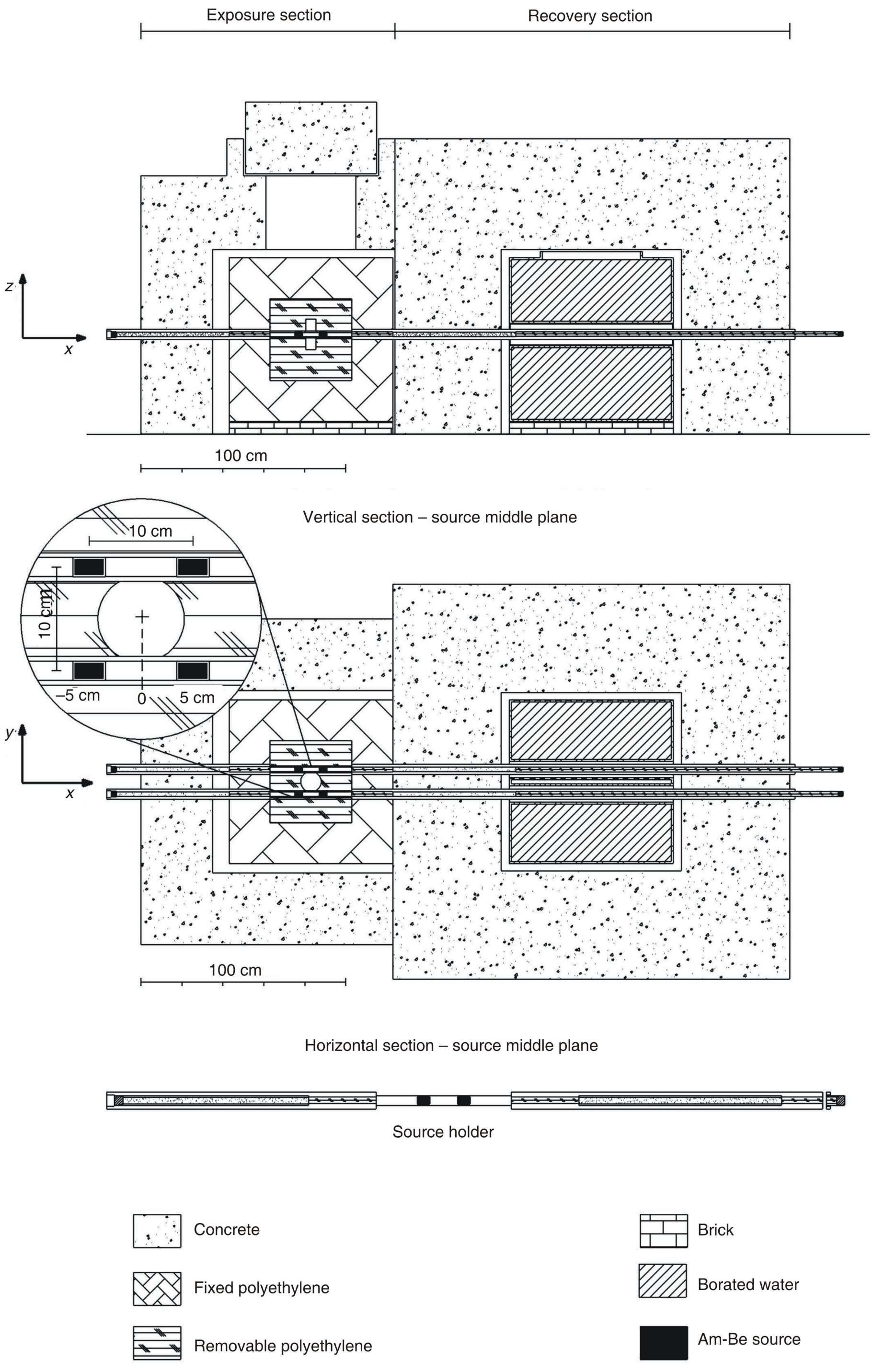

Figure 4. Schematic diagram of the final configuration of MNI

configuration (7.5 μSv/h), whereas gamma radiation is the more significant component in the thermal configuration (0.73 μSv/h).

The recovery facility consists of a special fiberglass container filled with borated water surrounded by thick concrete. A watertight opening in the container permits the crossing of source rods. On the outside of the recovery box, the effective dose rate on each of the faces of the cubic concrete shielding ranges between 0.12 (edges) up to 1.71 μSv/h (center). The final value leads to an annual effective dose lower than 0.5 mSv, with no limit on occupancy time around the facility.

The contribution of gamma radiation with dose values ranging between 0.007 to 0.06 μSv/h, mainly related to 2.23 MeV gamma radiation produced by the

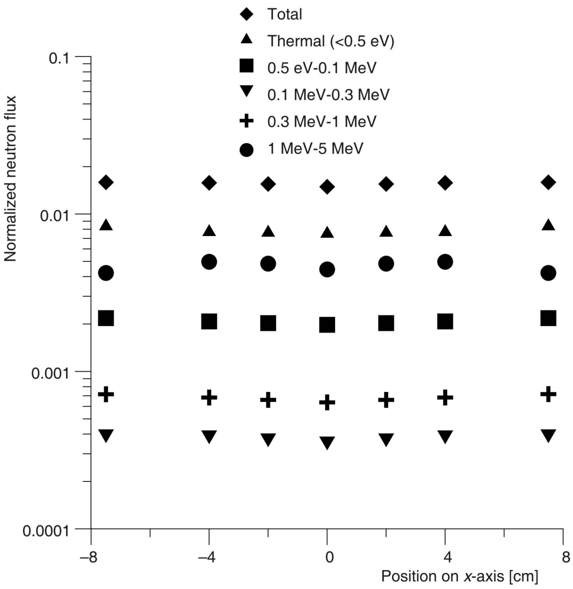

Figure 5. Behavior of predicted normalized neutron fluence in MNI with respect to distance along x-axis from the center of the irradiation cavity; the plane section considered does not include any source; MCNP5 simulation with empty cavity irradiation and neutron sources inside a compact polyethylene cube – thermal neutron flux prevalence

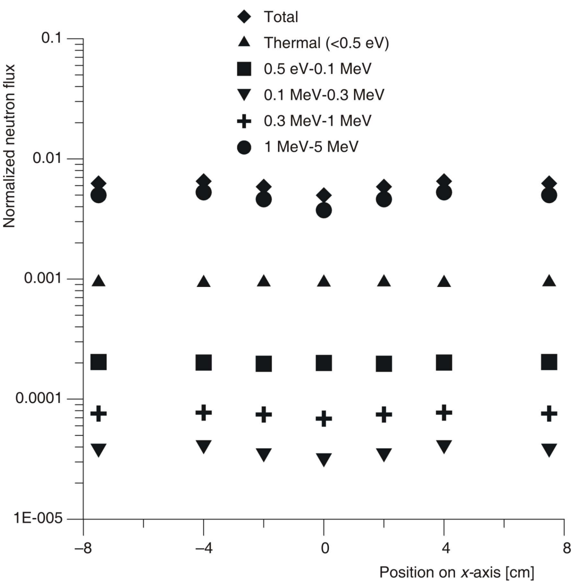

Figure 6. Behavior of predicted normalized neutron fluence in MNI with respect to distance along x-axis from the center of the irradiation cavity; the plane section considered does not include any source; MCNP5 simulation with empty cavity irradiation and neutron sources in air – fast neutron flux prevalence

radioactive capture by hydrogen in the recovery container, is hardly of any significance. These values were determined with a 3% boric acid concentration in water. It should be noted that a change in concentration of up to 4.5% does not lead to a significant decrease of the values of the effective dose rate, ranging from 2.3 μSv/h without boric acid (0%) up to 1.71 μSv/h (4.5%). A considerable decrease, instead, occurs in the gamma component, within the range of 0.74 μSv/h to that of 0.060 μSv/h (4.5%), probably due to an increase of neutron capture in water with respect to the concurrent radioactive capture reaction with hydrogen.

Numerical simulations are very useful for designing and verifying, as well as for optimizing tool configurations. In fact, in order to improve the main flux characteristics, MC simulation was also used to optimize the distance between the sources inside the rods. For symmetry, the sources were placed inside the median plane of the irradiation chamber, as shown in fig. 4, so that their positions could be varied inside the rods themselves. To verify the effect of the distance between the sources on neutron flux behavior along the *z*-axis of the irradiation cavity, some MC simulations were carried out by means of following configurations:

– sources placed at –5 cm and 5 cm positions along the source holder axis (distance between the sources: 10 cm; the origin of the source holder axis (0 cm) is the point corresponding to the axis of the irradiation cavity (see fig. 4),
– sources placed at –6 cm and 6 cm (distance between the sources: 12 cm), and
– sources placed at –7 cm and 7 cm (distance between the sources: 14 cm).

As highlighted in figs. 7 and 8, the configuration that provides a flow value higher at the center of the cell of irradiation is the first cited one (10 cm). However, at a distance of 12 cm or 14 cm, there was a more

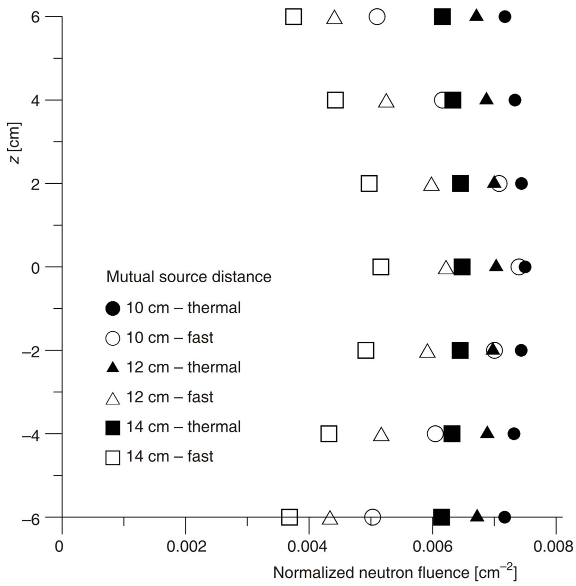

Figure 7. Behavior of MCNP5-predicted normalized neutron fluence in MNI with respect to distance on the axis of the irradiation cavity; MCNP5 simulation with empty cavity irradiation and neutron sources inside a compact polyethylene cube – thermal neutron flux prevalence

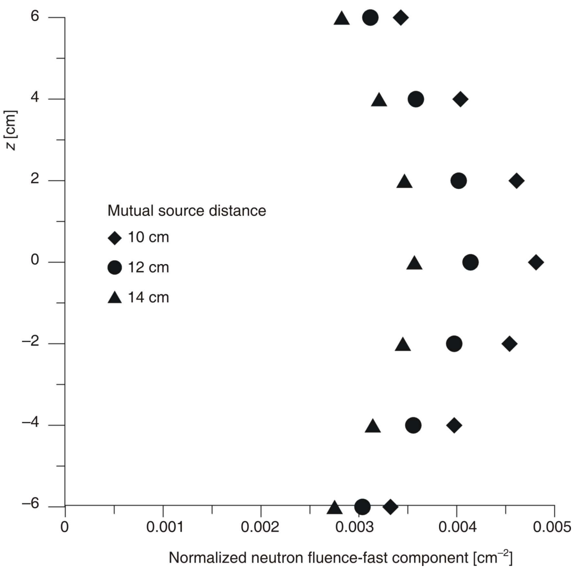

Figure 8. Behavior of MCNP5-predicted normalized neutron fluence in MNI with respect to distance on the axis of the irradiation cavity; MCNP5 simulation with empty cavity irradiation and neutron sources in air – fast neutron flux prevalence

pronounced discrimination between the thermal and fast neutron fluxes. In these instances, an optimized distance of 10 cm was chosen for the final configuration. However, the results of simulations with other distances should be taken into account on a case to case basis.

As a reference accident, a complete break with the emptying of the borated water container was assumed. Accident simulation analysis assesses dose values ranging from 15 to 138 μSv/h for neutron components and those from 0.3 to 2.3 μSv/h, for gamma components. Both ranges can still be considered acceptable, if both the probability of the occurrence of an accident and the operator presence-time near the irradiator are taken into consideration. However, to prevent an accident, a water level indicator monitors the recovery of the water level and an alarm is activated if the level is too low. In the latter case, an operator will transfer the sources into a third emergency container filled with water, thus allowing the technicians to safely repair the equipment.

To check the activation effects on polyethylene and stainless steel after 15 years of continuous irradiation, induced activities were evaluated. For both types of used steels, AISI 304 and AISI 316, induced activity values are significant for short-lived radionuclides (lower than 30 Bq for ^{56}Mn and 15 Bq for ^{59}Fe) while, in the case of ^{55}Fe, whose gamma emission energy is very low, the induced activity values in only 9 Bq. Spectrometric measurements of samples irradiated in the channel of the AGN-201 COSTANZA research reactor [18] for about 2 hours, with a thermal neutron flux of about 2.5 10^8 $cm^{-2}s^{-1}$, equivalent to a 1-year integrated dose of a neutron irradiator, highlighted only radioactive species with a short half-life.

Initial experimental dose tests were performed by irradiating a series of BD-PND and BDT bubble detectors (Bubble Technology Inc.), TLD dosimeters and gold foils. The mean value of 5 10^4 15% $cm^{-2}s^{-1}$ for the thermal neutron flux proved to be near the predicted value. For the fast configurations, suitable BD-PND detectors and CR39 dosimeters furnished an average value of 3.5 10^4 20% $cm^{-2}s^{-1}$, with an error of 26% in respect to the simulation value. To assess the experimental determination of neutron flux values to be considered as calibration data, further experiments and calibration procedures (as reported, *e. g.* in [19]) are also in progress.

CONCLUSIONS

The availability of a modular neutron irradiator allows the calibration of neutron detectors and dosimeters, as well as the testing of materials and devices outside the nuclear reactor. The flexibility of the irradiation facility regarding the parameters of the setting permits the achievement of favorable irradiation conditions, although absolute neutron flux values achieved are still many orders of magnitude lower than those of complex reactors or accelerator-based plants.

Monte Carlo simulation is, thus, confirmed as a useful technique for studying neutron irradiators with different configurations and the preliminary assessment of neutron flux characteristics originating from various sources.

By identifying the most promising facility assessment, the studied model can also be useful for planning future researches or particular applications of neutron irradiation.

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AUTHOR CONTRIBUTIONS

The simulation analysis with the MCNP5 code and the final design of the irradiator are to be attributed to P. Buffa. S. Rizzo provided the theoretical basis and nuclear data, as well as a critical review of the design procedure. Validation experiments were carried out by E. Tomarchio. For the most part, the manuscript was written and the majority of figures prepared by E. Tomarchio. All contributing authors analyzed and discussed the results reached.

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Пјетро БУФА, Салваторе РИЦО, Елио ТОМАРКИО

МОНТЕ КАРЛО ДИЗАЈН МОДУЛАРНОГ ^{241}Am-Be НЕУТРОНСКОГ ОЗРАЧИВАЧА

У овом раду приказано је проучавање усмерено ка дизајнирању модуларног ^{241}Am-Be неутронског озрачивача уз помоћ Монте Карло методе за тестирање неутронских детектора и личних дозиметара и обраде великих количина узорака. Урађена је процена облика и дебљине модератора и заштитних материјала уз помоћ Монте Карло програмског пакета за симулацију MCNP5. Поузданост симулације је експериментално потврђена активирањем златних фолија и мерењима термолуминесцентним дозиметрима у озраченој ћелији постављеној у центру конфигурације за тестирање.

Кључне речи: неутронски озрачивач, ^{241}Am-Be извор, Монте Карло метода