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Original Article

Characterization of neutron spectra for NAA irradiation holes in H-LPRR through Monte Carlo simulation

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A B S T R A C T

The Korea Atomic Energy Research Institute (KAERI) has designed a Hybrid-Low Power Research Reactor (H-LPRR) which can be used for critical assembly and conventional research reactor as well. It is an open tank-in-pool type research reactor (Thermal Power: 50 kW_{th}) of which the most important applications are Neutron Activation Analysis (NAA), Radioisotope (RI) production, education and training. There are eight irradiation holes on the edge of the reactor core: IR (6 holes for RI production) and NA (2 holes for NAA) holes. In order to quantify the elemental concentration in target samples through the Instrumental Neutron Activation Analysis (INAA), it is necessary to measure neutron spectrum parameters such as thermal neutron flux, the deviation from the ideal 1/E epithermal neutron flux distribution (α), and the thermal-to-epithermal neutron flux ratio (f) for the irradiation holes. In this study, the MCNP6.1 code and FORTRAN 90 language are applied to determine the parameters for the two irradiation holes (NA-SW and NA-NW) in H-LPRR, and in particular its α and f parameters are compared to values of other research reactors. The results confirmed that the neutron irradiation holes in H-LPRR are designed to be sufficiently applied to neutron activation analysis, and its performance is comparable to that of foreign research reactors including the TRIGA MARK II.

1. Introduction

The Neutron Activation Analysis (NAA) began to be widely known as an analysis method with good sensitivity at a time when science and technology that used research reactors was developing greatly. The NAA can be largely divided into the Instrumental Neutron Activation Analysis (INAA) and Radiochemical NAA (RNAA), and the determination of elemental concentrations based on the conventional INAA are usually made by the ratios between the specific activities of the target material and the Standard Reference Materials (SRMs) [1]. The method of comparing ratio can ignore the systematic errors generated from the self-shielding and multi-scattering effects of thermal and epithermal neutrons, but it has several disadvantages: the preparation of target material and SRMs under the same neutron irradiation conditions, enough space for SRMs in irradiation capsules/rigs, and the long time required to measure SRMs activities [2].

The k_0-standardization method was developed by F. De. Corte et al. to improve the accuracy and precision of the measurement data, and it is simpler experimentally but more complex in the concepts and procedures for calculation [3]. In the process of calculating the concentration of elements, the k_0-standardization method considers various input parameters: (1) thermal and epithermal neutron fluxes, (2) the deviation from the ideal 1/E epithermal neutron flux distribution (α), and (3) thermal-to-epithermal neutron flux ratio (f) [4]. Those input parameters can be evaluated by activation experiments and computer simulations, but the experiments using the activation foils should take into account the corrections for all neutron effects in order to decrease the systematic errors. Hence, the computer simulation has been widely applied to obtain these parameters, and especially Monte Carlo codes are mainly used for simulating the complex geometry and various neutron reactions [5].

Over 10 years, the Korea Atomic Energy Research Institute (KAERI) has developed a new concept reactor called the Hybrid-Low Power Research Reactor (H-LPRR) that can simultaneously serve as a research reactor and a critical assembly. Recently, the existing model has been modified to increase flexibility and cost efficiency, and its own safety functions have been strengthened to prevent power excursion due to the reactivity insertion accident [6]. The H-LPRR has 8 irradiation holes located outside the reactor core which can be

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used for medical Radioisotope (RI) production (e.g., ^{32}P, ^{99m}Tc, ^{131}I, and ^{198}Au) and NAA. In this study, the characteristics and performance for irradiation holes in H-LPRR were analyzed using some parameters applied in the k_0-standardization method, and those parameters were evaluated using the MCNP6.1 [7] and FORTRAN 90 language. Among them, a detailed analysis of α and f parameters was performed, and the performance of neutron irradiation holes was evaluated through comparison with values in other studies.

2. Core configuration of the H-LPRR

The H-LPRR was designed in an open-tank-in-pool type with 50 kW thermal power, a maximum thermal neutron flux of ~4.0×10^{12} n/cm^2-sec, and a light water coolant. The reactor core is filled with UO_2 fuel assemblies and two kinds of reflectors composed of beryllium and graphite, which are widely used in research reactors, and the reactor core is cooled by natural convection during normal operation. The fuel assembly consists of 3×3 square arrangements of UO_2 fuel rods, and the basic specifications (e.g., enrichment, radius, material, etc.) are similar to those used in OPR-1000 except for the axial length. The Control Absorber Rods (CARs) is filled with natural B_4C, and four CARs are symmetrically installed in the east, west, north, and south based on the center of the core in order not to deflect the distribution of neutron flux when controlling the core reactivity (see Fig. 1 (a)). Beryllium is used as an inner reflector, and they are surrounded by a graphite outer reflector canned with aluminum. The beryllium and graphite reflectors are placed on the grid plate without an auxiliary fastening device and are designed to be replaceable with the fuel assemblies. The irradiation holes in the reactor core are divided into two types: IR (for RI production) and NA (for NAA) holes, and in particular, NA holes are located in the left corner of the reactor core in consideration of the arrangement of the interior space and facilities of the building. Two aluminum tubes connected to the top of the reactor are installed inside the NA irradiation holes, and the polyethylene irradiation capsule moves inside the tube with a relatively large radius. In addition, the irradiation capsule is moved to the reactor core through pneumatic pressure and is designed to be located in the axial center of the reactor core with a relatively high neutron flux (see Fig. 1 (b)).

3. Determination method of α and *f* parameters

The distribution of epithermal neutrons is known to be inversely proportional to neutron energy, but in actual research reactors, deviation from the ideal 1/E distribution of the epithermal neutrons (α) may occur in the irradiation hole. Due to this deviation, an error may occur in the resonance integral of the neutron capture cross-section, and a $1/E^{(1+\alpha)}$ distribution of the epithermal neutrons has been introduced to correct this phenomenon [2]. To accurately estimate the effective resonance integral of the neutron capture cross-section, it is necessary to determine the parameter α for a specific irradiation hole, and the relationship between the resonance integral I_0 and the effective resonance integral $I_0(\alpha)$ is expressed by the following equation:

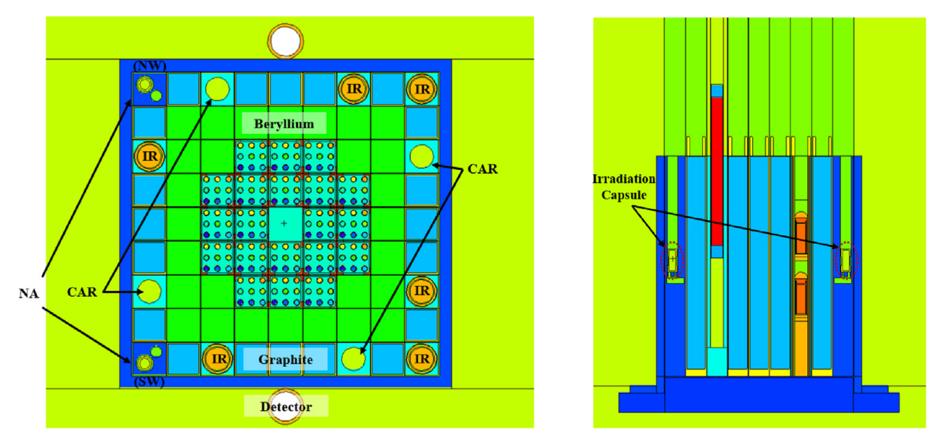
$$I_0(\alpha) = \frac{I_0 - 0.429\sigma_0}{(E_r)^{\alpha}} + \frac{0.429\sigma_0}{(2\alpha+1)\times(E_{cd})^{\alpha}} \quad (1)$$

where E_r is the effective resonance energy (in eV), σ_0 is the 2200 m/s neutron capture cross-section, E_{cd} (=0.55 eV) is the cadmium cut off energy, and α is the deviation from the ideal 1/E epithermal neutron flux distribution. $Q_0(\alpha)$ is the ratio of the resonance integral in a $1/E^{(1+\alpha)}$ epithermal neutron flux distribution to the neutron capture cross-section (σ_0), which can be presented, as follows:

$$Q_0(\alpha) = \frac{I(\alpha)}{\sigma_0} = \frac{Q_0 - 0.429}{(E_r)^{\alpha}} + \frac{0.429}{(2\alpha+1)\times(E_{cd})^{\alpha}} \quad (2)$$

Since the thickness of the irradiation sample and monitor are not negligibly thin enough to ignore changes in the neutron flux distribution, correction for the neutron self-shielding effect must be considered in the neutron activation analysis. The correction factors for thermal, epithermal, and resonance neutron self-shielding effects are defined as G_{th}, G_{epi}, and G_{res}, respectively, and these parameters are evaluated through a calculation using Monte Carlo codes or experiments. Moreover, the experimental methods for determining parameter α include bare, Cd-covered, and Cd-ratio methods developed by F. De. Corte et al., which are classified into dual or multi-monitor methods depending on the number of specimens used [8]. In order to accurately measure the parameter α, suitable monitors with effective resonance in the energy range (a few eV ~ a few keV) of the epithermal neutron must be selected/used, and the widely used monitors and their characteristics are shown in Table 1.

The shape factor (α) representing the non-ideal $1/E^{(1+\alpha)}$ epithermal neutron flux distribution is required for transforming the resonance integral to thermal cross-section ratio Q_0 into $Q_0(\alpha)$. When a set of n monitors are irradiated with and without Cd-cover, the parameter α can be obtained as the slope(-α) of the linear $\log T_i$ versus $\log(E_{r,i})$. The parameter T_i can also be determined through



(a) Horizontal Cross-section (b) Axial Cross-section

Fig. 1. MCNP model of the H-LPRR.

Table 1
Typical monitors and their nuclear data for Cd-ratio multi-monitor method [2,9,10]

Reaction	E_r [eV]	Q_0	G_{epi}	$T_{1/2}$
^{55}Mn(n,γ)^{56}Mn	468	1.053	1.0	2.579 h
^{59}Co(n,γ)^{60}Co	136	1.99	0.95	5.271 y
^{94}Zr(n,γ)^{95}Zr	6,260	5.31	0.982	64.02 d
^{96}Zr(n,γ)^{97}Zr	338	251.6	0.9728	16.74 h
^{197}Au(n,γ)^{198}Au	5.65	15.71	1.0	2.695 d
^{232}Th(n,γ)^{233}Th	54.4	11.53	1.0	22.3 m
^{238}U(n,γ)^{239}U	16.9	103.4	1.0	23.50 m

repeated least square fitting to the regression line (see Eq. (3)).

$$T_i = \frac{E_{r,i}^{-\alpha}}{(F_{cd,i}R_{cd,i} - 1)Q_{0,i}(\alpha) \times \frac{G_{epi,i}}{G_{th,i}}} \tag{3}$$

where, F_{cd} = Cadmium Transmission Factor,R_{cd} = Cd-ratio.

The parameter f is defined as the ratio of the thermal neutron flux and epithermal neutron flux and is taken into account when considering epithermal neutron activation. Also, it is generally

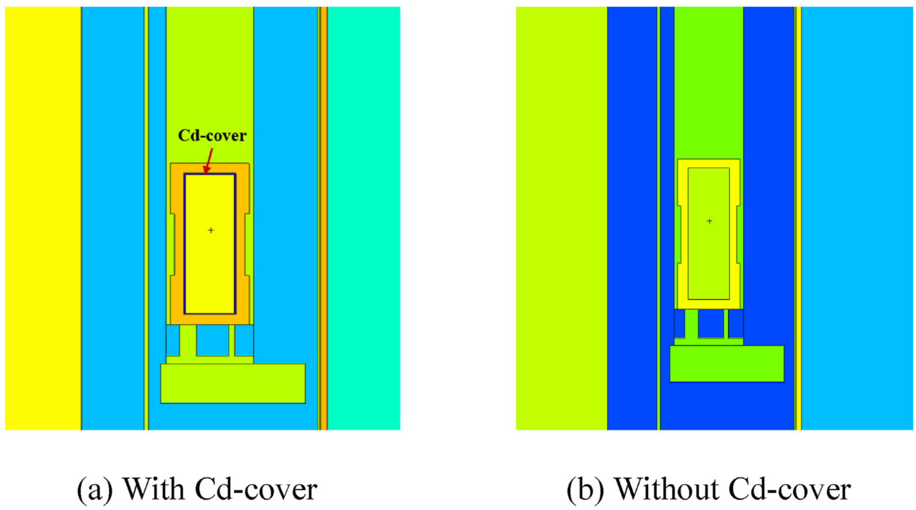


Fig. 2. MCNP model for NAA irradiation capsule.

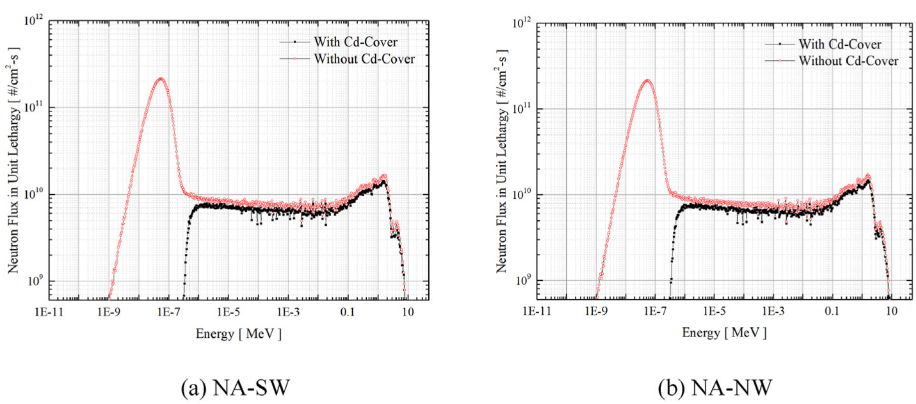


Fig. 3. Neutron flux distribution in NAA irradiation holes.

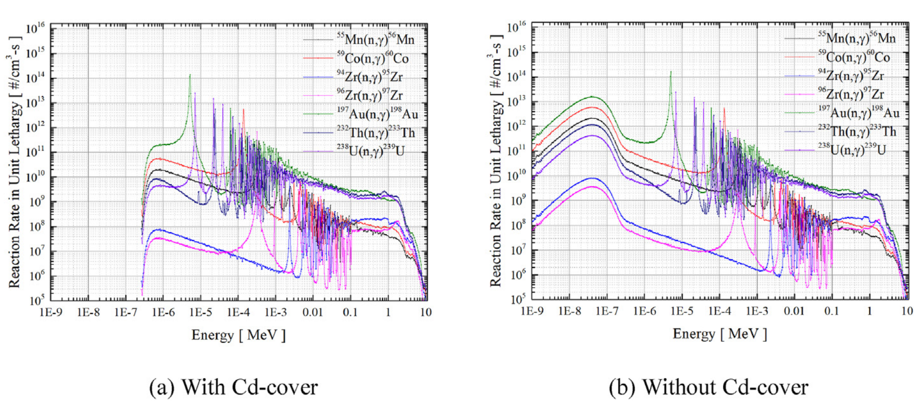
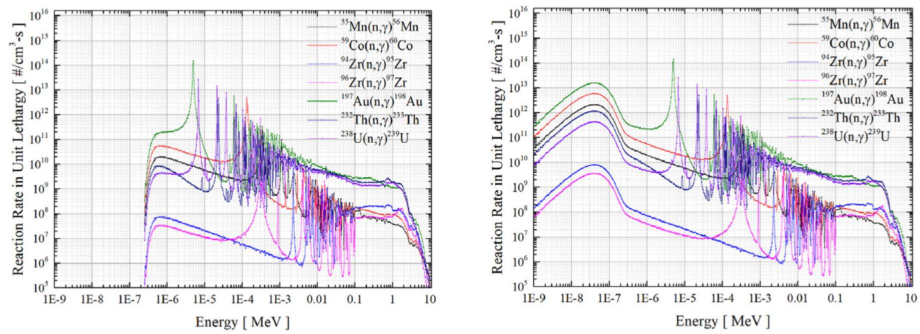


Fig. 4. Neutron reaction rate distribution in NA-SW irradiation capsule.

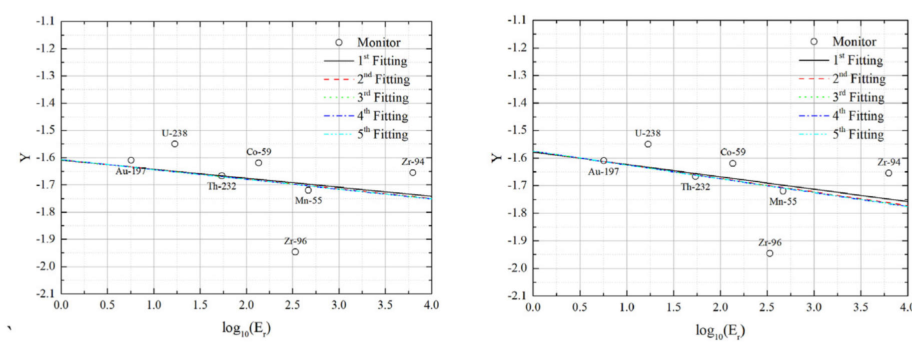


(a) With Cd-cover (b) Without Cd-cover

Fig. 5. Neutron reaction rate distribution in NA-NW irradiation capsule.

Table 2
Nuclear reaction rate and cadmium ratio of each monitor.

Reaction	NA-SW			NA-NW		
	w/ Cd [#/cm³-s]	w/o Cd [#/cm³-s]	R_{cd}	w/ Cd [#/cm³-s]	w/o Cd [#/cm³-s]	R_{cd}
^{55}Mn(n,γ)^{56}Mn	9.84×10^{10}	4.77×10^{12}	48.90	9.77×10^{10}	4.78×10^{12}	48.47
^{59}Co(n,γ)^{60}Co	5.87×10^{11}	1.35×10^{13}	22.37	6.07×10^{11}	1.36×10^{13}	23.06
^{94}Zr(n,γ)^{95}Zr	2.08×10^{9}	1.95×10^{10}	9.32	2.09×10^{9}	1.95×10^{10}	9.37
^{96}Zr(n,γ)^{97}Zr	2.68×10^{10}	3.65×10^{10}	1.36	2.66×10^{10}	3.62×10^{10}	1.36
^{197}Au(n,γ)^{198}Au	1.35×10^{13}	4.75×10^{13}	3.67	1.31×10^{13}	4.80×10^{13}	3.53
^{232}Th(n,γ)^{233}Th	6.31×10^{11}	3.18×10^{12}	4.94	6.50×10^{11}	3.21×10^{12}	5.03
^{238}U(n,γ)^{239}U	2.26×10^{12}	3.16×10^{12}	1.44	2.22×10^{12}	3.19×10^{12}	1.40



(a) NA-SW (b) NA-NW

Fig. 6. Iteration Calculation for α Determination.

Table 3
α and *f* values of NAA Irradiation Holes at Some Research Reactors.

Country	Reactor	Power	Irradiation hole	α	*f*
South Korea	HANARO	30 MW	NAA #1	0.097	1,148
	H-LPRR	50 kW	NA-SW NA-NW	0.036 0.050	40.49 37.56
Japan	JRR-3M	20 MW	PN-2 PN-3	0.140 0.043	188 2776
Vietnam	TRIGA MARK II (Dalat)	500 kW	Rotary Rack Channel 13-2	0.073–0.040	35.7 21.5
Belgium	Thetis (INW)	150 kW	Channel 3 Channel 15	0.015 0.084	25 72
Malaysia	TRIGA MARK II	1 MW	Rotary Rack	-0.010	22.6
Hungary	WWRS (KFKI)	10 MW	Mila	0.015	32

determined using the Cd-ratio (R_{cd}). Au is mainly used as the monitor, but other monitors (Co, Zr, etc.), which have well-known nuclear data, are also widely applied in the analysis. The parameter *f* can be derived using the equation presented below, and in practice, $f = 10^{-(y\text{-intercept})}$ can be evaluated using the y-intercept value of the trend line that determines α.

$$R_{cd} = \frac{(A_{sp})_{bare}}{(A_{sp})_{cd}} = \frac{[\phi_{th}\sigma_0 + \phi_{epi}I_0(\alpha)]}{F_{cd}\phi_{epi}I_0(\alpha)} \quad (4)$$

where $(A_{sp})_{bare}$ = Specific Count Rate for an Interesting γ-peak without Cd-cover $(A_{sp})_{cd}$ = Specific Count Rate for an Interesting γ-peak with Cd-cover.

4. Calculation results and discussion

The MCNP6.1 code with the ENDF/B-VII.0 library was used for characteristics evaluation of NAA irradiation holes in H-LPRR, and the main components in the reactor core, including irradiation holes and capsules, were specifically reflected in the 3-D MCNP model. Fig. 2 shows the vertical cross section of the irradiation capsule in the NA-SW irradiation hole, and it is possible to check the model difference between the case where the Cd-cover (Thickness: 0.5 mm) is included in the irradiation capsule and the case where the Cd-cover is missing. This model was used to calculate the thermal and epithermal neutron flux distributions and the neutron reactions with various monitors, and a sufficient number of neutron histories are used for a MCNP run to obtain results with reasonable stochastic uncertainties. About 170 million neutron histories (4,200 active batches and 400,000 histories per batch) are usually used, which results in a fractional standard deviation of 0.002% in k_{eff} and 1% in neutron flux distributions and neutron reactions.

The neutron flux distributions were analyzed in a relatively small cylindrical volume (NAA irradiation capsule) with a diameter of 2.5 cm and a height of 6 cm by applying the F4 tally function in the MCNP code. The energy range is divided into 600 groups from 0.001 eV to 10 MeV, and the calculated results for each irradiation hole are shown in the Fig. 3. Since NA-SW and NA-NW holes are symmetrically located in the reactor core, it can be confirmed that the neutron flux distributions in each hole are almost the same.

Figs. 4 and 5 show the distribution of the neutron reaction rate generated in NA-SW and NA-NW irradiation capsules. As shown in the figure, the resonance peaks of the seven (n,γ) reactions are uniformly distributed in the epithermal neutron energy region. Moreover, the effective resonance energy peak ($E_r = 5.65$ eV) of the ^{197}Au(n,γ)^{198}Au reaction exists in the lowest energy region, whereas the effective resonance energy peak ($E_r = 6{,}260$ eV) of the ^{94}Zr(n,γ)^{95}Zr reaction is located in the highest energy region (see Table 1). The total reaction rate and cadmium ratio (R_{cd}) of each nuclear reaction are shown in Table 2. It can be seen that the NA-SW and NA-NW irradiation capsules have almost the same values, and the R_{cd} values range from 1.36 to 48.90. In particular, the R_{cd} of the ^{96}Zr(n,γ)^{97}Zr is the lowest, while the R_{cd} of the ^{55}Mn(n,γ)^{56}Mn is the highest.

Fig. 6 shows the fitting curves for α determination. As shown in the figures, the iterative fitting calculations converge in five times, and the convergence condition is less than 0.1% of the slope change. Table 3 presents the values of α and *f* in the NAA irradiation holes of the H-LPRR and foreign research reactors. The α values for the NA-SW and NA-NW irradiation holes of the H-LPRR are 0.036 and 0.050, respectively, which are positive and indicate a soft epithermal neutron spectrum. Moreover, their *f* values ($f_{\text{NA-SW}} = 40.49$ and $f_{\text{NA-NW}} = 37.56$) show that the neutrons at these irradiation holes are not sufficiently thermalized by beryllium and graphite reflectors/moderators, so the contribution of the epithermal neutrons to the (n,γ) reaction is quite significant. Nevertheless, the values of α and *f* of NAA irradiation holes in the H-LPRR are expected to be sufficiently utilized for NAA as the same level as those of other irradiation holes in foreign research reactors [2].

5. Conclusion

The neutron spectrum characteristics of NAA irradiation holes (NA-SW and NA-NW) in the H-LPRR were analyzed by applying the k_0-standarization method. In particular, we determined the spectral properties such as α (the deviation from the ideal 1/E epithermal neutron flux distribution) and *f* (thermal-to-epithermal neutron flux ratio) of the neutron flux distribution using the Cd-covered method. The MCNP6.1 code with the ENDF/B-VII.0 library was applied for accurate simulation of the distributions of the neutron flux and the nuclear reaction rate for seven monitors (Mn-55, Co-59, Zr-94, Zr-96, Au-197, Th-232, and U-238). The results showed that the parameter α values of NA-SW and NA-NW irradiation holes were 0.036 and 0.050, respectively, and their *f* values were 40.49 and 37.56, respectively. These values were similar to those of foreign research reactors, and the H-LPRR are expected to be sufficiently utilized for neutron activation applications. Moreover, the Monte Carlo calculation of the neutron flux distribution in irradiation holes can provide detailed information for spectral characteristics analysis and is considered a good method for verifying experimental results.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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