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Original Article

Determining PGAA collimator plug design using Monte Carlo simulation

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ABSTRACT

The aim of this work is to help inform the decision for choosing a convenient material for the PGAA (Prompt Gamma Activation Analysis) collimator plug to be installed at the tangential channel of the Moroccan Triga Mark II Research Reactor. Two families of materials are usually used for collimator construction: a mixture of high-density polyethylene (HDPE) with boron, which is commonly used to moderate and absorb neutrons, and heavy materials, either for gamma absorption or for fast neutron absorption. An investigation of two different collimator designs was performed using N-Particle Monte Carlo MCNP6.2 code with the ENDF/B-VII.1 and MCLIP84 libraries. For each design, carbon steel and lead materials were used separately as collimator heavy materials. The performed study focused on both the impact on neutron beam quality and the neutron–gamma background at the exit of the collimator beam tube. An analysis and assessment of the principal findings is presented in this paper, as well as recommendations.

1. Introduction

Prompt Gamma Activation Analysis (PGAA) is a very widely applied technique to simultaneously qualify and quantify the elemental composition of unknown samples, ranging in mass from micrograms to a few grams. This non-destructive technique typically measures in several minutes to several hours per sample. The possibility of detecting a small amount of light elements in a matrix comprised of high-Z elements is another characteristic [1,2]. The shape of the analyzed sample is a comparatively low matrix dependency.

The PGAA instrument comprises, in addition to the beam collimator, the following main elements: i) the primary beam shutter, which consists of two sections dedicated to opening and closing the neutron beam; ii) the supermirror neutron guide, which mainly comprises multilayer-coated mirrors (glasses) treated with Ti and Ni; and iii) the beam shaper connected to the end of the supermirror guide for controlling the beam size, depending on sample size. The input beam size measures 2.5 cm × 10 cm, while three output sizes are available, namely 2.5 cm × 2.5 cm, 2 cm × 2 cm, and 1 cm × 1 cm. In order to collect prompt gamma rays emitted by irradiated samples, a 25% high-purity semiconductor germanium detector (HPGe) is used. The beam stop, meanwhile, consists of alternating layers of neutron- and gamma-absorbing materials to prevent the beam from crossing the sample.

The purpose of the collimator is to reduce beam divergence with a minimum induced background. This will be installed in the NB1 beam tube of the Moroccan Triga Mark II reactor (TMII). The beam tube comprises inner and outer segments which have internal radiuses of 15 cm and 20 cm, respectively. The collimator is generally constructed from two main materials for the absorption of neutron and gamma radiation, basically consisting of heavy metals [4]. For this purpose, two materials were evaluated, namely lead and carbon steel. Lead has been used to fabricate collimator components at several facilities, such as the University of Texas TRIGA reactor in Austin [5] and the 10 MW Budapest Research Reactor at Budapest KFKI [6]. Carbon steel, meanwhile, is used for the collimator installed at the Oak Ridge National Laboratory [7]. Lead, in its pure state, has the main disadvantage of softness. This is generally

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overcome by adding around 5% weight of antimony to enhance hardness. This improvement is important for making both the commissioning and decommissioning stages stack free [8]. However, it can also impact the performance of measurements. Carbon steel also has its own drawbacks, namely corrosion problems and/or chlorine hydrolase in humid areas. These phenomena are avoided through a nickel- or chromium-based coating process, which is an efficient way to prevent corrosion propagation inside the reactor vessel, particularly when the collimator is inserted inside the beam tube.

In order for this study to be useful for the Moroccan PGAA instrument under a wide range of conditions and circumstances, an additional investigation was pursued using lead and nickel-coated carbon steel. Two MCNP6.2 models were developed based upon two different configurations where a moderator and neutron absorber were a common part. This common part comprised borated polyethylene (HDPE+25%B_4C), which is the most widely used material for this purpose [9]. The first configuration comprised a heavy material block (for fast neutron reduction/absorption) followed by a moderator and thermal neutron and gamma absorber. The second one, meanwhile, involved a sandwich of materials comprising heavy material, moderator and thermal neutron gamma absorber. Our attempt to figure out the respective material and configuration for an adequate profile was based firstly on evaluating the neutron and gamma fluence rates at the exit of the collimator plug and outside the beam streamline background. Next, an assessment of gamma-induced activity versus irradiation time determination was considered, because this induced activity could make an additional contribution to gamma background at the measurement stage. Both assessments were performed for the two abovementioned configurations and for both raw and modified lead and carbon steel materials.

All calculations were performed using the MCNP6.2 Monte Carlo code [10] developed by the Los Alamos National Laboratory. The ENDF/VII.1 cross-section library was used to apply neutron-induced cross-sections and the MCPLIB84 library was used for photon transport. Calculating the activation within the neutron transport was run using the "ACT" card. The treatment used data from the DELAY_LIBRARY_V5.dat library and produced delayed neutrons for fission and activation where needed. Delayed gamma emission was calculated from ENDF/B-VII.1 data contained in CINDERGL.dat. Delayed betas, alphas, and positrons were sampled solely from the DELAY_LIBRARY_V5.dat data.

2. Methodology and components description

2.1. Neutron and gamma source

The input source was obtained using a TRIGA MARK II reactor MCNP6.2 model with fresh fuel [11]. Both the obtained neutron and gamma spectra were divided into seven angular bins. Each bin was 1° and discretized to 35 energy groups (Figs. 2 and 3). The decision was made to locate this source at the connection of the inner and outer segments of the beam tube, which was close to the extremity of the collimator located inside the outer segment (Fig. 1). The time-dependent source was used by SI and SP commends. The irradiation time was assumed to be 2.5 years with a 2 MW operating reactor power [12]. The delayed particles available in MCNP6.2— namely neutron, gamma, beta, alpha, and positron—were also taken into consideration.

2.2. Collimator

As mentioned previously, carbon steel and lead were used as candidate materials for collimator construction within this study.

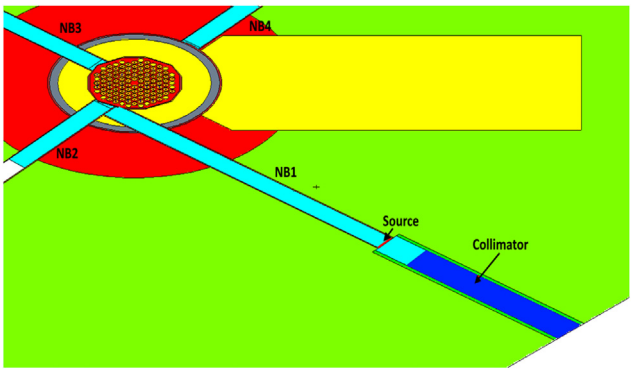

Fig. 1. Scheme presenting a horizontal cross-sectional view of the TMII reactor core and NB1 beam tube.

We studied two different configurations (labeled 1 and 2), each of which were used for the different materials. The indexes "a" and "b" correspond to the carbon steel and lead materials, respectively.

2.2.1. Configurations 1a and 1b

As shown in Fig. 4, configuration 1a consists of three main parts: The first part was the carbon steel rings with an aperture of 5 cm, length of 100 cm, and outer diameter of about 20 cm. The second part comprised 35 cm-long high-density polyethylene/boron carbide composite (HDPE+25% B_4C) rings with the same diameter and aperture used in the first part. Finally, for tightening purposes, an additional (third) part was added in the form of a 5 cm-thick carbon steel ring to tighten the whole collimator [11]. It is worth mentioning here that configuration 1b is largely identical to 1a, with the exception that it uses lead instead of carbon steel.

2.2.2. Configurations 2a and 2b

Configuration 2a comprises two kinds of 20 cm diameter rings with an aperture diameter of 5 cm. These rings are carbon steel and (HDPE+25%B_4C) composite, as depicted in Fig. 5 (2a). These are positioned alternately throughout the beam tube to get a total length of 140 cm. As in the previous case, configuration 2b deviates from 2a only by substituting lead for carbon steel.

3. Results and discussion

In this investigation, all calculation results were obtained assuming that the collimator aperture was an active section and that the collimator outer ring was a passive section, as demonstrated in Fig. 6.

In general, the hot spot of the beam should be as maximal as possible to achieve the best detection limits, although better beam quality can be achieved with the addition of sapphire and bismuth filters. On the one hand, the beam should not be projected directly onto the guide mirrors. For this reason, the aperture of the collimator is generally smaller than that of the guide. The aperture diameter is determined by taking into consideration the beam divergence angle to avoid high fluence neutron radiation at the guide mirrors. On the other hand, neutrons diverging from the beam, which are calculated at the passive section, will be projected laterally to the guide mirrors. This will particularly damage the first section of the guide mirrors, as well as lower the performance of other surrounding instruments. Thus, an estimation of the beam at the passive section is necessary to apply corrections at the level of the beam shutter.

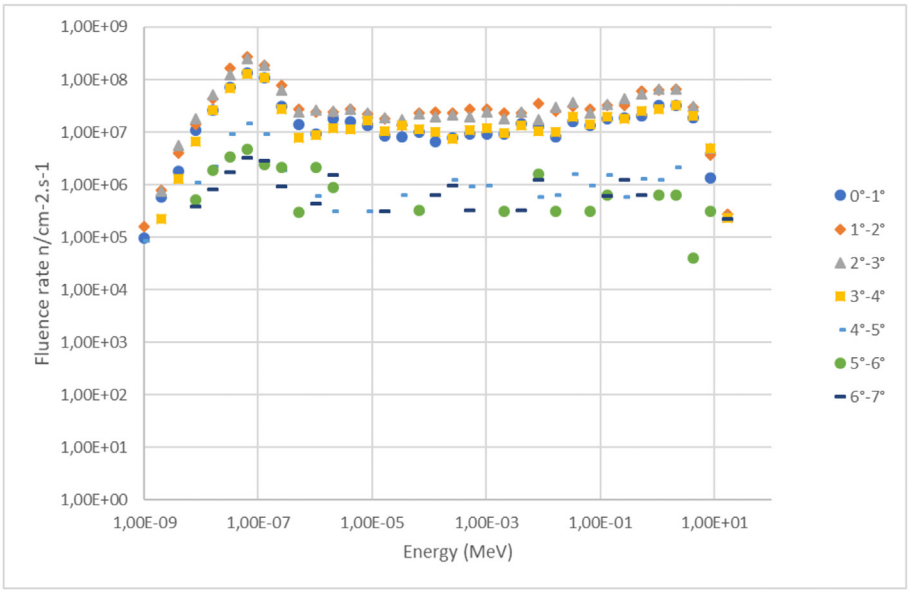

Fig. 2. Spectrum of neutron fluence rate per energy per angle of emission.

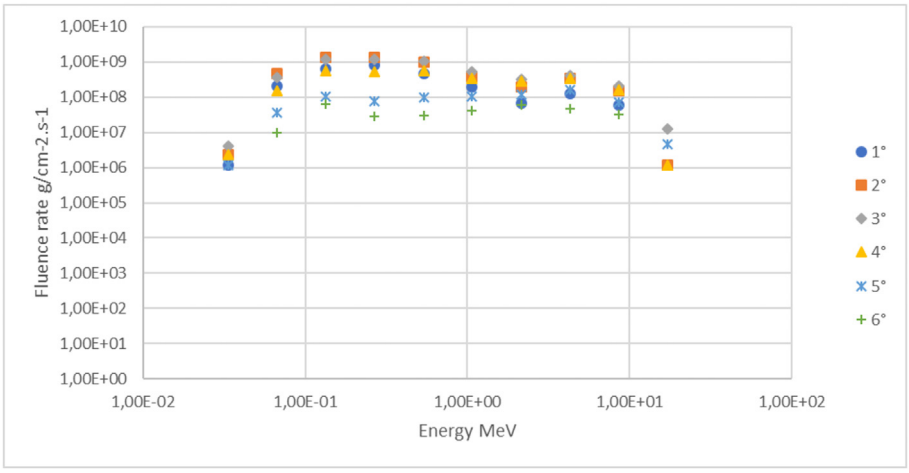

Fig. 3. Spectrum of gamma fluence rate per energy per angle of emission.

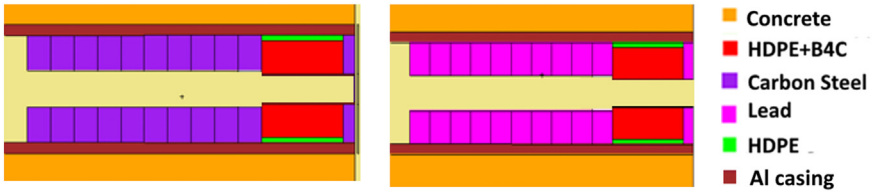

Fig. 4. MCNP models of the two variants of configuration 1 (left side—configuration 1a, right side—configuration 1b).

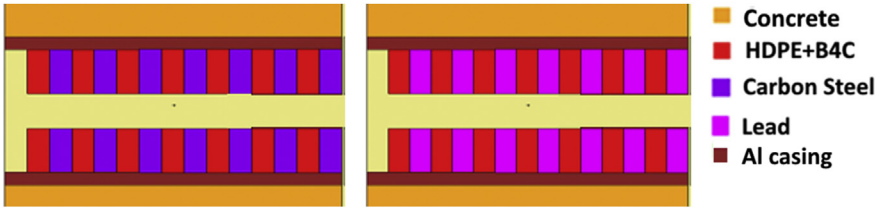

Fig. 5. MCNP models of the two variants of configuration 2 (left side—configuration 2a, right side—configuration 2b).

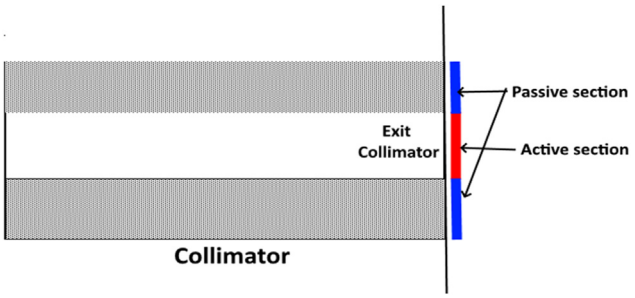

Fig. 6. Scheme presenting the active and passive section positions at the exit of the collimator.

3.1. Neutron fluence rate

Tables 1 and 2 show—for configurations 1b and 2b, respectively—the neutron fluence rate component calculation results at the exit of the collimator in the active and passive sections. The neutron energy bins generated ranged as follows: 0eV–0.5eV, 0.5eV-1keV, 1keV-1MeV, and 1 MeV–17.2 MeV. There was no significant impact on the neutron fluence rate components at the active region when swapping carbon steel with lead within the same configuration. This is primarily due to dominance of geometrical constraints. However, the slight differences that were observed were mainly due to varying scattering cross-sections between carbon steel and lead (Fig. 7). Furthermore, configuration 1 demonstrated a fluence rate intensity that was 24% higher than that of configuration 2. This is probably mainly due to configuration 2's use of neutron absorber along the collimator, whereas configuration 1 has neutron absorber material at the end of the collimator, where neutron scattering is low.

From the perspective of investigating the neutron background, it was noted that configuration 2 has a lower background in the passive section. The use of carbon steel in configuration 1 decreased the thermal and fast neutron backgrounds by more than 29% and 13%, respectively, when compared to using lead. Similarly, the use of carbon steel in configuration 2 decreased the thermal and fast neutron backgrounds, to 38.9% and 16.2%, respectively, when compared to using lead. This is most likely due to the absorption cross-section differences for the two studied materials (Fig. 8). It was also observed that the neutron background generated when using configuration 2 was on average 24% lower than that produced when using configuration 1.

3.2. Intrinsic gamma dose

In order to consider all gamma radiation in this evaluation, two calculations were performed for gamma radiation originating from the reactor core and that induced by neutron–matter interaction (neutron activation). The fluence-rate-to-dose function introduced by the International Commission on Radiological Protection (ICRP) was defined using the DE and DF cards in the MCNP code to calculate volume averaged doses and reaction rates. The calculated findings for the gamma equivalent dose rate (in Sv/h) at both the active and passive sections of the collimator are presented in Tables 3 and 4, respectively. It can be see how lead reduces gamma noise in the neutron beam by up to 4% for both configurations, while configuration 2 is better in terms of gamma noise contribution to the neutron beam. Meanwhile, the neutron activation gamma noise generated when using carbon steel is 10% higher than when using lead. This large difference can be explained through the configuration of the collimator and the differences between the absorber cross-sections when using lead and carbon steel.

On the other hand, based on the data in Table 4, configuration 1a has a 57%[3] lower total gamma background than configuration 2a, while for configuration 1b, it is 67%[4] lower than for configuration 2b. In addition, according to the total gamma cross section (Fig. 9), lead is more efficient at stopping high-energy gamma rays originating from the reactor core.

Table 1
Comparison of the components of neutron fluence rate ($cm^{-2}.s^{-1}$) in the active section at the exit of the collimator.

Energy	Config 1a	Config 1b	Diff %[1]	Config 2a	Config 2b	Diff %[2]
0eV-0.5eV	4.63E+8	4.71E+8	1.70	3.49E+8	3.52E+8	0.85
0.5eV-1keV	1.83E+8	1.82E+8	−0.55	1.37E+8	1.35E+8	−1.48
1keV-1MeV	2.08E+8	2.08E+8	0.00	1.57E+8	1.53E+8	−2.61
1 MeV-17.2 MeV	1.34E+8	1.37E+8	2.19	1.03E+8	1.03E+8	0.00
Total	9.87E+8	9.98E+8	1.10	7.45E+8	7.43E+8	−0.27

Table 2
Comparison of the components of neutron fluence rate ($cm^{-2}.s^{-1}$) in the passive section at the exit of the collimator.

Energy	Config 1a	Config 1b	Diff %	Config 2a	Config 2b	Diff %
0eV-0.5eV	4.28E+6	6.10E+6	29.8	3.17E+6	5.19E+6	38.9
0.5eV-1keV	3.72E+6	3.77E+6	1.3	2.98E+6	3.02E+6	1.3
1keV-1MeV	4.82E+6	5.09E+6	5.3	3.89E+6	4.11E+6	5.4
1 MeV - 17.2 MeV	1.26E+6	1.45E+6	13.1	9.80E+5	1.17E+6	16.2
Total	1.41E+7	1.64E+7	14.00	1.10E+7	1.35E+7	18.52

3.3. Gamma decay

Gamma decay emission induced from neutron activation radioisotopes (e.g., $Sb^{121}(n,G)Sb^{122}$, $Sb^{123}(n,G)Sb^{124}$, $Fe^{54}(n,G)Fe^{55}$ …) could have a relative impact on the quality of the analysis results. To quantify this impact, it was assumed that the collimator had been activated for 2.5 years. Many decay steps were considered—namely a week, a month, three months, six months, one year, and two years—to evaluate its evolution over time. As mentioned in the introduction, within the same investigation, the modified variants of the studied materials were also investigated in this section to cover all possible situations.

Firstly, ambient equivalent dose rates generated at the exit of the collimator were observed for configurations 1 and 2 using both uncoated carbon steel and nickel-coated carbon steel. The results are presented in Table 5. Nickel plating was used to protect the general mild steel from corrosion in high humidity conditions. The nickel coating was assumed to be 10 μm thick for these calculations. The results show that the induced gamma radiation almost stopped inside the collimator. It is clear that the nickel coating had no significant impact on the dose rate for configuration 1 using carbon steel. Conversely, for configuration 2, the calculated equivalent dose rate was 1.7 time > higher at measurement time when the carbon steel was nickel coated compared to uncoated carbon steel. No impact was observed in the collimator surroundings when the beam was closed.

Secondly, the results for the decayed gamma ambient equivalent dose rates at the exit of the collimator for the lead and lead plus 5% antimony variants are presented in Table 6. Antimony is usually added to lead to prevent it changing shape, which can cause many problems when manipulated. It can be seen in Table 6 that neither the configuration nor lead composition variations had a significant impact on the dose rates at the exit of the collimator, although the addition of antimony did increase the equivalent dose rate by about

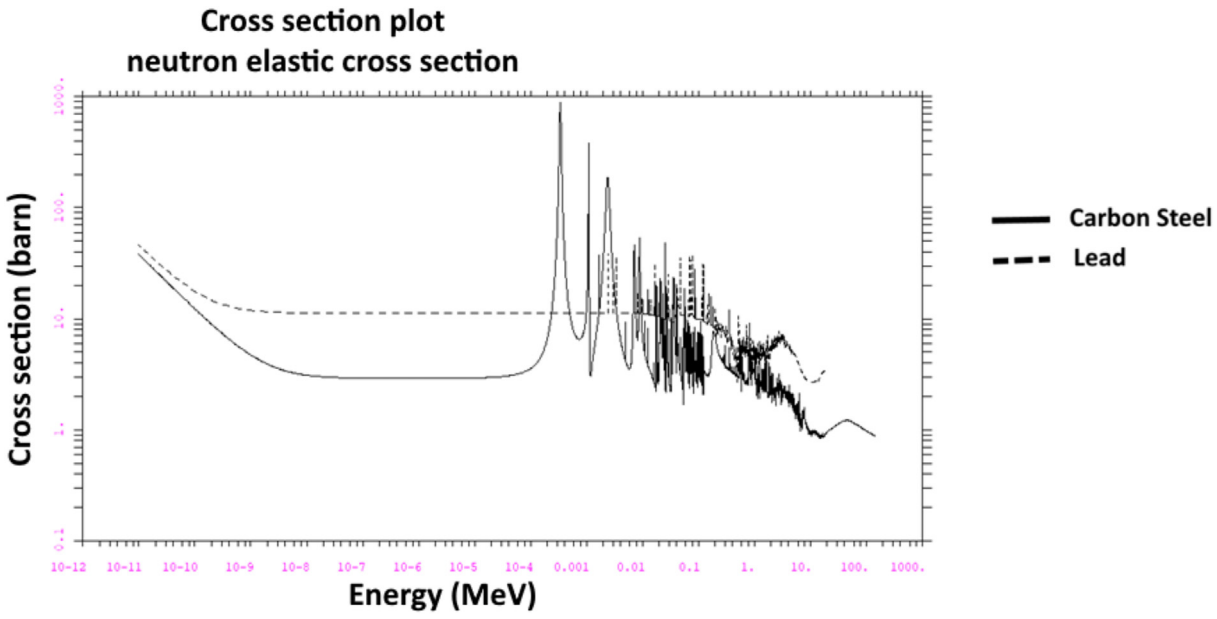

Fig. 7. Neutron elastic scattering cross-section for carbon steel and lead.

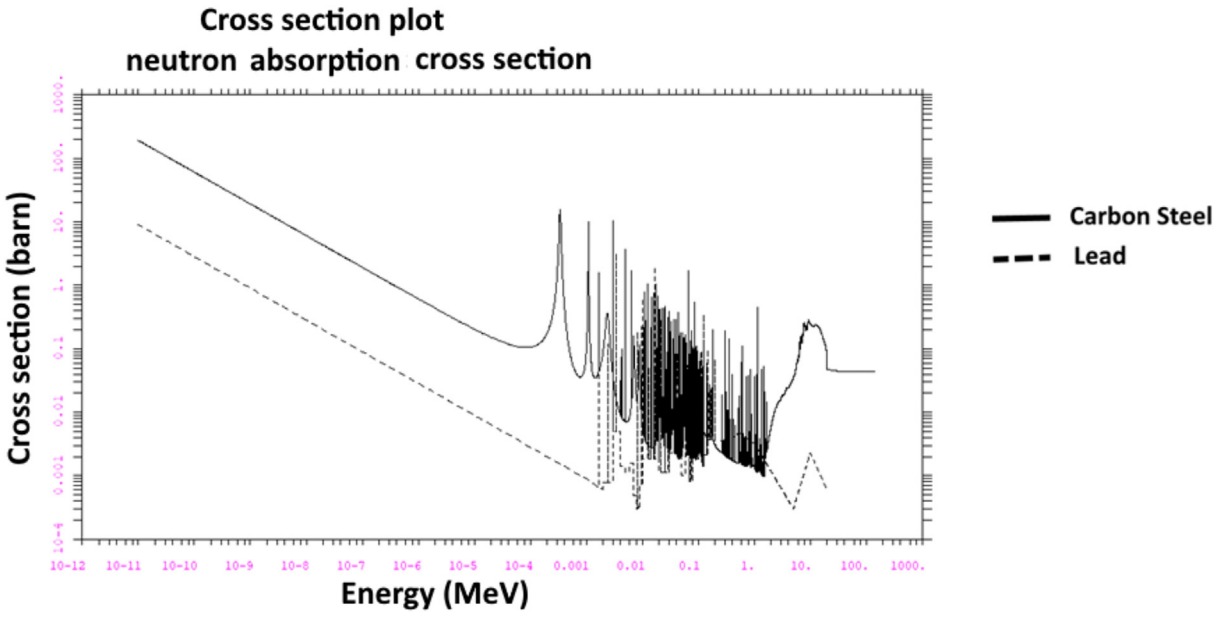

Fig. 8. Neutron absorption cross section for carbon steel and lead.

Table 3
Gamma ambient equivalent dose rate at the exit of the beam tube in the active section for the two gamma sources (Sv/h).

Type of gamma source (Sv/h)	Config 1a	Config 1b	Diff %	Config 2a	Config 2b	Diff %
Gamma dose from neutron activation	2.88	2.61	−10.34	2.19	1.94	−12.89
Gamma dose from reactor core	14.6	14.2	−2.82	11.6	11.3	−2.65
Total	17.48	16.81	−3.99	13.79	13.24	−4.15

7.60% and 17.14% for configurations 1 and 2, respectively.

4. Conclusions

To decide the most convenient material and collimator configuration for a PGAA system, two configurations were separately investigated using carbon steel and lead as heavy materials. A collimator configuration using carbon steel makes it possible to gain 24% in the intensity of the neutron fluence rate compared to using lead. This makes it a suitable choice for facilities requiring

Table 4
Gamma ambient equivalent dose rate at the outer section of the collimator in the passive section for the two gamma sources (Sv/h).

Type of gamma source (Sv/h)	Config 1a	Config 1b	Diff %	Config 2a	Config 2b	Diff %
Gamma dose from neutron activation	0.396	0.308	−28.57	1.28	1.1554	−10.78
Gamma dose from reactor core	0.177	0.0786	−125.19	0.0567	0.0341	−66.28
Total	0.573	0.3866	−48.22	1.3367	1.1895	−12.37

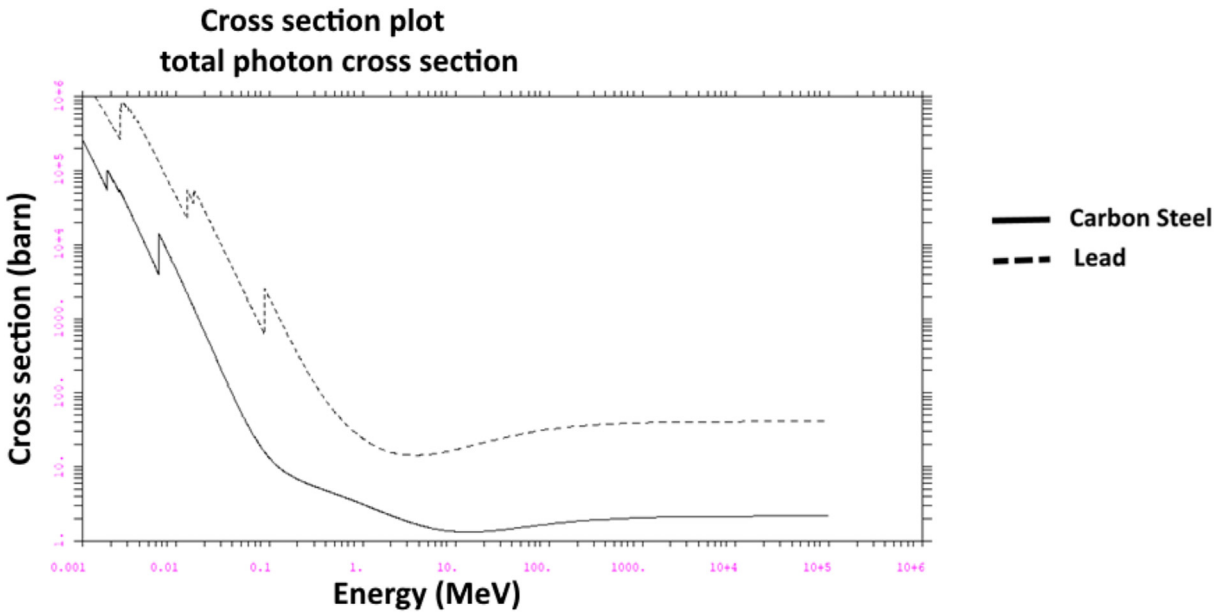

Fig. 9. Gamma total cross-section for carbon steel and lead.

Table 5
Ambient equivalent dose rate in (Sv/h) for delayed gamma after activation of collimator for 2.5 years for coated and uncoated carbon steel at the exit of the collimator.

	Configurations 1a and 2a							
		T = 0	1 Week day	1Month decay	3 Months day	6 Months day	1 Year day	2 Years day
Carbon steel without nickel coating	Configuration 1a	2.24	<1E-7(5)	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7
	Configuration 2a	1.66	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7
Carbon steel with nickel coating	Configuration 1a	2.26	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7
	Configuration 2a	2.84	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7

Table 6
Ambient equivalent dose rate in (Sv/h) for delayed gamma after activation of collimator for 2.5 years for lead and lead+5% antimony at the exit of the collimator.

		Configurations 1a and 1b						
		T = 0	1 Week day	1 Month decay	3 Months day	6 Months day	1 Year day	2 Years day
Lead	Configuration 1b	1.70	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7
	Configuration 2b	1.45	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7	<1E-7
Lead + 5% Antimony	Configuration 1b	1.84	3.11E-05	2.62E-05	2.24E-05	3.01E-06	9.34E-07	1.98E-07
	Configuration 2b	1.75	2.04E-05	1.69E-05	2.79E-06	2.08E-06	9.39E-07	1.65E-07

such a high fluence rate intensity. In addition, regardless of the collimator configuration, the use of carbon steel decreases the neutron background by more than 14% when compared to lead. Configuration 1, however, is unsuitable for measurement facilities installed close to the beam tube due to the higher neutron background when compared to configuration 2. Furthermore, the use nickel-coated or uncoated carbon steel has no significant effect on the level of gamma background around the beam tube.

On the other hand, configuration 1 can be regarded as more suitable than configuration 2 for facilities installed closer to the beam tube due to its lower gamma background, especially when using lead instead of carbon steel. For long-term irradiation, the use of lead with antimony generates an additional background that can influence facilities' surrounding measurements. Lead with antimony is therefore not suitable to be used around measurement instruments.

In conclusion, given that the PGAA facility requires a high fluence rate, combined with the fact that the measurement system is

far from the exit of the beam tube, configuration 1 with carbon steel rings is deemed to be more appropriate for the Moroccan PGAA collimator.

From another perspective, an investigation of the radioactive waste generated using these two materials at the decommissioning stage and their behavior over time under high neutron fluence rates will help give a general overview for making an adequate choice with respect to both facility requirements and neutron source characteristics.

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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