IMPROVING THE PERFORMANCE OF ^{241}Am-Be FOR PGNAA APPLICATIONS USING A PROPER SHIELDING FOR NEUTRON SOURCE AND THE NaI DETECTOR

by

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The gamma ray spectrum resolution from a ^{241}Am–Be source-based prompt gamma ray activation analysis set-up has been observed to increase in the energy region of interest with enclosing the NaI detector in a proper neutron and gamma ray shield. We have investigated the tact that the peak resolution of prompt gamma rays in the region of interest from the set-up depends on the source activity to the great extent, size and kind of the detector and the geometry of the detector shield. In order to see the role of a detector shield, five kinds of the detector shield were used and finally the proper kind was introduced. Since the detector shield has an important contribution in the reduction of the undesirable and high rate gamma rays coming to the gamma ray detector, a good design of a proper shield enables the elimination of the unwanted events, such as a pulse pile-up. By improving the shielding design, discrete and distinguishable photoelectric peaks in the energy region of interest have been observed in the spectrum of prompt gamma rays.

Key word: prompt gamma ray activation analysis, NaI(Tl), neutron source, gamma ray, neutron shield, boric acid

INTRODUCTION

The prompt gamma neutron activation analysis (PGNAA) technique has a wide range of applications, such as process control tasks in manufacturing industry, contraband detection and well logging in oil exploration [1-3]. Detecting concealed threats in hidden compartments of maritime vessels, and detecting buried land mine and explosive materials are only some of the examples of PGNAA method applications as a non-intrusive tool [4, 5].

Another application of PGNAA is in diagnostic medicine. The human body, as well as the majority of explosives, contains a significant amount of nitrogen. After the capture of a thermal neutron, the nitrogen nucleus emits a characteristic 10.8 MeV gamma ray [6]. The energy of this gamma ray is well above the characteristic energy of gamma rays from neutron capture on other elements that may usually exist in the sample.

This gamma ray signature appears in the 9-11.5 MeV energy region of the detected spectrum. The region of interest (ROI), or nitrogen energy region, is critical in neutron activation analysis applications for detecting nitrogen.

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NaI detectors are still used frequently in industrial PGNAA applications such as "total body nitrogen estimations" [6-8]. They have the advantages of being efficient for high-energy gamma rays, rugged, and they can be used without cooling.

Inadequate shielding of the gamma ray detector against unwanted radiation can cause detrimental effects on the detector responses and acquired spectra.

In the present paper the proper shield for a 3 3 NaI scintillation detector was investigated by comparison of a certain number of spectra acquired from different shield designs.

The primary purpose of this paper is to achieve an easy design in which the three well-known peaks of nitrogen, 10.8 MeV (photopick), single escape (SE) and double escape (DE), as a track of the existence of nitrogen in the sample, have the highest resolution.

METHODS AND PROCEDURES

The geometry of the ^{241}Am-Be source-based PGNAA set-up employed in the present study is shown in fig. 1. In order to get relatively maximum yield in the acquired prompt gamma rays spectra, the optimum moderator length and the optimum position

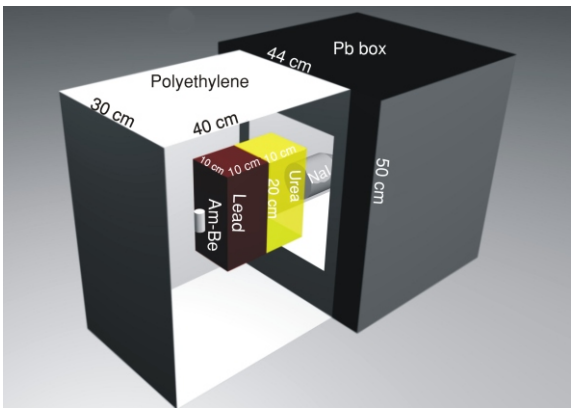

Figure 1. Schematic set-up of the ^{241}Am-Be source based PGNAA set-up with the source-inside-moderator geometry

of the ^{241}Am-Be neutron source inside the moderator were implemented. The optimum position is achieved when the maximum number of thermal neutrons collides with the sample. The geometry consists of a cubical sample (10 cm 10 cm 20 cm) and a cubical Pb shield (10 cm 10 cm 20 cm) enclosed in a cubical high-density polyethylene assay (97%, 30 cm 40 cm 50 cm), in a way that the sample is positioned between the cubical Pb shield and a gamma ray detector. A Pb room as a low background space enclosing detector is also considered to omit the background gamma rays.

The cubical Pb shield 10 cm thick placed between the ^{241}Am-Be source and the sample is there in order to obtain a satisfying gamma spectrum compared to samples' library spectrum, without interfering with the ^{241}Am-Be gamma ray component (4.438 MeV) and its subsequent Compton-scattered gamma rays from the source capsule. This shield attenuates the 4.438 MeV gamma rays resulting from the first excited 2+ state in ^{12}C in the ^{241}Am-Be neutron source [9]. In this way this Pb shield not only filters the ^{241}Am-Be gamma ray component but also moderates neutrons and slightly increases the neutron flux via (n, Xn) interaction. Depending on the neutron energy, X can be 2 or 3 [10]. Also, due to the fact that natural Pb has a negligible (n, γ) interaction cross-section compared to the sample neutron capture cross-section [10], the selection of Pb as a gamma shield for the neutron source in this PGNAA set-up is the best choice.

The prompt gamma ray spectrum measurements were performed with a 3 3 NaI detector and a ^{241}Am-Be source with 5 Ci (1 Ci = 3.7 10^{10} Bq) of activity contained in a standard Amersham X.14 capsules format (code AMN24) that was placed 25 cm from the detector surface. To reduce a significant background response in the NaI detector due to the prompt and radioisotope gamma rays produced by neutron activation in the NaI crystal, a neutron shield must be employed. The radioactive capture cross-section (n, γ) for thermal neutrons by ^{127}I is about 10^{-27} m^2. Figures 2 (a) and 2 (b) show neutron

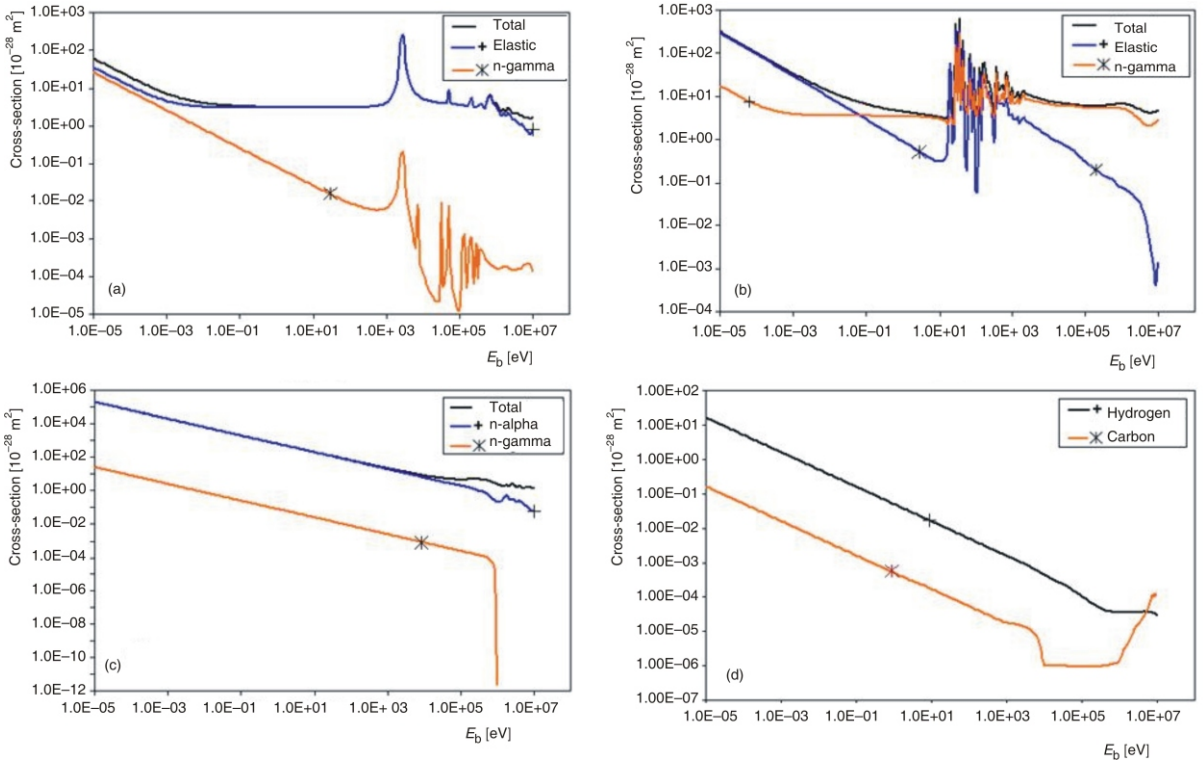

Figure 2. Neutron cross-section: (a) ^{23}Na, (b) ^{127}I, (c) ^{10}B, and (d) ^{1}H and ^{12}C

cross-sections for ^{23}Na and ^{127}I, respectively [10]. It can be seen that the role of iodine in absorbing low-energy neutrons is greater than that of sodium. Also the half-life of the radioactive product ^{127}I is much shorter (24.99 minutes) than that of ^{24}Na (14.98 hours).When thermal neutrons are captured, in addition to the prompt gamma rays, the residual elements, ^{24}Na and ^{128}I, still emit some beta and gamma radiation. Tables 1 and 2 list the radiation resulting from the decay of ^{24}Na and ^{128}I [10]. In assessing the extent of the activation of a NaI(Tl) detector, we should take into account all the possible neutron reactions with the components of the scintillator crystal. Thermal neutrons usually lead to (n, γ) reactions, while fast neutrons are generally associated with scattering reactions, and if they occur within the NaI crystal, they will result in a crystal defect. The contribution of the neutron capture in any neutron shielding to the final gamma rays spectrum must also be considered. The types of reaction in these shields depend on the neutron energy. For absorbing thermal neutrons, boron compounds are highly effective [11].

Table 1. Gamma and beta rays from ^{24}Ne

Radiation	Energy [MeV]	Intensity [%]
γ	1.368	99.992
γ	2.754	99.944
β	0.277	0.053
β	1.390	99.944

Table 2. Gamma and beta rays from ^{128}I

Radiation	Energy [MeV]	Intensity [%]
γ	0.443	0.16
γ	0.527	0.0154
β^-	1.682	15
β^-	2.125	77
β^+	0.511	0.14
β^+	1.254	6

In this work boric acid (H_3BO_3 fine powder, density of 0.89 g/cm^3 was used as a neutron shield material for the NaI(Tl) detector. Neutrons in the vicinity of the detector may be absorbed by ^{10}B present in the boric acid. The thermal neutron cross-section of ^{10}B is 3840 10^{-28} m^2. Figure 2(c) shows the variation of the ^{10}B cross-section as a function of the neutron energy [10].

In the experiment with the configuration shown in fig. 1, by using the proper detector shield and a thick layer of Pb in front of the neutron source, the concentration of the "spiked" element was such that prompt gamma ray photopeaks of interest could be clearly observed, as well as the peaks' centroids determined without interference from other photopeaks or continuum. The other gamma rays which were generated by the sample with the lower energy may have vanished in the acquired spectrum by those high rate prompt gamma rays produced due to the existence of carbon and hydrogen in the moderator. Table 3 shows the energies of prompt gamma rays with respect to ^{1}H and ^{12}C targets that can be observed in the experimental spectra [12]. Figure 2(d) shows the neutron cross-sections of the (n, γ) interaction by ^{1}H and ^{12}C targets as a function of neutron energy [10].

Table 3. Thermal neutron capture gamma rays

Target	γ [MeV]	Intensity [%]
^{1}H	2.223	100.00
^{12}C	0.595	0.36
	1.262	47.96
	1.857	0.24
	3.089	0.64
	3.684	47.64
	4.945	100.00

The accumulation of these high-rate and undesirable gamma-rays especially (2.22 MeV by hydrogen) in the detector volume and consequently simultaneous pulses that can be piled up highly distort the spectra in the energy ROI.

Pile-up usually has two disturbing effects on the spectrum. The first is increasing the continuous background in the ROI [13] due to the pile-up of low-energy pulses, and the second is the escape of the pulses from ROI to the higher energy region when the energy of pile-up pulse is more than the upper level of the ROI.

In order to solve this problem, the NaI detector itself must be shielded by Pb in a way which filters undesirable gamma rays while "welcomes" the prompt gamma rays originating from the sample.

The first thought is to use an annular Pb shield for the NaI detector. In this way just the front face of the detector is exposed to radiation.

Some types of shields for the 3 3 NaI(Tl) detector were tested but for comparison and for drawing conclusions easily just five of them which have significant effects on the acquired spectrum are shown in fig. 3.

Figure 3 shows five types of detector shields made of boric acid (thermal neutron shield) with different thicknesses of the cylindrical layer ranging from 1 to 3 cm.

Each layer of boric acid shield composed of boric acid fine powder housed in a thin-walled aluminum container in which can surround the NaI detector.

The other types of shield are not only consisting of boric acid but also consist of annular Pb shield covering the NaI detector side.

Spectra of prompt gamma rays with these kinds of shield in an one hour were measured by position-

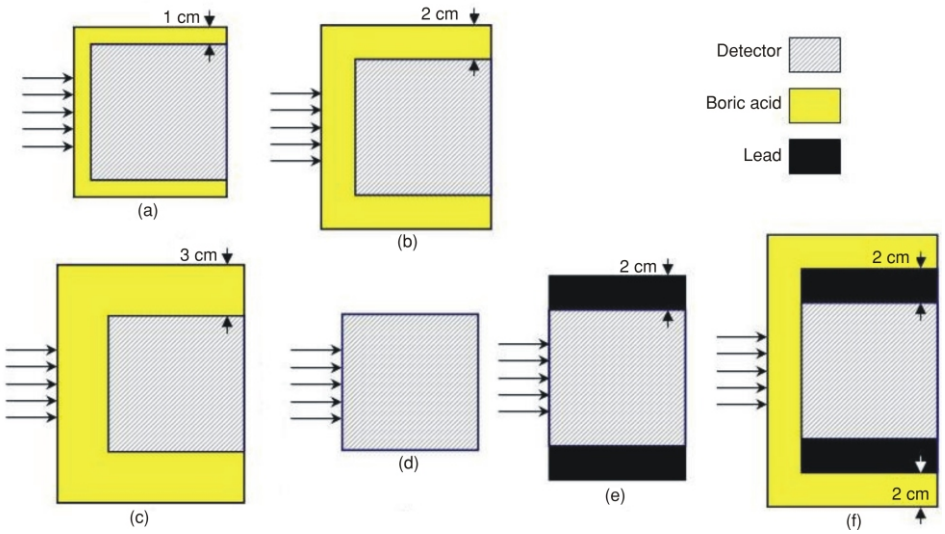

Figure 3. Detector shield configurations made of a boric acid cylindrical layer and an annular Pb shield

ing blocks of urea fine powder (CH_4N_2O, assay 99%, 10 cm 10 cm 20 cm) as a sample on the detector side, between the cubical Pb shield and NaI(Tl) detector.

The pulse height spectra were acquired with an Aptec model 5004 multi channel analyzer (MCA) set, so that the first 1500 channels corresponded to an energy range of about 0-12 MeV. The total time for each measurement was only one hour and the MCA was operated in the stabilized data acquisition mode.

RESULTS AND DISCUSSION

In the PGNAA experiments five types of detector shield, shown in fig. 3, were positioned around the detector one by one.

Each of the following figures is based on the one type of the detector shield. Figure 4 is the spectrum of urea when the configuration of the detector shield is the (a) structure shown in the fig. 3. Figures 5 and 6 are corresponding to the (b) and (c) structures, respectively. These show that by increasing the thickness of the cylindrical layer of the boric acid shield the resolution of the spectrum in the ROI will be higher and the peaks will be more distinguishable.

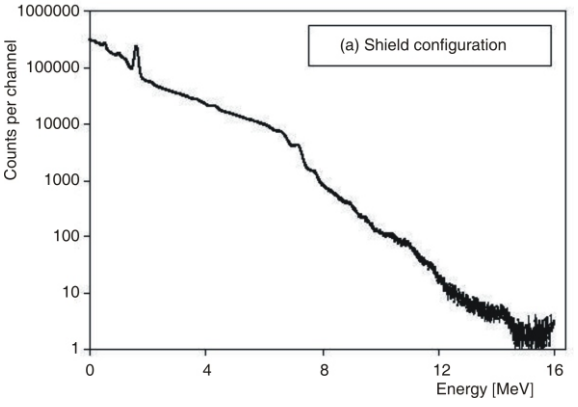

Figure 4. Prompt gamma ray spectrum of urea with (a) structure

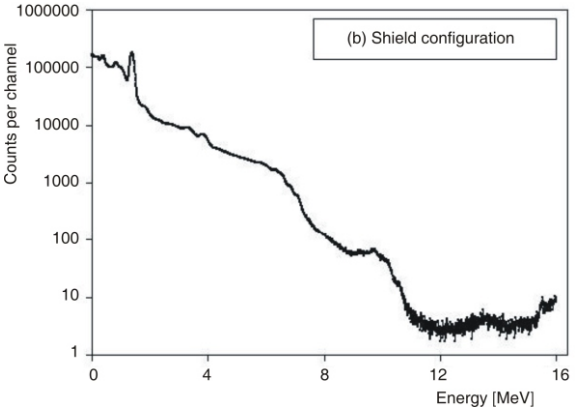

Figure 5. Prompt gamma ray spectrum of urea with (b) structure

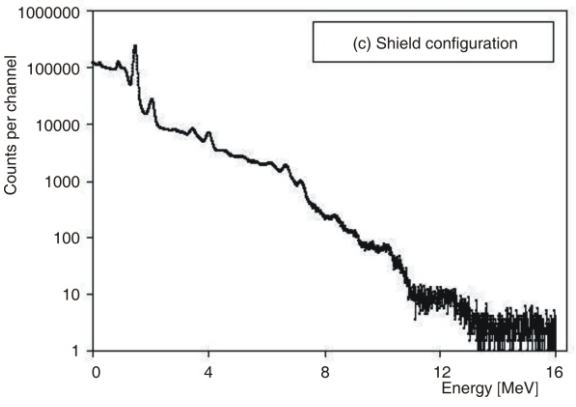

Figure 6. Prompt gamma ray spectrum of urea with (c) structure

Note that by increasing the thickness of the boric acid shield we cause the reduction of the total gamma ray flux coming to the detector to a certain extent. As we can see, having a good resolution in the acquired spectrum by a thicker layer of the boric acid shield is not an uncommon reason for a little pile up. Figure 7 is the spectrum of urea using a bare NaI detector. Figures 8 and 9 are relative to the (e) and (f) structures, respectively, shown in fig. 3.

According to fig. 9 and a comparison between the other spectra, we can be sure that the best kind of shield between the used structures is the (f) kind.

A 2 cm thick layer of boric acid in the (f) configuration is the optimum for one hour acquisition time, while in comparison using a 3 cm thick layer of boric acid to get the similar spectrum and resolution in the ROI, more than one hour is needed.

To confirm the suitability of using a thick layer of Pb (10 cm) in front of the neutron source (behind the sample) and to see its consequent effects on the acquired spectra, another spectrum is also recorded when the cubical Pb shield is replaced with a cubical polyethylene (the moderator material).

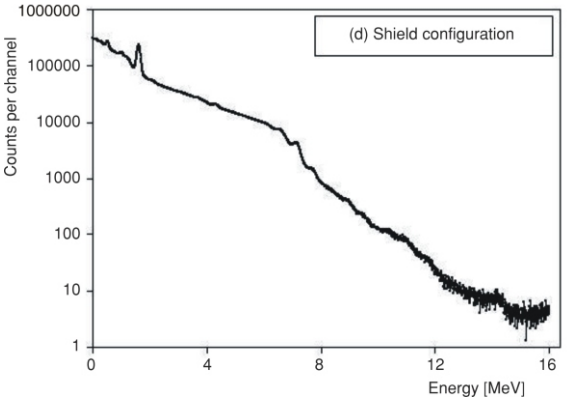

Figure 7. Prompt gamma ray spectrum of urea with (d) structure

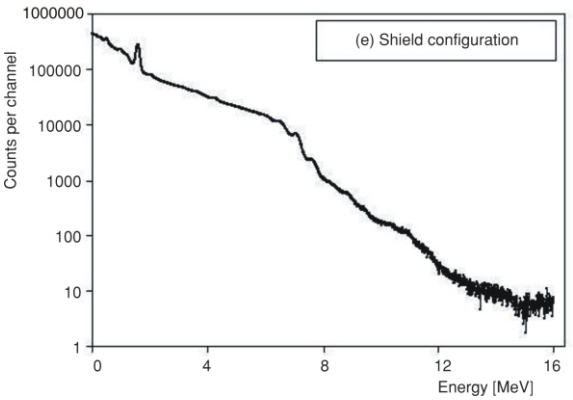

Figure 8. Prompt gamma ray spectrum of urea with (e) structure

Figure 10 shows the acquired spectrum when the cubical Pb shield in front of the neutron source was replaced by a moderator material. In this spectrum a high rate neutron source gamma ray component (4.438 MeV) completely distorts the whole spectrum, especially in the ROI. This shows that the gamma activity of the neutron source is sufficiently intense to lead pulse pile up and significant background in the ROI.

Also in the high energy region above the 12 MeV a "footprint" of a high energy gamma ray can be observed. It is not a real gamma ray emitted from the sample or source. As was stated previously, at high rates, pulse pile up can result in apparent peak amplitudes for gamma rays which are considerably larger than any individual pulse [13].

Comparing figs. 9 and 10, the idea to use gamma shield in front of the neutron source behind the sample position to reduce the portion of the source gamma ray component in the ROI is highly strengthened. If we use a neutron source that emits only neutrons this concern would vanish.

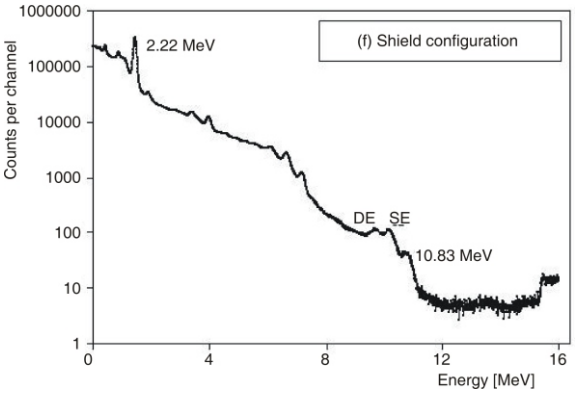

Figure 9. Prompt gamma ray spectrum of urea with (f) structure

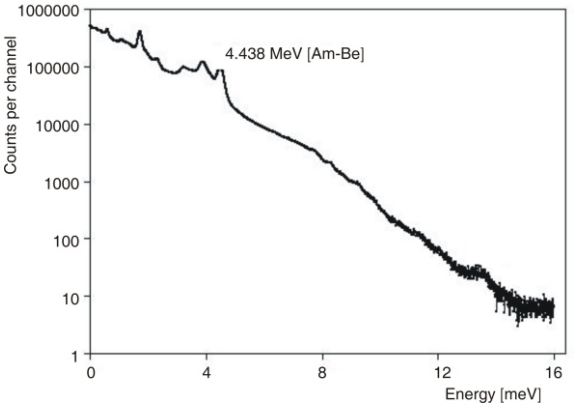

Figure 10. Prompt gamma ray spectrum of urea with (f) structure and the cubical Pb shield in front of the neutron source replaced with cubical polyethylene

CONCLUSIONS

This work gives an improved design of a detector shield based PGNAA set-up and suggests the proper and optimum size of the detector shield. The optimum size of the moderator and the distance between the source and the detector lead to reduce the acquisition time to get a relatively "good" spectrum for a special sample. By using a proper shield, the acquisition time and dead time fall down significantly and a distinguishable spectrum (without the disturbing effects of pile up) in the ROI can be achieved in a shorter time (*e*. *g*. one hour). On the whole, a large portion of undesirable gamma rays are subtracted from the total amount of incidence of radiation to the detector. Thus we can achieve the better resolution in the recorded peaks, which is important in the elemental analysis.

Comparing all of the detected spectra, it is well understood that the (f) structure can delete the effects of the interfering gamma rays in the ROI not completely but sufficiently to measure precisely the relative percentage of the existence of an special element in a composite material.

Note that providing appropriate shielding for the detector will thus not only improve the observed spectra but also increase the life of the detector.

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УНАПРЕЂИВАЊЕ РАДНИХ ОСОБИНА ^{241}Am-Be ИЗВОРА У АКТИВАЦИОНОЈ АНАЛИЗИ КОРИШЋЕЊЕМ ПОГОДНОГ ШТИТА

При активационој анализи промптним гама зрачењем уочено је да се резолуција спектра гама зрачења из ^{241}Am-Be извора увећава у енергетској области од интереса уколико се NaI детектор окружи погодном заштитом од неутрона и гама зрачења. Испитано је у којој мери резолуција пика спектра гама зрачења у области од интереса зависи од активности извора, величине и врсте детектора и геометрије детекторске заштите. У циљу да се одреди утицај штита детектора, употребљено је пет врсти штитова и, на крају, изабран прави, Како штит значајно доприноси умањењу нежељеног гама зрачења које стиже у детектор, ваљан пројекат погодног штита спречава нежељене догађаје, на пример, нагомилавање импулса. Унапређењем пројекта заштите у спектру промптног гама зрачења примећени су дискретни, раздвојени фотоелектрични пикови у испитиваној енергетској области.

Кључне речи: активациона анализа промптним гама зрачењем, NaI(Tl), неутронски извор, гама зрачење, неутронски штит, борна киселина