Measurements of the Energy Spectra of Photoneutron Reaction Ga (γ, n) in the Giant Dipole Resonance

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The development of intensive neutron source based on an electron accelerator without fissile materials is promising approach for the production of radioactive isotopes for medical purposes. The basis of the technical approach is to combine the technology of intense beams of high energy electrons and liquid gallium as a target, whose activation by generated bremsstrahlung gamma-rays leads to the formation of short-lived radioactive nuclei. As a result, gallium is environmentally safe working body target with indefinite useful, and technology for intensive neutron radiation based on gallium target does not require processing and storage of radioactive waste.

The purpose of this work is to investigate neutronic properties of gallium target irradiated by pulsed source of electrons with an energy of 35 MeV (IREN Facility, JINR, Dubna). A series of experiments were made to measure such neutronic characteristics of gallium target, as neutron yields, energy spectra of photoneutrons and the time dependence of gamma rays intensity of activated gallium target).

Experimental set-up

Measurements of yields and energy distributions of neutrons generated by 35 MeV electrons in gallium target were made with the help of methods based on the activation threshold detectors and ^{3}He - neutron spectrometer. The IREN Facility (JINR, Dubna) was used as an electron accelerator. IREN Facility operates in a pulsed mode with a pulse repetition rate of 25 Hz. The energy of electrons incident on the gallium target was 33.58 MeV. The pulse width was 0.1 ms, the current value in the electron pulse was 1 A. The neutron flux monitor was installed in the experimental hall to control the stability of the electron beam.

The gallium target was designed in the form of stainless steel cylinder filled by high purity gallium. The target diameter is 60 mm, height – 150 mm, weight of gallium – 2220.5 g, empty target weight – 371.5 g. The gallium target temperature during irradiation session was monitored by three thermocouples located at the top and the bottom end, and in centre of target back surface. At 50 mm from the entrance window of the target a technological hole of 12 mm diameter was made with the purpose to place there the activation detectors and gallium samples. The gamma ray activities data obtained with the help of activation detectors and Ga samples was used to determine the neutron energy spectrum and neutron flux density inside the gallium target. Arrangement of the gallium samples and activation monitors inside target is seen in Figure 1, which shows a section of a gallium target at a distance of 50 mm from the input window of the electron beam.

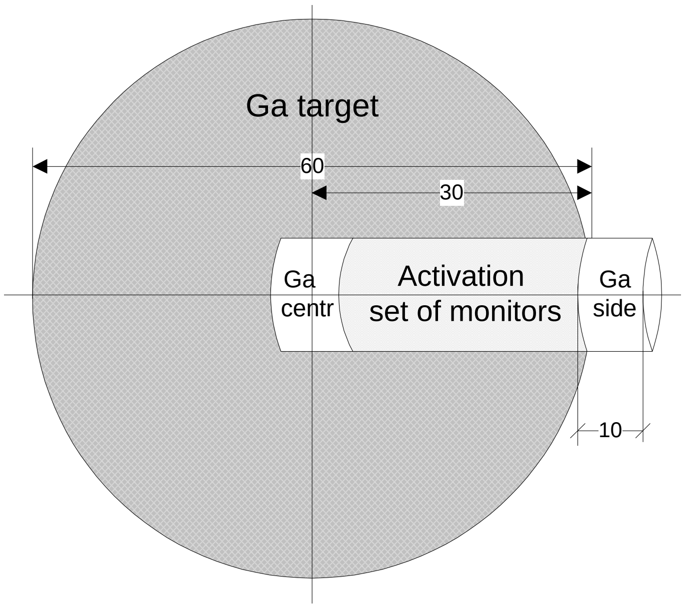

Figure 1. Arrangement of monitors and the Ga samples in gallium target.

A photoneutron energy spectrum from outside of the gallium target was measured with the help of ^{3}He-spectrometer. The experimental set-up for measurement of the energy spectra of neutrons emitted during the interaction of the electron beam with the gallium target is presented in Figure 2 (Ga - gallium target; Pb - 15 cm of lead shielding; PA - preamplifier; Canberra - signal processing system; National Instruments - data acquisition system based on National Instruments modules). ^{3}He-spectrometer was installed at a distance of 10 m from the gallium target. In front of the entrance window of the spectrometer a lead filter was set. The thickness of lead filter was 15 cm. The filter designed to reduce the intensity of bremsstrahlung gamma rays produced by deceleration of electrons in the Ga target.

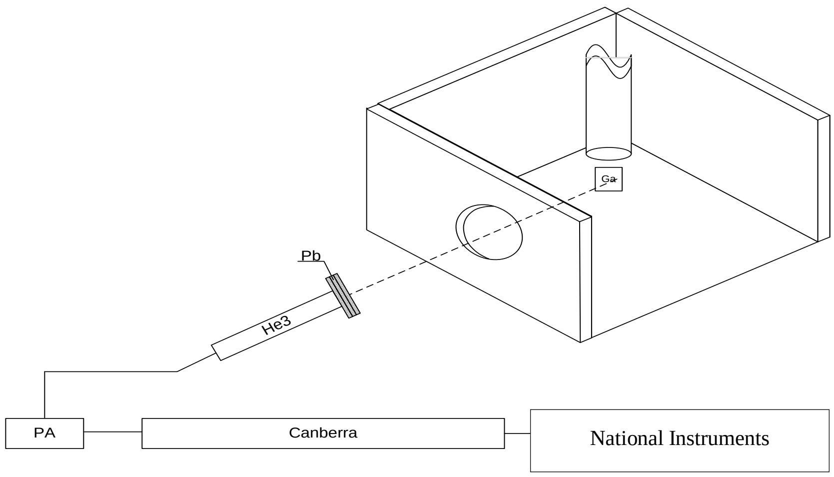

Figure 2. Experimental setup based on ^{3}He-spectrometer.

Measurements and data processing in the ^{3}He-spectrometer based experiment

^{3}He-spectrometer was operated at a high level intensity background. As a result, the primary pulse spectrum of spectrometer includes a large number of piled-up pulses of gamma rays and high-energy neutrons just after electron pulse. Therefore, it was decided to record a time depended sequence of the waveform of the primary pulses in a cumulative binary file with a purpose of making pile-up corrections during processing of the accumulated data. The primary pulse spectrum, the pile-up pulse spectrum and pulse spectrum corrected for pile-up effect are presented in Figure 3.

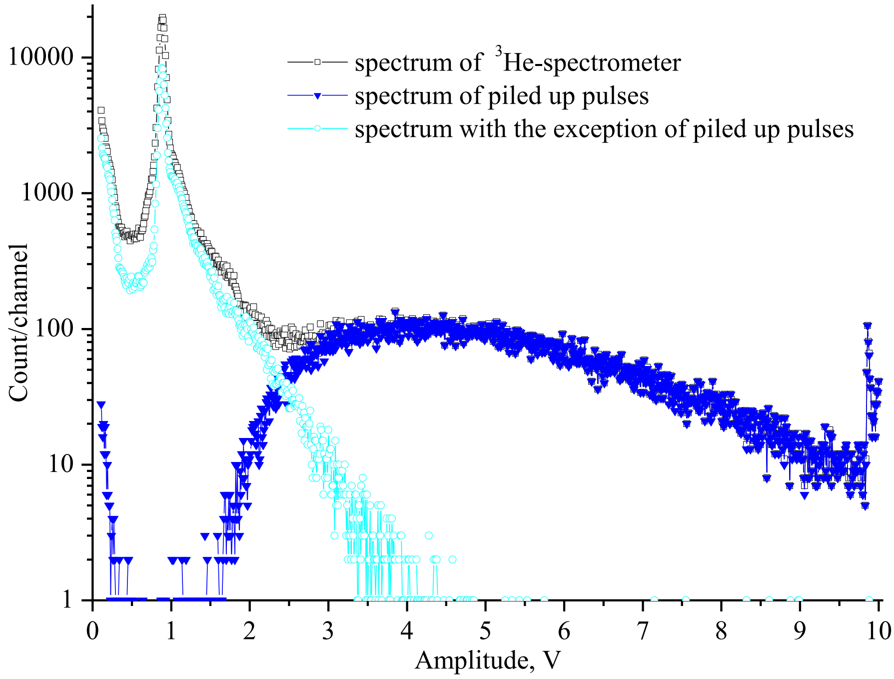

Figure 3. Instrumental neutron spectra obtained by processing the waveforms recorded during a session of gallium target irradiation by electrons with energies of 35 MeV.

After introducing the pile-up rejection the correction was made for distortion of the baseline of the ^{3}He-spectrometer which was induced by high count rates. Pole-zero compensation corrections was made in frame of specially developed mathematical procedure. Then the corrections for the ^{3}He recoil nuclei, wall effect, efficiency as well as for the attenuation of neutron flux in the lead slab.

Measurement and data processing in activation analysis experiment

The activation method for measuring neutron spectra uses the link between induced activity of detectors and neutrons flux density. The reactions are divided into two groups: reactions, which are most sensitive to low energy neutrons - (n, γ), (n, f) and reactions that are significant at neutron energies above a certain value, called the threshold energy of the reaction (threshold reaction). The main thresholds reactions are the neutron capture reactions with the emission of charged particles - (n, p), (n, α), an inelastic neutron scattering (n, n'), the neutron capture reactions with the emission of two neutrons (n, 2n) and the fission reaction (n,f).

In the present work the energy spectrum of neutrons generated into a gallium target by electrons with energies of 35 MeV in the range of 0.0001 eV to 16 MeV were measured by the activation method on the basis of reaction rate of following nuclear reactions: ^{56}Fe(n,p)^{56}Mn, ^{58}Ni(n,p)^{58}Co, ^{63}Cu(n,γ) ^{64}Cu, ^{113}In(n,γ)^{114m} In, ^{115}In(n,n')^{115m}In and ^{27}Al(n,α)^{24}Na. Activation detectors (monitors) were made in the form of disks with a diameter of 10 to 12.2 mm. The thickness of the monitors is in the range of 0.6 to 3.5 mm. All monitors were made of chemically pure metal.

For irradiation in neutron flux generated inside gallium target, monitors were placed in the channel of the target located at a distance of 50 mm from the entrance window. After irradiation the activation detectors were placed on the HPGe detector [1] for gamma ray measurements. An example of gamma ray pulse-height spectrum measured from Al detector is presented in Figure 4.

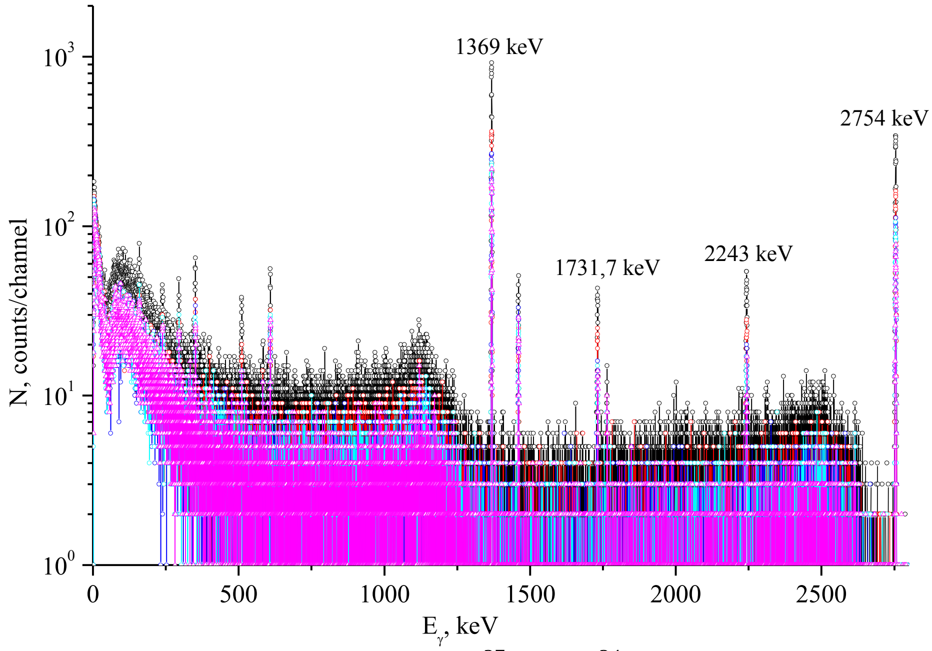

Figure 4. Pulse-height gamma ray spectra of the $^{27}Al(n,\alpha)^{24}Na$ reaction products in the sample of aluminum irradiated by the neutron flux generated in the Ga target.

All measured gamma ray pulse-height spectra were processed to obtain the photo-peak area of full energy deposition of gamma rays of a certain energy. The obtained values were used to calculate the reaction rate of the corresponding nuclear reactions. The measured values of nuclear reaction rates have been adjusted for the new recommended decay data [2, 3]. Adjusted reaction rate values of nuclear reactions R1 and R2 for two irradiation sessions with the different space arrangement of the monitors are given in Table 1. Table 1 lists the statistical error in the measured values of the reaction rates. In the measurements of the reaction rate of $^{63}Cu(n,\gamma)^{64}Cu$ reaction an additional source of error was the gamma ray background, which was attended by peaks with energy of 511 keV. The contribution of the background component into total absorption peaks of these gamma rays is about 30% of the total number of pulses in the peak. The background level was constant throughout all measurements.

The STAYSLF program was used to unfold the photoneutron spectrum from the reaction rates data presented in the Table 1. This program is based on the method developed in the code STAYSL [4] in which a Bayesian approach is used. In this study a priori information was taken from the experimental data obtained in the measurements of the neutron energy distributions of the photoneutron into energy range from 0.01 to 0.7 MeV using ^{3}He-spectrometer. An a priori data in the range below 0.01 MeV were obtained on the basis of calculations performed within the evaporation model.

Table 1. Measured values of nuclear reaction rates.

Reaction	Eγ, keV	R1, react./s·nuclei	σ1, %	R2, react/s·nuclei	σ2, %
$^{56}Fe(n,p)^{56}Mn$	847	$8.027\cdot10^{-19}$	16.9	$4.284\cdot10^{-19}$	8.3
$^{58}Ni(n,p)^{58}Co$	811	$1.381\cdot10^{-17}$	5.7	$1.362\cdot10^{-17}$	4.1
$^{63}Cu(n,\gamma)\ ^{64}Cu$	511.16	$3.27\cdot10^{-17}$	1	$2.477\cdot10^{-16}$	1
	1345.93	$4.426\cdot10^{-17}$	10	$3.13\cdot10^{-16}$	8.3
$^{113}In(n,\gamma)^{114m}In$	190.64	$2.95\cdot10^{-15}$	3.1	$3.805\cdot10^{-16}$	2.5
	558.56	$3.333\cdot10^{-15}$	2.9	$4.165\cdot10^{-16}$	7.4
	725.28	$3.277\cdot10^{-15}$	3.2	$4.192\cdot10^{-16}$	8.1
$^{115}In(n,n')^{115m}In$	336.53	$1.042\cdot10^{-16}$	2.2	$2.827\cdot10^{-17}$	4
$^{27}Al(n,\alpha)^{24}Na$	1369	$5.745\cdot10^{-19}$	4.4	$3.084\cdot10^{-19}$	12.6
	2754	$5.791\cdot10^{-19}$	6	$2.943\cdot10^{-19}$	17.9

Comparison of the energy dependence of the flux density of photoneutrons obtained on the basis of ^{3}He-spectrometer data and activation detectors data is shown in Figure 5.

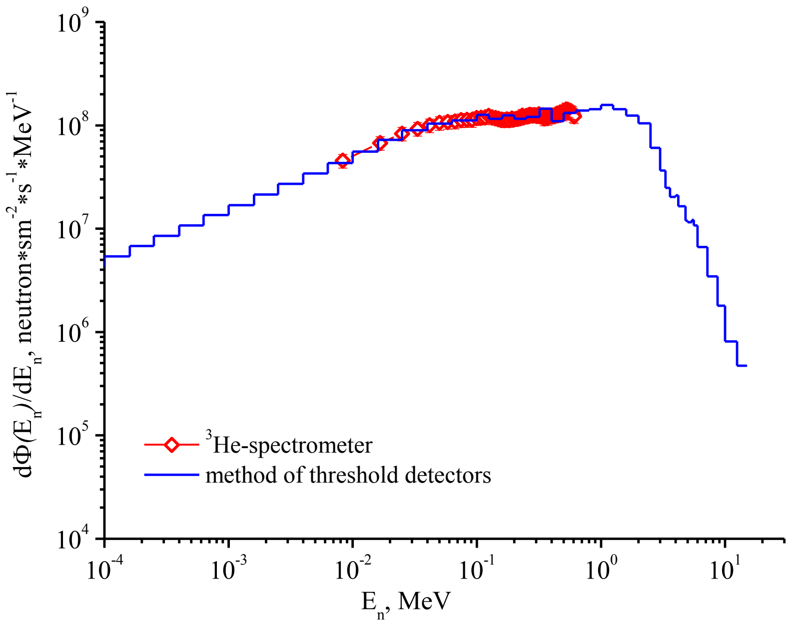

Figure 5. Comparison of the energy dependence of the flux density measured by the activation method and ^{3}He-spectrometer.

The average energy of the photoneutron spectrum presented in the Fig.5 is 2.17 MeV. The flux density obtained on the basis of these data is $1.7\cdot10^{9}$ neutron·cm^{-2}·s^{-1} for the average electron current of 2.5 mA. It should be noted that the energy range of photoneutron spectrum in the present study significantly expanded towards of soft neutrons (the low boarder obtained from the data measured by the ^{3}He-spectrometer was about 10 keV) as compared to the spectra of photoneutrons obtained in other studies [5]. The low–boarder of photoneutrons spectra presented in the review [6] in the vast majority of works varies from 1 to 2 MeV. A distinctive feature of these spectra is the presence of fast neutron components, the average energy of which is substantially higher than the average energy of neutrons originated from evaporation process.

Radial flux density distribution in the Ga target

The photoneutron spectra obtained in the center of gallium target and on its surface were integrated by energy to obtain the flux density values at corresponding points of the target. Obtained values are shown in Figure 6.

To verify experimental values of the ratio of flux density in the center and on the target surface obtained on the basis of the energy spectra, we carried out additional experiments in which we measured the activity of saturation of the samples of gallium in the center and on the surface of the irradiation device. For this purpose, Ga samples of known mass were placed in a specially made channel of the irradiation device located at a depth of 5 cm from the input window of the irradiation device and on its surface at a distance of 5 cm from the point of input of the electron beam at the target. After irradiation, the gamma-ray spectra of samples were measured at certain time intervals for a fixed time.

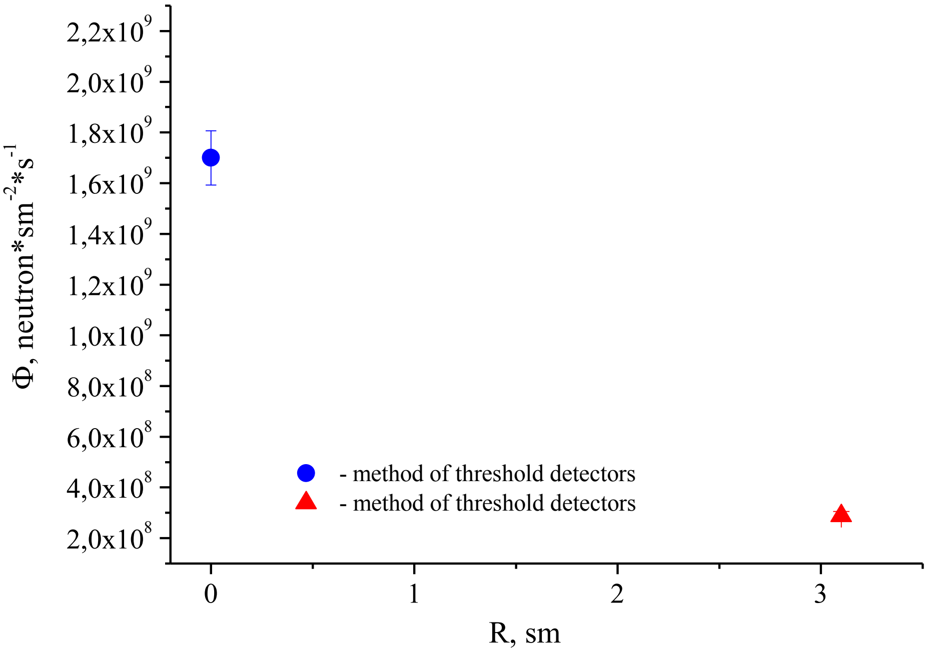

Figure 6. Radial distribution of photoneutrons flux density into gallium target at a depth of 5 cm from the entry point of the electron beam.

The resulting spectra were processed to obtain peak area of total absorption of gamma rays of a certain energy. In turn, the obtained values used to calculate the count rate depending on time. The results obtained for activity decay of the gallium samples set in the center and on the wall of the irradiation devices are shown in Figure 7. Processing the decay curves of activity of the gallium samples in the center and on the surface of the target by the least squares method yielded activity values of the samples at the moment of switching off the current of electrons (activity saturation).

Ratio between the activity of saturation of gallium samples for gamma rays with energies of 834 keV was 4,76 ± 0,24, which is consistent with the results of spectral measurements obtained with threshold detectors.

Measurements of characteristics of dose of the gallium target after irradiation session are produced by detection unit BDKS-96. A result of analysis the received data was found that the background level on the surface of the irradiated target gallium achieved within 4.77 hours.

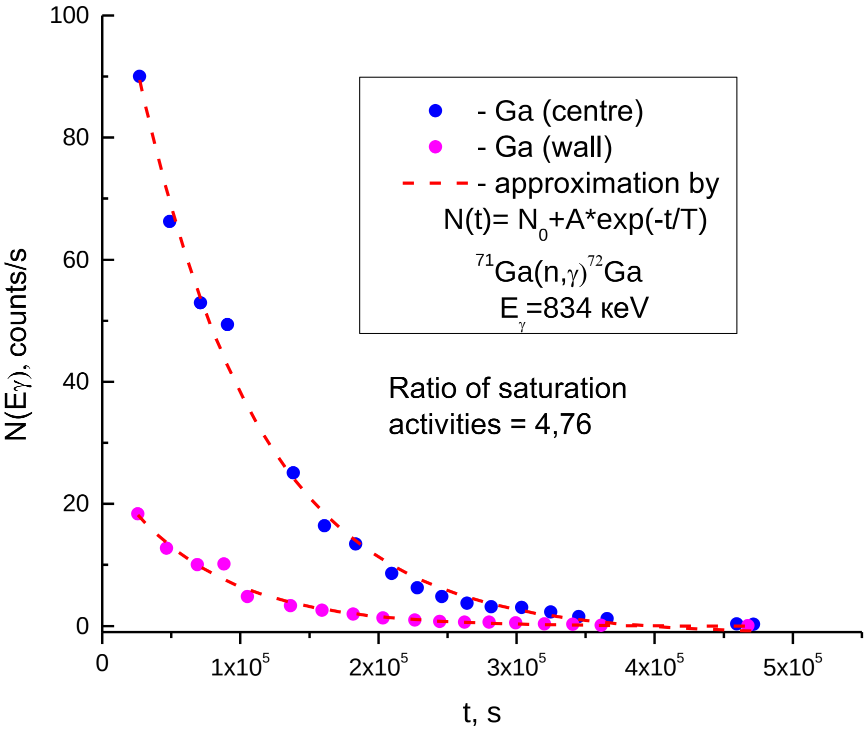

Figure 7. Time dependence of activity of gallium samples placed in the center and on the wall of the Ga target.

Conclusion

In the present work the energy spectrum of photoneutrons generated into a gallium target by electrons with energies of 35 MeV in the energy range of 0.0001 eV to 16 MeV were measured by the activation method on the basis of the reaction rate of the following nuclear reactions: $^{56}Fe(n,p)^{56}Mn$, $^{58}Ni(n,p)^{58}Co$, $^{63}Cu(n,\gamma)$ ^{64}Cu, $^{113}In(n,\gamma)^{114m}$ In, $^{115}In(n,n')^{115m}In$ and $^{27}Al(n,\alpha)^{24}Na$. The ^{3}He-spectrometer was used to measure the spectrum of photoneutrons in the energy range from 10 to 700 keV. The average energy of the photoneutron spectrum is 2.17 MeV. The flux density in the centre of gallium target obtained on the basis of these data is $1.7 \cdot 10^9$ neutron$\cdot$cm$^{-2}\cdot$s^{-1} for the average electron current of 2.5 mA. The radial distribution of photoneutron flux density inside gallium target at a depth of 5 cm from the entry point of the electron beam was measured with the help of activation detectors. Ratio of the saturation activities of gallium samples placed in the centre and on the surface of the target for gamma rays with energy of 834 keV was 4,76 ± 0,24 that is consistent with the results of spectral measurements obtained with the activation detectors. Measurements made by BDKS-96 dosimeter show that the background level of gamma radiation on the surface of the irradiated gallium target is gained within 4.77 days.

References

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