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COUNTING EFFICIENCY OF REGISTRATION OF CONTRIBUTIONS OF FAST NEUTRON REACTION PRODUCTS BY DETECTORS BASED ON OXIDE SCINTILLATORS $ZnWO_4$, $Bi_4Ge_3O_{12}$, $CdWO_4$ and Gd_2SiO_5

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The results of the study of the contributions of the interaction reactions of fast neutron sources of ^{239}Pu-Be and ^{252}Cf to the counting efficiency of registration by oxide scintillators $CdWO_4$, $ZnWO_4$, $Bi_4Ge_3O_{12}$ and Gd_2SiO_5, presented. The amount of gamma quanta per input neutron emitted from final nuclei excited in the reactions of inelastic scattering $(n, n'\gamma)_{in}$, resonant scattering $(n, n)_{res}$ and capture $(n, \gamma)_{res}$ and radiation capture $(n, \gamma)_{cap}$ was measured. PMT R1307 operating in single-electron mode was used as a photodetector, the background rate was $\sim 5\cdot10^3$ s^{-1}. The measured efficiency ε for scintillators ø40×40 mm was 752 for ZWO, 532 for CWO, 37 for GSO, and 23 for BGO in "counts/neutron" units, measurement error rate ~ 3-5%. The formation of the detector response is influenced by the parameters of the scintillator nuclei, such as the values of the interaction cross sections in the resonance region, the density of nuclear levels of the final nuclei, the lifetime of excited nuclear states, the upper limit of the resonance region of the cross section, as well as the scintillation time and geometric parameters of the scintillators. A phenomenological model of the response of an oxide scintillator to fast neutrons is proposed.
Keywords: *Oxide scintillator, ZWO, BGO, CWO, GSO, Fast neutrons, ^{239}Pu-Be, Resonance capture, Counting efficiency, Density of nuclear levels, Single photoelectron mode*
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Creation of compact, highly-sensitive detectors of fast neutrons emitted by sources of the Pu-Be, ^{252}Cf type is an actual task. Small-sized neutron and gamma-neutron radiation monitoring systems, designed to combat unauthorized transportation of fissile and radioactive materials, are especially needed in such detectors.

Registration of fast neutrons is possible with organic scintillators of the stilbene type however, detectors based on them require rather complex electronics, as a rule, they are grown in limited sizes. Detectors based on scintillation plastic have an insignificant efficiency of recording fast neutrons, ε ~ 0.1, while the necessary sensitivity is achieved due to the increase in the geometric dimensions and mass of the detector. The shortage of ^{3}He and the high cost of manufacturing ^{3}He counters stimulate the development of detectors based on new principles of recording fast neutrons.

The development of new fast neutron detectors requires information on the contributions to the detector response of the products of various mechanisms of interaction of neutrons with scintillator nuclei, since the energy of a fast neutron upon interaction with the substance of the detector changes approximately 10^8 times, from ~ 11 MeV to 0.025 eV. In addition, the magnitude of the detector's neutron response, in addition to the nuclear subsystem, is influenced by the atomic subsystem of the scintillator, namely the scintillation time, which must be considered when analyzing the contributions of mechanisms and developing new types of neutron detectors.

Fast neutrons from radioactive sources with energy $E \leq 10$ MeV experience such types of interaction with scintillator nuclei as elastic and inelastic scattering, resonance scattering, resonance capture, radiation capture. Therefore, three conditional energy intervals are distinguished:

$\Delta E_{in} \sim 10$ MeV – 0.5 MeV, $\mathbf{(n, n'\gamma)_{in}}$:

$$n+A \dashrightarrow (A+1)^* \rightarrow A^* + \mathbf{n'} + \gamma_{\mathbf{prompt}} \rightarrow A + \gamma_{\mathbf{delayed}}$$

$\Delta E_{res} \sim 0.5$ MeV – 0.1 keV, $\mathbf{(n, n')_{res}}$ and $\mathbf{(n, \gamma)_{res}}$:

$$n + A \rightarrow (A+1)^* \rightarrow \mathbf{A} + \mathbf{n'};\ n + A \rightarrow (A+1)^* \rightarrow (\mathbf{A+1}) + \gamma_{\mathbf{delayed}}$$

$\Delta E_{cap} \sim 0.1$ keV – 0.025 eV, $\mathbf{(n, \gamma)_{cap}}$.:

$$n + A \rightarrow (A+1)^* \rightarrow (\mathbf{A+1}) + \gamma_{\mathbf{delayed}}$$

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In [1], it was shown that heavy elements can be used for calorimetry of fast neutrons, while the experimental efficiency of registration at the lowest registration threshold was ε ~ 0.4–0.5 instead of the expected ε ~ 0.1. Such a significant increase in the efficiency of the registration of high-energy neutrons was explained by the cascading action of the generation of secondary neutrons on heavy lead nuclei in the reaction of inelastic scattering. It was shown that the mechanism of inelastic scattering can be used to create highly efficient neutron calorimeters and neutron detectors of a new generation. In our work, heavy oxide scintillators such as ZWO (ZnWO4), BGO (Bi4Ge3O12), CWO (CdWO4), GSO (Gd2SiO5) were studied [2 -3, 26].

In these works, the inelastic scattering reaction $(n, n'\gamma)_{in}$ is the starting point that generates primary instantaneous and delayed gamma quanta from end-product nuclei and initiates the process of formation of secondary neutrons and multiple gamma quanta arising from the decay of excited states of final nuclei and increasing the statistics of events, generated by the primary fast neutron [4-7, 25,27-28].

At the first stages of research [8], the contribution of high-energy gamma quanta (E ~ 0.5-3 MeV) from the inelastic scattering reaction (n, n′γ) arising during the discharge of excited solid-state scintillator nuclei was carried out by small-sized oxide scintillators, V ~ 1 cm^3. At the same time, the efficiency of the registration of fast neutrons, measured by a spectrometric path with an integration time of τ = 30 us and a registration threshold of ~ 20 keV, was ε ~ 0.4 - 0.5 counts/neutron. Such a choice of the detector signal generation time was due to the need to suppress secondary cascade gamma quanta, and the small size of the detectors did not allow neutrons to reach the resonant energy range. Thus, for the reaction of inelastic scattering, the oxide scintillators looked practically equivalent.

In the future, in order to increase the counting efficiency of fast neutron registration, in our works [2, 3, 26] it was proposed to register multiple (cascade) gamma quanta generated between the highly excited transitions of final nuclei in the reactions of the interaction of neutrons with nuclei. For this purpose, it was proposed to register, in addition to the products of the inelastic scattering reaction, also the products of the resonance scattering and capture reactions.

The basis for such an approach is that the nuclei that make up some oxide scintillators (for example, Cd, W, Gd, Zn) have significant neutron interaction cross sections in the resonance region (~ 350 - 400 barn), while cross sections in the inelastic region are only ~ 2-3 barn.

In addition, in the reaction of inelastic scattering and resonance capture of secondary neutrons, both instantaneous gamma quanta from excited states of final nuclei (A+1)* with lifetimes of ~ 10^{-14} - 10^{-12} s are emitted, as well as delayed ones due to the lifetime consisting of final nuclei of several nanoseconds or more: – nucleus A* from the reaction $A(n, n'\gamma)_{in}A^*$ and (A+1)* from the capture reaction $A(n, \gamma)_{res}(A+1)^*$.

The intermediate (compound) nucleus (A+1)* receives the average excitation energy $E \sim S_n + E_{n\_kin}$ and, after the escape of the secondary neutron, which reduces the energy of the compound nucleus by the value of the neutron separation energy Sn, turns into the final nucleus A* with an excitation energy equal to energy of the incident neutron E_{n_kin}. At the same time, mainly gamma quanta with an energy of about 1-3 MeV are emitted, which are confidently registered by the electronic path even with moderate speed and sensitivity. Another secondary cascade gamma quanta is also produced, initiated by secondary neutrons from the reaction of inelastic scattering upon capture in the resonant region. That is why the cross section of the resonance zone becomes important.

It should be noted that in the reaction of inelastic scattering $(n, n'\gamma)_{in}$, the neutron ejected from the nucleus has a significantly lower energy (on average ~ 1-3 MeV) compared to the initial energy (~ 4-10 MeV), i.e. there is an effective slowing down of fast neutrons, which increases the probability of the subsequent capture of the escaped secondary neutron in the $(n, \gamma)_{res}$ reaction and the formation of a new compound nucleus.

In the case of capture of secondary slowed neutrons in the resonance region, it becomes possible to register additional instantaneous and delayed gamma quanta of small energies arising during the discharge of excited states of final nuclei (A+1)*, if the density of levels of excited nuclei is such that the distance between the levels exceeds excitation threshold of scintillator molecules.

Our estimate of the densities of the levels included in the considered scintillators, carried out in this work on the basis of the thermodynamic model of reactions [9, 10, 11], is consistent with estimates of the multiplicity of gamma quanta in resonance capture reactions and the data of works [12-13, 24], the results of the evaluation indicate the existence of low-energy gamma quanta emitted between the transitions of highly excited states of final nuclei, capable of effectively exciting the molecules of the oxide scintillator. The energy range of incident neutrons, for which the contributions of researched mechanisms of nuclear reactions involving neutrons can be realized, is also estimated.

To implement the possibility of registering low-energy gamma quanta from transitions between highly excited short-lived states of nuclei when their energy is close to the excitation threshold of scintillator molecules, for example, for ZWO, it was necessary to use a photodetector operating in the single-electron counting mode and to develop a high-speed, low-noise amplifier path operating in the current-voltage converter mode, which has a resolution time of ~ 1-2 ns (bandwidth Δf ~ 300 MHz), a high amplification factor (~ 3000), low noise and a low registration threshold. The average excitation energy of the scintillator molecule was estimated based on the light output of the scintillator. For example, for ZWO, the light output is ~ 10,000 photons/MeV, so the excitation threshold of the ZWO molecule is E_{thr} ~ 0.1 keV. To ensure the necessary time separation of signals from the interaction of primary and secondary neutrons, scintillation detectors with a sufficiently extended geometry were used, the thickness of the scintillator is ~ 40-50 mm.

Thus, the registration of cascades of gamma quanta of various energies in the range from a few MeV to hundreds of eV, genetically related to the input neutron emitted by excited nuclei, in the presence of a high-speed measuring path with a low registration threshold and a low noise level, made it possible to significantly increase the statistics of useful events, occurring per one input neutron.

Literary data also confirm the generation of a significant number of secondary cascade gamma quanta from inelastic scattering reactions [14], from radiation and resonance capture reactions [12, 15-16]. But, at the same time, it should be borne in mind that the data on the multiplicity of secondary gamma quanta, if not specified, were accumulated by the spectrometric technique during a long-time interval, and were processed in a delayed time mode.

In addition to theoretical estimates of the multiplicity of gamma quanta, this work developed a technique and provided measurements of the efficiency of recording fast neutrons, confirming the increase in the measuremented efficiency of the detector due to the use of secondary products of the resonance scattering reaction and capture.

The scintillator response model proposed in this paper also indicates the very significant role of the time of scintillation of the excited states of the atomic subsystem of the scintillator. The results of the estimation of the travel lengths of fast neutrons considering elastic and inelastic scattering point to a noticeable role of light oxygen nuclei, which are part of scintillators, for the process of slowing down neutrons.

1. EXPERIMENT

In Table 1 shows some characteristics of the investigated scintillators.

Table 1. Main characteristics of the investigated oxide scintillators

Scintillator	$ZnWO_4$	$CdWO_4$	$Bi_4Ge_3O_{12}$	Gd_2SiO_5
nucleus (Natural abundance) (cross section, barn)	part of nuclei in a molecule			
Zn (σ_{cap}= 1.079; σ_{res}=2.495; σ_{in}=0.491; σ_{el}=1.697; σ_{el_res}=97.22))	1/6			
W (σ_{cap}= 18.11; σ_{res}=355.1; σ_{in}=0.485; σ_{el}=2.978; σ_{el_res}=1118.5)	1/6	1/6		
Cd (σ_{cap}=2463; σ_{res}=71.935; σ_{in}=0.366; σ_{el}=2.650; σ_{el_res}=85.13)		1/6		
Gd (σ_{cap}= 48700; σ_{res}=398.3; σ_{in}=1.487; σ_{el}=1.937; σ_{el_res}=170.61)				2/8
Bi-209 (σ_{cap}= 0.0338 ; σ_{res}=0.1919;σ_{in}=0.392; σ_{el}=2.81; σ_{el_res}=144.48)			4/19	
Ge (σ_{cap}= 2.229; σ_{res}=5.935; σ_{in}=0.612; σ_{el}=1.686; σ_{el_res}=120.08)			3/19	
Si (σ_{cap}=0.160; σ_{res}=0,0834; σ_{in}=0.524; σ_{el}=0.734; σ_{el_res}=25.38)				
O (σ_{cap}= 0.00016 ; σ_{res}= 0.00016; σ_{in}=0.309; σ_{el}=0.957; σ_{el_res}=46.13)	4/6	4/6	12/19	5/8
Energy resolution,% (661.66 keV)	8.5	7	9	7.2
Density, g/cm^3	7.87	7.9	7.13	6.71
Effective atomic number	61	66	74	59
Light decay time, us	20	18	0.30	0.06/0.6
Light yield, pe/MeV	10000	20000	(8-10)*10^3	(8-12.5)*10^3
Gamma penetration depth, 1/e, cm (E_γ=0.662 MeV) 1/e, cm	1.44	1.45	1.40	1.49
Omission, (E_γ=0.662 MeV), a.u. (d = 4 cm)	0.062	0.064	0.058	0.069
The free path length of a neutron Σ, cm^{-1} Σ=Nσ, (E_n = 4 MeV)	0.389	0.373	0.390	0.394
Neutron penetration depth (1/e), 1/Σ, cm	2.571	2.682	2.563	2.540
Omission neutrons, a.u. (d = 4 cm)	0.211	0.225	0.210	0.207
Counting efficiency of registration of the fast neutrons, counts/neutron, ε, $(n,n'\gamma)_{in} + (n,\gamma)_{res}$,	574	443	19.3	22.2
Counting efficiency of registration of the fast neutrons, counts/neutron, ε, $(n,n'\gamma)_{in} + (n,\gamma)_{res} + (n,\gamma)_{cap}$	752	532	23	37
Size, mm	Ø 52x42	Ø 45x42	Ø 40x40	18x18x42

The structural scheme for measuring the efficiency of fast neutron registration in spherical geometry is presented in Fig. 1a, b [19]. In the neutron ^{239}Pu-Be source, accompanying gamma radiation from the reactions is present:

1) $^4He + {}^9Be \rightarrow {}^{13}C^* \rightarrow {}^{12}C^* + n \rightarrow {}^{12}C + \gamma$, ($^{12}C^*$, Eγ = 4.43 MэB);

2) $^4He + {}^9Be \rightarrow {}^{13}C^* \rightarrow {}^{13}C + \gamma$, ($^{13}C^*$, Eγ = 3.68 MэB).

The advantage of spherical geometry is the elimination of the need to introduce corrections for neutron scattering in the lead sphere, which is used to attenuate the accompanying gamma radiation from the source. The measurements used (α, n) a ^{239}Pu-Be source and a ^{252}Cf spontaneous fission neutron source with a flow of fast neutrons ~ 10^5 neutron·s^{-1}.

The source was placed inside a lead ball with a diameter of 100 mm with a channel to the center of the ball with a diameter of 20 mm. Photomultiplier R1307 Hamamatsu is used. The detector-source distance is 1000 mm. To weaken the external gamma background, a lead screen d = 5 mm is used.

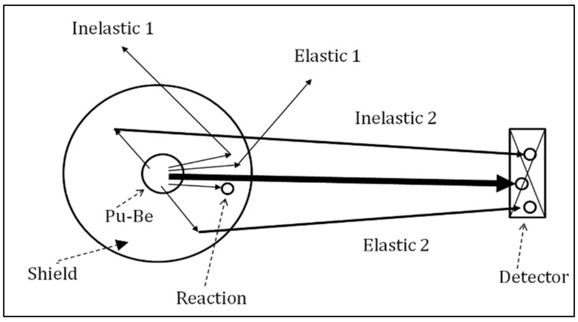


Figure 1a. Structural diagram of measurement of the counting efficiency of registration of fast neutrons in spherical geometry: 1 - photomultiplier R1307; 2 - scintillator under investigation; 3 – lead screen d = 5 mm for weakening the gamma background; 4 - lead layer Ø100 mm with a cylindrical channel Ø20×50 mm; 5 - 239Pu-Be neutron source; distance detector - source – 1000 mm

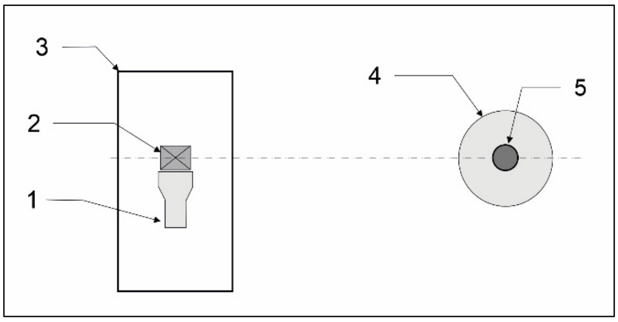


Figure 1b. The processes of scattering (→) and absorption (○) of fast neutrons in spherical geometry

The response to neutrons in the scintillators under study is formed by cascade gamma quanta emitted in the reactions of inelastic scattering $(n, n'\gamma)_{in}$, resonant scattering $(n, n)_{res}$ and capture $(n, \gamma)_{res}$, radiation capture $(n, \gamma)_{cap}$, as well as due to cascade gamma quanta that appear during the capture of secondary slowed neutrons. The value of the "count efficiency of neutron registration" (CENR) by oxide scintillators CWO, ZWO, BGO, GSO was measured in units of "counts/neutron", i.e. in units of the ratio of the detector count rate to the number of particles falling on the entire detector in 1s (the flow on the entire detector), i.e. CENR is the average number of pulses from the detector that fall on one input particle.

The source of neutrons is located in a lead sphere, which effectively scatters neutrons, both in elastic and inelastic scattering, while absorbing them very weakly, so that the number of neutrons emitted into space does not change and can be considered. The capture cross section of fast neutrons in Pb is ~ 1.11 mb.

Scattering in the direction of the detector, i.e. into the front hemisphere is completely compensated by backscattering into the detector from other points of the sphere, i.e. posterior hemisphere. Thanks to the spherical symmetry of the scatterer, the number of neutrons passing through each square centimeter of the spherical surface will also remain unchanged. Consequently, the lead spherical scatterer practically does not affect the number of scattered neutrons arriving at the detector, with the exception of neutrons absorbed in the lead layer due to the (n, γ) reaction. Thus, the scattering layer does not change the neutron count rate in the detector, performs the function of weakening gamma quanta, which leads to a decrease in the harmful influence of secondary gamma quanta from the Pu-Be source on the accuracy of measurements, reduces errors due to the "accumulation factor" of gamma quanta , scattered from the walls of the room. The contribution of scattered fast neutrons from the walls of the laboratory room was ~ 3%. Due to the increase in the effective size of the neutron source, it is necessary to carry out measurements at somewhat larger distances (~ 1 m) compared to the "narrow" geometry. Deviation from the law of inverse squares for a distance of 1.2 m does not exceed 3%.

The correction for the absorption coefficient of fast neutrons in the shielding sphere was measured by a LiI(Eu) detector for a source-detector distance of 2 m and amounted to ~ 2.5%. Scintillator size ^{6}LiI(Eu): Ø15 × 10 mm, ^{6}Li enrichment 96%. The thermal peak (α + t) for ^{6}LiI (Eu) had a gamma equivalent of 3.98 MeV, fast neutrons were registered in the energy range of 3.98 MeV ÷ 14 MeV.

The additional contribution from gamma quanta with energy $E\gamma$ = 4.43 MeV for the ZWO scintillator (correction for registration efficiency of gamma) amounted to ~ 7%, while the following was assumed: the γ/n ratio for the ^{239}Pu-Be source is 1, the transmission of gamma quanta with energy of 4.43 MeV by a lead sphere with a thickness of 50 mm ~ 9%, absorption of such gamma quanta in ZWO with an effective thickness of 46 mm ~ 76%.

The correction for the absorption coefficient of fast neutrons in the protective sphere was also measured using a ^{252}Cf source. A distinctive feature of the ^{252}Cf source is the absence of high-energy accompanying gamma radiation and a "softer" form of the spectrum, which can affect the efficiency of the excitation of the gamma quanta generation mechanisms. The difference in the measurement results for the ZWO scintillator was 11.8%, which can be explained by the presence of accompanying gamma quanta with an energy of E = 4.43 MeV (7%) and a harder form of the spectrum of the Pu-Be source, which leads to an increase in the output of secondary gamma quanta (4.8 %). The background attenuation coefficient in the energy range of 10–150 keV by a 5-mm-thick lead shield around the scintillator was ~3.

In order to reduce the influence of fluctuations of the cosmic neutron background, the measurement of the registration efficiency was carried out by five exposures - "100 s - background measurement, then 100 s - signal measurement". The statistical error for each exposure was ~1%.

On Fig. 2 shows the structural diagram of the measuring path, consisting of three channels - single-electron (counting channel) ($\tau \sim 1$ ns), fast spectrometric ($\tau \sim 50$ ns, Amptek DPP PX-5), slow spectrometric (linear spectrometric amplifier, $\tau \sim 1$ us). The photodetector is a Hamamatsu R1307 PMT, the rise time of the signal is ~ 7-8 ns. The single-electronic account mode is used to lower the registration threshold, increase sensitivity, and reduce the registration resolution time.

To amplify signals from PMT, a current-voltage converter was developed, Fig. 3, amplifying signals with a rise time of $\tau_{ris} \sim 0.5$ ns. The first stage is made according to the "current - voltage converter" scheme on OP type 4817, the input bias current is 2 nanoamps, the total voltage gain of the second - seventh stages is $k_{amp} \sim 740$, which made it possible to register low-energy (E ~ 0.1 keV) gamma quanta. As the output driver, the 7th stage on the opamp type 7171 was used.

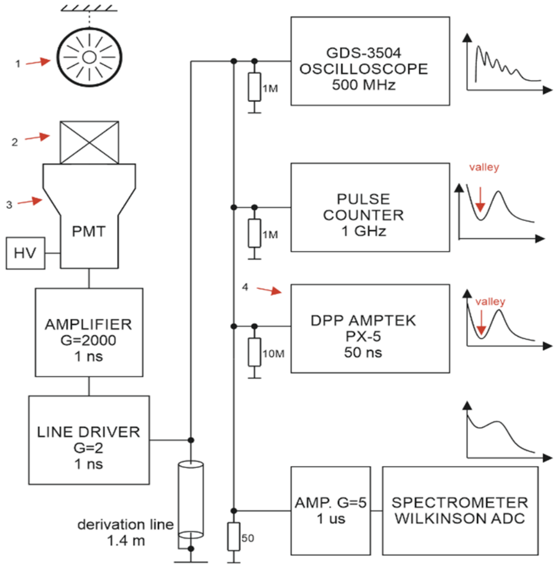


Figure 2. Structural diagram of the electronic path for measuring the counting efficiency of registration of fast neutrons by scintillators. 1 – neutron source, 2 – scintillator, 3 – PMT R1307, 4 – digital pulse processor

In order to reduce the dead time of the tract and quickly restore the baseline of the tract, shortening (differentiation) of signals using a cable delay line was used.

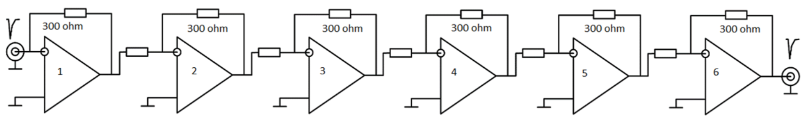


Figure 3. Structural diagram of a broadband pulse preamplifier on OP type ADA4817. The input resistors in the OP are 100 Ohm. The rise time of the first cascade is τris ~ 0.5 ns

The total dark noise of the PMT base line and the electronic preamplifier noise is 50 mV at a voltage of 1250 V on the PMT. The dark loading of the measuring path in the single-electron mode for this PMF sample and a ø45×40 mm scintillator is ~ 5×10^3 c^{-1}. A DPP Amptek digital processor, signal processing time of 50 ns, and a pulse counter with a bandwidth of $\Delta f \sim 1$ GHz were used as a recorder.

On Fig. 4 shows the response of the ZWO scintillator to neutrons from the ^{239}Pu-Be source in the single-electron (upper figure) and spectrometric ($r_{rise} \sim 1$ µs, lower figure) modes. The speed of the electronic path $\tau_{rise} \sim 1$ ns.

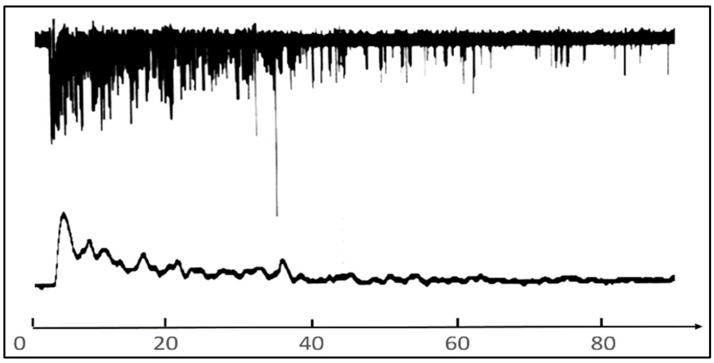


Figure 4. The response of the ZWO scintillator to fast neutrons from the ^{239}Pu-Be source in the single photoelectron ($\tau_{rise} \sim 1$ ns, upper figure) and spectrometric ($\tau_{rise} \sim 1$ µs, lower figure) modes. Here X is time, µs

On Fig. 5 shows the waveform at the output of the broadband channel during the registration of neutrons from the ^{239}Pu-Be source by the ZWO scintillator. The duration of the leading edge of the signals $\tau \sim 8$ ns is determined by the PMT parameters.

The hardware spectrum measured by DDP Amptek during irradiation of the ZWO scintillator with neutrons from the ^{239}Pu-Be source is shown in Fig. 6. Signal processing time is 50 ns.

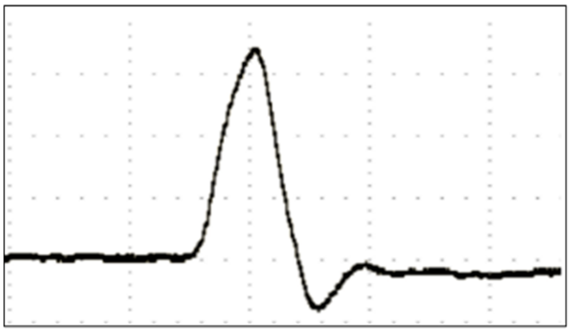


Figure 5. Response to neutrons from the ZWO scintillator at the output of the broadband channel. The dropout at the trailing edge of the signal is due to the shortening delay line, $2\tau = 20$ ns. Scale along the X axis is 25 ns/div

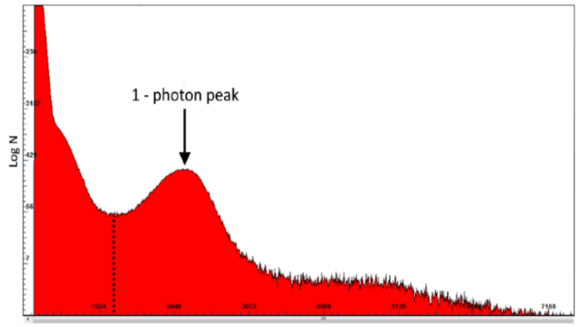


Figure 6. Hardware spectrum of the ZWO scintillator on PMT R1307, HV = 1250 V. Y axis – Log(Counts), X axis – channels

2. METHODOLOGY FOR THE STUDY OF CONTRIBUTIONS OF PRODUCTS OF REACTIONS WITH NEUTRONS TO THE SCINTILLATOR RESPONSE

The processes of generation of cascade gamma quanta and secondary neutrons in the reactions of elastic and inelastic scattering, resonant elastic scattering and capture of fast neutrons on the nuclei of the ZWO scintillator are schematically presented as follows:

– **elastic processes** – $(n, n')_{el}$: n+ (A) --> n' + (A); → n' -> . . .
$\sigma(n,n) = 0.957$b (O-16, E=14MeV)

– **inelastic processes** – $(n, n'\gamma)_{in}$: $n+(A) \to (A+1)^* \to n'+(A)^*_{final}$ → γ_{promt} → γ_{delay}
$\sigma(n, n'\gamma)_{in}$= 0,491 b (Zn);
$\sigma(n, n'\gamma)_{in}$= 0,485 b (W)

– **resonance elastic scattering** – $(n,n')_{res}$: n+(A) -> n'+ (A) → n' -> . . .
$\sigma(n', n'')_{res}$=113 b (Zn);
$\sigma(n', n'')_{res}$=136 b (W)

– **resonance capture** – $(n', \gamma)_{res}$: $n'+(A) \to (A+1)^*_{final} \to (A+1)$ → γ_{del}
$\sigma(n, \gamma)_{res}$=2,495 b (Zn);
$\sigma(n,\gamma)_{res}$ = 355.1 b (W)
$E_n \sim 8$ keV – 600 keV

– **resonance capture** – $(n', \gamma)_{cap}$: $n'+(A) \to (A+1)^* \to (A+1)$ → γ_{del}
$\sigma(n, \gamma)_{cap}$=1,079 b (Zn)
$\sigma(n, \gamma)_{cap}$=18.11 b (W)

In the reactions of inelastic scattering $(n, n'\gamma)_{in}$ on heavy nuclei of scintillators, such as Bi, W, Gd, Cd, neutrons can lose energy up to 2-3 MeV on average, slowing down considerably. Along with inelastic scattering, neutrons are effectively slowed down in the elastic scattering reaction on light nuclei that are part of oxide scintillators, such as ^{16}O.

Secondary neutrons can re-enter the resonance capture reaction $(n, \gamma)_{res}$. At the same time, the role of such nuclei as ^{64}Zn, in which $E_{res} \sim 0.6$ MeV, can be very significant. When neutrons interact with nuclei, both excited compound nuclei (A+1)* and final excited nuclei A* appear. In $(n, n'\gamma)_{in}$ reactions from (A+1)*, as a rule, 2-4 instantaneous gamma quanta with different energies fly out on average, but all of them will be registered in real time as one response due to the small the lifetime of compound nuclei.

An insignificant amount of gamma quanta with different energies from the final nuclei A* from the $(n, n'\gamma)_{in}$ reaction can be recorded separately in real time due to the low excitation energy, because some A* nuclei can be in an excited state for a few nanoseconds or more.

In the capture reactions $(n, \gamma)_{res}$ from the final nuclei (A+1)*, due to the significantly higher excitation energy than in the $(n, n'\gamma)_{in}$ reaction, from several tens to several hundreds of instantaneous gamma quanta fly out with different energy, but their registration on a real time scale will depend on the life time of excited states.

In works [12, 24] the values of the multiplicity of gamma quanta arising in the radiation capture of neutrons on nuclei are given, i.e. from excited nuclei (A+1)*. The value of the multiplicity of N_γ during the capture of thermal neutrons by nuclei of average atomic weight (these are Si, Zn, Ge isotopes) fluctuates within the limits of N_γ = 17-175 counts/neutron. For nuclei of high atomic weight (isotope W), the value of multiplicity varies within N_γ= 75-225 counts/neutron.

In the inelastic scattering of neutrons on the nuclei, there is also a certain multiplicity of gamma quanta emitted by final excited nuclei A*. This is confirmed by the data of works [14, 20]. The value of the multiplicity of N_γ in the interaction of neutrons with nuclei of average atomic weight is in the range of 4-14 counts/neutron. For heavy nuclei, the value of multiplicity N_γ is in the range of 70-130 counts/neutron.

The smaller value of the multiplicity in inelastic scattering compared to the resonance capture reaction can be explained by the fact that the final nuclei A* in inelastic scattering have a significantly lower excitation energy than the final nuclei of radiation capture due to neutron escape, which sharply reduces the excitation energy by the amount of nuclear binding energy.

It should be noted that separate hardware registration of cascaded gamma quanta in real time is possible only if the lifetimes of excited states, the intervals between cascade events, and the transport delay of secondary neutrons when moving into the scintillator material until the moment of capture are units of nanoseconds or more.

The use of a high-speed measuring path capable of processing signals with a rise time of ~ 0.5 nanoseconds, for example, a PMT path in the single photoelectron counting mode, contributed to a significant decrease in the energy threshold of registration to the energy of gamma quanta, which constitutes the excitation threshold of oxide scintillator molecules, about E_{thr} ~ 0.02 -0.1 keV.

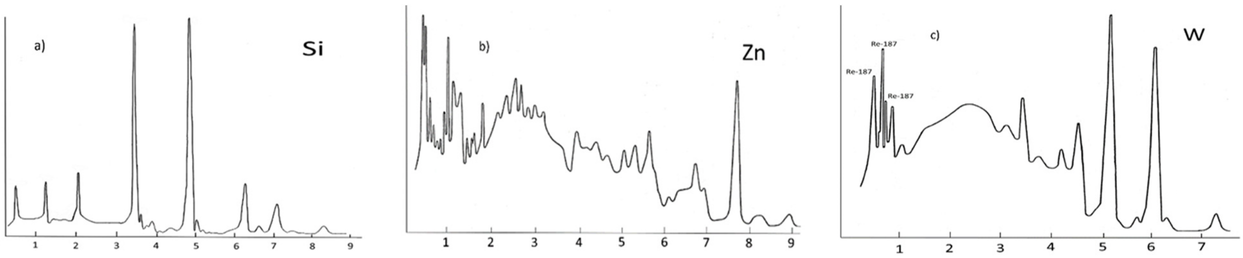
Lowering the threshold of registration made it possible to increase the contribution to the detector response of low-energy gamma quanta from high-lying transitions of excited final nuclei (A+1)*, especially from the reaction of resonance capture on heavy nuclei, for example W, Gd.

This paper presents the experimental results of measurements of contributions of multiple gamma quanta generated both in inelastic scattering and in resonance and radiation capture of neutrons by oxide scintillators. In addition, it was experimentally confirmed in our work [3] that the growth of useful statistics of events due to cascade gamma quanta, which are caused by incident neutrons, ultimately leads to an increase in the sensitivity of the neutron detector.

Secondary neutrons from the inelastic scattering reaction (n, n'γ) have significantly lower energy and after additional deceleration on light nuclei can be captured more effectively in the resonance region of Zn (E_{res_max}(Zn) ~ 600 keV) and W (σ_{el_res}=355.1 b), which will contribute to an increase in the multiplicity of secondary neutrons and an increase in the statistics of events.

2.1 ESTIMATION OF THE MULTIPLICITY OF GAMMA QUANTA FROM THE INELASTIC SCATTERING REACTION

In reactions with neutrons, four types of spectra are experimentally observed: – a monoline with an energy equal to the binding energy of a neutron, if an excited light nucleus emits a gamma quantum, it is in the ground state; – cascades of gamma quanta with discrete, energy-separated levels for light nuclei (Si) (Fig. 7a); – cascades of gamma quanta overlapping in the low-energy region, but still separated in the high-energy region (medium nuclei, Zn) (Fig. 7b). As the atomic weight increases in the region of 1.5-3.5 MeV, a wide hump appears, which is formed by gamma quanta from overlapping highly excited compound nuclei (heavy nuclei, W) (Fig. 7c) [16].



a b c

Figure 7. Types of spectra observed during the interaction of fast neutrons from the reaction $(n, n'\gamma)_{in}$ with nuclei of different atomic weights:
a) – light Si nucleus; b) – the nucleus of the average atomic weight of Zn, the hump in the region of 1.5-3.5 MeV begins to form; c) – heavy nucleus W, a hump is formed in the region of 1.5-3.5 MeV. The Y axis is the number of counts, the X axis is the energy, MeV [16]

The region of the spectrum in the interval 1.5-3.5 MeV is formed in the reaction of inelastic scattering by gamma transitions of excited states of the final nucleus. In this interval, the levels in the middle nuclei of Zn are resolved by energy. It can be seen in Fig. 7 c that in the interval of 1.5-3.5 MeV in the heavy nucleus W, the levels are not resolved in terms of energy, therefore, the energy of gamma-quantum transitions between these levels may not be large enough for reliable registration. This fact can be explained by the high density of excited levels in heavy final nuclei, because the responses of the levels located in the high-density zone merge into one instantaneous response of gamma quanta.

Both excited compound nuclei (A+1)* and final excited nuclei of scintillators A* participate in the formation of cascade gamma quanta. In addition, secondary neutrons from the reactions of inelastic scattering $(n, n'\gamma)_{in}$ and resonant scattering $(n, n)_{res}$ can experience capture again, thus allowing to separate in time the cascade events initiated by the primary neutron and increase the useful statistics of gamma quanta generated primary neutron.

In Table 2 presents the neutron cross sections of reactions of resonance capture $(n, \gamma)_{res}$, elastic resonance scattering $(n, n)_{res}$, inelastic scattering $(n, n'\gamma)_{in}$, radiation capture $(n, \gamma)_{cap}$ for a natural mixture of nuclear isotopes entering in the compound of the studied oxide scintillators $ZnWO_4$, $Bi_4Ge_3O_{12}$, $CdWO_4$, Gd_2SiO_5. [21-22].

Table 2. Cross sections of neutron reactions $(n, n'\gamma)_{in}$, $(n, \gamma)_{res}$, $(n, n)_{el}$, $(n, \gamma)_{cap}$ for a natural mixture of nuclear isotopes included in scintillators $ZnWO_4$, $Bi_4Ge_3O_{12}$, $CdWO_4$, Gd_2SiO

	0.0253 eV, (b) $(n, \gamma)_{cap}$	0.5 eV-10 MeV, (b) $(n, n)_{res}$	0.5 eV-10 MeV, (b) $(n, \gamma)_{res}$	14 MeV, (b) $(n, n'\gamma)_{in}$	14 MeV, (b) $(n, n)_{el}$	Threshold, keV $(n, n'\gamma)_{in}$	E_{res_max} keV
(A)	(A+1)	(A)	(A+1)	(A)	(A)		
$_{48}$Cd-nat	2463.26	85.13	71.935	0.366	2.650	247.6	8
$_{74}$W-nat	18.11	1118.48	355.1	0.48505	2.798	46.7	6
$_{30}$Zn-nat	1.079	97.224	2.4955	0.49025	1.697	94.7	600
$_{83}$Bi-nat	0.0342	144.48	0.19196	0.3923	2.810	900.7	200
$_{32}$Ge-nat	2.229	120.08	5.9348	0.61168	1.686	0.595	15
$_{64}$Gd-nat	48699	170.61	398.3	1.4872	1.937	54.88	8
$_{14}$Si-nat	0.1604	25.378	0.0834	0.52419	0.734	1.779	1800
$_{8}$O-16	1.6e-4	46.13	1.6e-4	0.3095	0.957	0.71	

In our opinion, the most significant parameters of the indicated nuclei, which are used in the analysis of deposits of neutron interaction products, are the resonance capture cross section, the density of nuclear levels of the final nuclei, and the elastic scattering cross section in the resonance region $(n, n)_{res}$.

It should be noted that since the Gd and W nuclei have similar parameters of the resonance capture zone (sections and the upper limit of the resonance zone), then, from the point of view of the nuclear subsystem, the GSO and ZWO scintillators should have approximately the same CENR, but this is not observed in reality. Apparently, an equally important role in the formation of the scintillator response is played by the atomic subsystem of the scintillator, i.e., time of scintillation. Indeed, the ratio of the efficiencies of the scintillators ZWO – $\varepsilon = 574$ cnt/neutron and GSO - $\varepsilon = 22.2$ imp/neutron correlates with the ratio of the durations of the scintillation times: ZWO – $\tau = 20$ us and GSO - $\tau = 0.6$ us.

The work [13] (Fig. 8) shows the densities of the levels of final nuclei depending on the mass number of nuclei. Ellipses highlight regions of ^{64}Zn and ^{182}W nuclei. The upper dashed line is the density of levels corresponding to the transition energy equal to E ~ 0.001 keV – this is the lower threshold of the single-electron registration mode, the lower dashed line is the average excitation energy of the ZWO molecule, E ~ 0.1 keV. It is possible to conclude that practically all gamma quanta from reactions of inelastic scattering and resonance capture on Zn and W nuclei can be registered in the single-electron mode and will contribute to increasing the counting efficiency of registration.

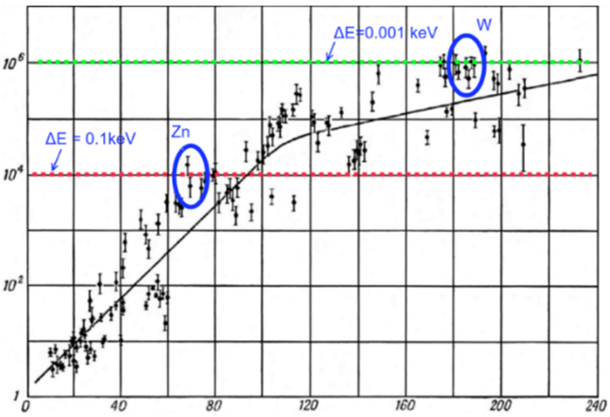


Figure 8. Density of levels of final -state nuclei ρ depending on the mass number of nuclei. Here Y is the density of levels, MeV^{-1}; here X is the mass number of nuclei. Ellipses highlight regions of ^{64}Zn and ^{182}W nuclei. The upper dotted line is the lower threshold of registration E_{th}~ 0.001 keV in the single-electron mode, the lower dotted line is the average excitation energy of the ZWO molecule, E~ 0.1 keV [13]

In this paper, based on the thermodynamic model of nuclear reactions, some parameters of the levels of the final nuclei included in the studied scintillators [9-11] are estimated. The density of the nuclear levels ρ of the excited states of the final nuclei from inelastic scattering reactions $(n, n'\gamma)_{in}$ and resonant capture $(n, \gamma)_{res}$ on the nuclei are estimated. The working interval of energies of incident neutrons, in which gamma quanta are generated, registered by the photodetector, the energy of the secondary neutron is estimated. (Table 3). The possibility of a statistical description of nuclei is associated with a large number of system states in the energy interval of averaging, excited in compound nuclei and final nuclei during neutron capture.

The basis of the model of the reaction $a + A -> C^* -> b + B$, passing through a compound nucleus, is the description of the distribution of ejected particles by energy (spectrum): $W(E) \sim Eb\ \sigma_C(E)\ \rho_B(E_{Bb} - E)$, where

E is the energy of the ejected particles, $\sigma_C(E)$ is the formation cross section of the compound nucleus, $\rho_B(E_{Bb} - E)$ is the density of levels of the final nucleus, $E_B = E_{Bb} - E$ is the excitation energy of the final nucleus, E_{Bb} is the energy of particle b, E_{Bb}^{max} is the maximum energy b corresponding to the formation of the final nucleus B in the ground state, E_a is the energy of the incident particle.

Table 3. Comparative evaluation of the parameters of light and heavy final nuclei excited by fast neutrons in the reactions of inelastic scattering and resonance capture

Reaction A(a, b)B	$^{64}Zn(n, \gamma)_{res}{}^{65}Zn$		$^{64}Zn(n, n'\gamma)_{in}$		
The energy of the incident neutron E_a, MeV	0,01	0,600	1.3	5	12.5
Neutron attachment energy	7.981		7.981		
1 level energy E_1	-	-	0.992		
Excitation energy compound nuclei, MэB	7.99	8.58	8.29	12	19.5
Excitation energy end nuclei state, макс., E_B, MeV	-	-	0.31	4.0	11.5
E^*_{Bb} max, max energy b, (n′)	-	-	0.31	4.0	11.5
End nuclei temperature, Θ, MeV	1.348	1.397	0.27	0.96	1.63
Level density ρ, MeV^{-1}	2201	2937	106	259	10200
Levels distance, D=1/ρ, keV	0.45	0.34	9.4	3.9	0.1
$E_{kin.\ out.}$ (n′), 2Θ, MeV	-	-	0.53	1.92	3.26
Reaction A(a, b)B	$^{182}W(n, \gamma)_{res}{}^{183}W$		$^{182}W(n, n'\gamma)_{in}$		
The energy of the incident neutron E_a, MeV	0,001	0,01	1.01	2.0	2.58
Neutron attachment energy	6.191		6.191		
1 level energy E_1	-	-	0.1001		
Excitation energy compound nuclei, MэB	6.19	6.20	7.1	8.09	8.67
Excitation energy end nuclei state, макс., E_B,MeV	-	-	0.91	1.90	2.48
E^*_{Bb} max, max energy b, (n′)	-	-	0.91	1.90	2.48
End nuclei temperature, Θ, MeV	0.711	0.711	0.27	0.39	0.45
Level density ρ, MeV^{-1}	9.6e+05	9.7e+05	944	4200	9710
Levels distance, D=1/ρ, keV	0.001	0.001	1.06	0.24	0.1
$E_{kin.\ out.}$ (n′), 2Θ, MeV	-	-	0.55	0.79	0.90

Considering the nucleus as a degenerate Fermi gas, the Weiskopf formula can be used for $\rho(E_B) = (C/E_B^2)\,exp(2\sqrt{aE_B})$, where E_B is the excitation energy of the final nucleus, a constant C ~ 1, the constant a was determined by the density of single-particle compounds on the Fermat surface according to the formula $a = \left(\frac{\pi^2}{2}\right)\left(\frac{A}{E_F}\right), E_F \sim 37\ MeV$. The temperature of the final nuclei B was estimated for a given excitation energy according to $\theta = \sqrt{E_B/a}$. The temperature of the compound nuclei C was estimated as $\theta = \sqrt{E_{Bb}^{max}} = \sqrt{E_a/a}$.

The probability of recording gamma quanta from inelastic scattering reactions on W nuclei is significantly lower due to the high density of W levels. Despite the high gamma multiplicityN_γ for W, from the results of the estimates given in Tab. 3 shows that the working interval of W nuclei (ΔE=1-2.6 MeV) is significantly smaller than that of Zn (ΔE= 1.3-12.5 MeV). Only gamma quanta from the discharge of some long-lived states can contribute to multiplicity, and the registration of short-lived low-energy gamma quanta from highly excited states requires a highly sensitive single-electron path.

From Tab. 3 shows that low-energy cascade gamma quanta with energy from E ~ 0.1 keV and higher are generated in transitions of highly excited states of final nuclei (neutron energy range E ~ 1.3 – 12.5 MeV for Zn and ~ 1.0 - 2.6 MeV for W). At the same time, the final nuclei of average weight Zn* in the inelastic scattering reaction have the density of nuclear levels from $1.06*10^2$ MeV^{-1} to ~$1.0*10^4$ MeV^{-1}, which corresponds to the transition energy from 9.4 keV to 0.1 keV. Gamma quanta in this energy range can excite ZWO molecules and be recorded in the single-electron mode.

In the final heavy nucleus - W* in the reaction of inelastic scattering, the density of nuclear levels ranges from 944 to ~$9.7*10^3$ MeV^{-1} in the energy interval of incident neutrons 1-2.6 MeV, which corresponds to the distance between levels from 1 keV to 0.1 keV. Gamma quanta in this energy range can also excite ZWO molecules and be recorded in the single-electron mode.

Consequently, model calculations indicate that in inelastic scattering reactions, the multiplicity of cascaded gamma quanta of final nuclei of medium weight will be greater than that of final heavy nuclei due to a wider operating range of neutron energies.

At the same time, the energy of the cascade gamma quanta of final medium-weight nuclei will be greater than for heavy nuclei due to the lower density of levels in excited medium-weight nuclei, i.e. the reaction of inelastic scattering from the point of view of neutron detection is more productive for nuclei of medium weight, if the threshold of registration of electronics is big.

The operating range of neutron energies for Zn nuclei is ΔE~ 11 MeV (from E~ 1.3 to 12.5 MeV), and it is limited from below by the excitation energy, and from above by the density of nuclear levels of the order of 10^4 MeV^{-1}. The operating range of neutron energies for W nuclei is much narrower, ΔE ~ 1.6 MeV (from E ~ 1 to 2.6 MeV) due to the lower value of the nucleon separation energy in W (6.191 MeV) than in Zn (7.981 MeV).

2.2. ISOLATION OF THE CONTRIBUTION OF INELASTIC SCATTERING

In inelastic scattering through a compound nucleus n + A -> (A + 1)* -> A* + n' + γ_{prt} -> A + γ_{del}, two types of emitted gamma quanta are distinguished. First, these are instantaneous gamma quanta of the discharge of the compound nucleus (A+1)*, since they are emitted during the short lifetime of the compound nucleus (~ 10^{-14} s). As a rule, the number of them is small, from 1 to 4 counts/neutron. [20]. Their separate registration is practically impossible, all the energy of nuclear decay is concentrated in one pulse of the scintillator. Secondly, these are delayed gamma quanta discharges of excited long-lived states of a final nuclei, for which separate registration is possible, and which form cascades of gamma quanta.

The work [14] presents data on the multiplicity of cascade gamma quanta in inelastic reactions with fast neutrons. For example, the gamma multiplicity of final nuclei $^{64,66,68}Zn$ is Nγ = 18, 8, 7 counts/neutron, respectively. The gamma multiplicity of nuclei $^{182,184,186}W$ is Nγ = 126, 71, 86 counts/neutron, respectively. The gamma multiplicity of nuclei $^{156,158,160,155,157}Gd$ is Nγ=15, 17, 14, 11, 7 counts/neutron respectively. The gamma multiplicity of $^{110,112,114}Cd$ nuclei is Nγ = 62, 62, 71 counts/neutron, respectively.

Gamma quanta of the discharge of compound nuclei are quite simply isolated, for example, by using small-sized scintillators, or by filtering the signal stream, i.e. by an integrated filter with a time constant from units to tens of microseconds. Integration provides an efficient combination of cascaded gamma quanta into a single signal and eliminates signal delays during the deceleration of secondary neutrons in d ~ 4-5 cm samples. As a rule, the gamma multiplicity remains low and the efficiency does not exceed the value of 0.5–0.6 counts/neutron for small-sized (d ~ 1 cm) of scintillators and is close to 1 counts/neutron for scintillators with a size of d ~ 4-5 cm. When using a highly sensitive broadband preamplifier and integrating the signal with a spectrometric amplifier (τ ~ 1 ns + 1 us), the counting efficiency of registration remains practically at the same level - 3 - 4 counts/neutron for sizes d ~4 cm.

Thus, the filtration mode with the appropriate selection of the formation time value effectively suppresses cascade gamma quanta of low energies from highly excited transitions in the final nuclei, i.e. estimate the contribution of the inelastic scattering mechanism to the counting efficiency of registration. For example, the efficiency for small-sized ZWO scintillators with a size of 10×10×10 mm in the spectrometric (i.e., without the use of a broadband preamplifier) mode of τ=1 us is ~ 0.73 counts/neutron, and increases to only 2.3 counts/neutron for a large-sized ZWO with a size of Ø52×42 mm with the use of a broadband preamplifier (mode τ=1 ns+1 μs, amplification factor ~3000). Similarly, in BGO and GSO scintillators in the filtration mode τ=1 ns+1 μs, the mechanism of inelastic scattering $(n, n'\gamma)_{in}$ is also distinguished, while the recording efficiency ε_n ~ 1 counts/neutron for BGO and ε_n ~1.9 counts/neutron for GSO.

Due to the fact that the inelastic scattering reaction produces secondary neutrons with significantly reduced energy, they are more likely to enter the resonance capture reaction on the nuclei. Also, due to the transport delay in time of the secondary neutron until the possible subsequent capture, the use of a high-speed counting path allows to increase the number of registered cascade gamma quanta from the capture reaction.

2.3. ESTIMATION OF THE MULTIPLICITY OF GAMMA QUANTA FROM THE RESONANCE CAPTURE REACTION

The contribution of gamma quanta from the reactions of inelastic scattering $(n, n'\gamma)_{in}$ and resonance capture $(n, \gamma)_{res}$ to the spectrum of gamma radiation on heavy and medium nuclei was studied in [16]. On Fig. 9 a, b show the averaged spectra of gamma radiation from the reactions of inelastic scattering $(n, n'\gamma)_{in}$ at the excitation energy E_n=7.5 MeV and resonance capture $(n, \gamma)_{res}$ at the neutron energy En=0.1 MeV on the heavy nucleus W and the medium nucleus weights of Cu.

It can be seen that the N_γ-multiplicity from the inelastic scattering reaction (n,n′γ)in on nuclei of medium and heavy weight exceeds the Nγ-multiplicity from the resonance capture reactions $(n, \gamma)_{res}$.

The fact that the increase in energy release after the capture of a resonant neutron with an energy of E = 0.1 MeV for the medium Cu nucleus was noticeably higher than for the heavy W nucleus can be explained by the higher density of the final W nucleus, which means the emission of lower-energy gamma quanta from excited levels of final nuclei W, for the registration of which the sensitivity of the electronic path was not high enough, therefore, with the same registration threshold, significantly less of them will be registered from W than from Cu.

This is confirmed by our estimates according to the evaporation model, which give the value of the density of the levels of the final nuclei in the capture reaction for W – ρ = $9.6\cdot10^5$ MeV^{-1}, and for Zn – ρ = 2.2-$2.9\cdot10^3$ MeV^{-1}. In addition, model estimates performed for the inelastic scattering reaction give a value of the density of levels for Zn nuclei that is lower than for W nuclei, and in a wider energy interval (Zn, ΔE=1.3 ÷ 12.5 MeV; W, ΔE=1 ÷ 2.6 MeV) (see Table 3).

It should be noted that the multiplicity of Nγ, as a rule, is measured in a spectrometry mode, i.e. in a delayed mode, so it may not agree with the value of the estimated registration efficiency, which is measured in real time.

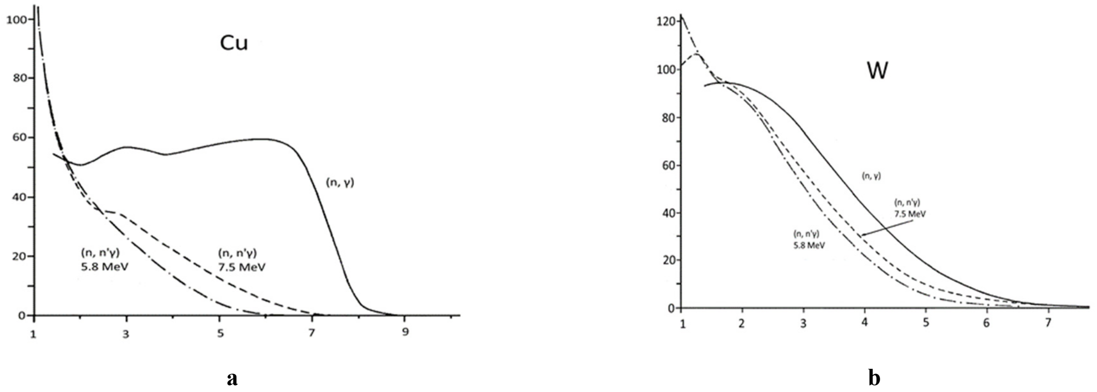


Figure 9. Averaged spectra of gamma radiation from reactions $(n,\gamma)_{res}$ and $(n,n'\gamma)_{in}$. a) – heavy nucleus W, solid line – $(n, \gamma)_{res}$, dashed and dash-dotted – $(n, n'\gamma)_{in}$; b) – nucleus of average atomic weight Cu, solid line – $(n, \gamma)_{res}$, dashed and dash-dotted – $(n, n'\gamma)_{in}$; The spectra were measured using a NaI crystal. The spectra (n, γ) were measured at neutron energy E = 0.1 MeV. Here Y is the number of accounts (E*ρ(E)), i.e. conditionally, Nγ is multiplicity), and X is MeV [16].

In our experiments on measuring the efficiency of recording fast neutrons by scintillators containing nuclei of medium and high atomic weight, the contribution of resonance capture (ε = 574 counts/neutron) exceeds the contribution of inelastic scattering (3 counts/neutron) to the experimental efficiency of registration by more than 2 orders.

This fact can be explained by the combined effect of the following factors: – high sensitivity and speed of the electronic circuit operating in single-electron registration mode; – the generation of sufficiently slowed secondary neutrons from the reactions of inelastic scattering and resonant scattering; – additional slowing down of neutrons on light oxygen nuclei; – by the large resonance capture cross-section of heavy W nuclei (σ_{res} = 361.6 b) and the emission of low-energy, short-lived cascade gamma quanta in the transitions of highly excited states of the final nucleus; – contributions of long-lived states of the final nucleus (A+1)*, for example, W-183, E=453 keV, τ=18 ns, W-185, E=244 keV, τ=19.3 ns; – the contribution of a significant amount consists of a lifetime of ~ 1-2 ns; – a considerable time of luminescence of the scintillator ZWO (20 us).

It should be noted that the low efficiency of registration in BGO can be explained by the absence of resonances in the heavy Bi nucleus - σ_{res}=0.192 b and the insignificant value of the scintillation time - τ_{scint} = 300 ns, and in GSO it can be explained by the small value of the scintillator scintillation time - τ_{scint} = 600 ns compared to τ_{scint}= 20 us for ZWO. Resonance capture on nuclei of medium weight (Z_n, σ_{res} = 2.49 b) cannot be productive due to the low cross section in resonance zone.

Also, a significant increase in the counting efficiency of registration for ZWO – with ε = 574 counts/neutron to ε = 752 counts/neutron with additional slowing down of neutrons flying out of the scintillator confirms the fact that the main mechanism for increasing the registration efficiency is capture of neutrons, primarily resonant, then, as the neutrons slow down – radiation capture.

In addition to the fact that the density of nuclear levels plays an important role in the formation of the response of the scintillator to neutrons, one should also consider the fact that sufficiently long-lived states can be excited in final nuclei in the resonance capture reaction on W nuclei, for example, W-183, E=453 keV, τ=18 ns, W-185, E=244 keV, τ=19.3 ns. Also, long-lived states can be excited in the final nuclei of inelastic scattering W, for example, E=2230 keV, τ=1.4 us, E=1286 keV, τ=8.3 us.

In medium-weight Zn isotopes, long-lived states can be excited in the final nuclei of resonance capture, for example, Zn-65, E = 53.9 keV, τ = 1.6 us; Zn-67, E = 93 keV, τ=9.2 us.

Therefore, it can be noted that the results obtained in our experiments to determine the efficiency of registration of oxide scintillators using a high-speed electron path do not contradict the results of work [16] on increasing the contribution to the energy release of scintillators due to the neutron capture reaction.

2.4. ISOLATION OF THE RESONANCE CAPTURE CONTRIBUTION

In resonant capture through the compound nucleus n+A -> (A+1)* -> (A+1)+γ_{del} , as in the case of inelastic scattering, the appearance of delayed gamma quanta of the discharge of excited long-lived states of the final nucleus (A +1), for which separate registration is possible and which form cascades of gamma quanta, can be observed.

In work [12, 24] values of the multiplicity of gamma quanta arising in the radiation capture of neutrons on nuclei, i.e. from excited state nuclei (A+1)*. The value of the multiplicity of N_γ during the capture of thermal neutrons by nuclei of average atomic weight (these are Si, Zn, Ge isotopes) fluctuates within the limits of Nγ = 17-175 counts/neutron. For nuclei of high atomic weight (isotope W), the multiplicity varies within the range of Nγ =75–225 counts/neutron. It is necessary to keep in mind that data on the multiplicity of secondary gamma quanta, unless otherwise specified, are accumulated by the spectrometric technique during a long-time interval, and are processed in a delayed time mode. In this case, the registration threshold is usually ~ 1-10 keV units and higher. This leads to the fact that there is always a significant discrepancy in the value of the efficiency ε (CENR) and the multiplicity Nγ.

The estimation of the level densities in the resonance capture reaction by the studied scintillators based on the thermodynamic model of reactions (see Tab. 3) gives a very high value of the level density for heavy nuclei – approximately 10^6 MeV^{-1}. Therefore, the value of the observed counting efficiency of registration can be explained by the total contribution as long-lived states of the final nucleus (A+1), for example, W-183, E=453 keV, τ=18 ns, W 185, E=244 keV, τ=19.3 ns, as well as a significant number of low-energy gamma quanta from transitions with a lifetime of ~ 1-2 ns. In this case, the use of a high-sensitivity, high-speed, low-noise measurement path operating in the single-electron mode (τ=1 ns) makes it possible to effectively register gamma quanta of short-lived compounds, which leads to an increase in the efficiency (CENR) of neutron registration. It should be noted that when switching to the spectrometric mode, when suppressing short-lived gamma quanta, the efficiency decreases by more than two orders of magnitude - from ε=574 counts/neutron to ε=3 counts/neutron for ZWO.

Also, the increase in the contribution of resonance reactions contributes to the increase in the number of slowed down secondary neutrons from the reactions of inelastic scattering and resonance scattering, the presence of oxygen nuclei, which participate in the effective slowing down of neutrons.

Thus, for effective registration of the products of the resonance capture reaction - cascade gamma quanta - it is necessary to use a specialized broadband electronic path operating in the single-electron mode. In addition, scintillators should contain nuclei with a high cross-section of resonant capture.

2.5. CONTRIBUTIONS TO THE EFFICIENCY OF RECORDING GAMMA QUANTA OF SCINTILLATOR NUCLEI OF DIFFERENT ATOMIC WEIGHTS

Based on the experimental results of measuring the efficiency of recording fast neutrons of ^{239}Pu-Be and ^{252}Cf sources by GSO (ε=22.2 counts/neutron) and ZWO (ε=574 counts/neutron) scintillators and estimates of the densities of the levels of final nuclei according to the thermodynamic model for of Zn and W nuclei in the energy range of incident neutrons 1 – 2.6 MeV (W) and 1.3 – 12.5 MeV (Zn), it can be concluded that nuclei of average atomic weight (for example, Zn) provide a significant decrease in the energy of secondary neutrons from the inelastic scattering reaction, and at the same time, they can generate gamma quanta from transitions of final nuclei with energy from ~0.1 keV to ~9.4 keV. Resonance capture on medium nuclei (Zn) can also contribute to gamma multiplicity both in the presence of long-lived stable final nuclei and generate cascade gamma quanta from transitions of final nuclei with energy from ~0.34 keV to ~0.45 keV. But in this case, one should take into account the insignificant value of the cross section for the inelastic scattering and resonant capture of nuclei of middle weight.

A more important role in increasing the efficiency of registration is played by heavy nuclei with a high cross-section of resonance capture - W, Gd nuclei. Unlike BGO (efficiency ε=19.3 counts/neutron), which includes heavy nuclei (Bi, σ=0.1919 b, short scintillation time – τ=300 ns), which do not have resonances, the ZWO scintillator (efficiency ε = 574 counts/neutron) has a significant a higher multiplicity of secondary gamma quanta, which can be explained by a combination of resonant capture (W, σ_{res}=361.6 b) and a long scintillation time (20 μs).

Heavy nuclei (W), according to estimates of the level density (Table 3), contribute to the efficiency of neutron registration (CENR) in the inelastic scattering reaction in a narrow range of energies (E~ 1–2.6 MeV) (D ~ 1.06–0.1 keV).

The contribution to the gamma multiplicity of the resonance capture reaction on heavy W nuclei is provided by both long-lived states of final nuclei, for example, τ=18 ns, τ=19.3 ns, and a significant amount of low-energy gamma quanta from transitions with a lifetime of ~ 1-2 ns , which is confirmed by the high density of levels ($\sim 9.6 \cdot 10^5$).

The use of a high-speed, highly sensitive measuring path operating in the single-electron mode (τ~1-2 ns) can ensure the registration of short-lived cascade gamma quanta up to energy ~0.1 keV and below, emitted by final heavy nuclei from the reactions of inelastic neutron scattering and resonance capture secondary neutrons.

2.6. DELAYED GAMMA QUANTA FROM FINAL NUCLEI INCLUDED IN THE ZWO SCINTILLATOR

In Tab. 4 presents data on the lifetimes of Zn and W isotopes excited in the reactions of inelastic scattering and resonance capture, which are part of the ZWO scintillator, which were used in the analysis of the results of measurements of the efficiency of fast neutron registration [23].

It can be noted that despite the high multiplicity of Nγ, which is significantly greater than unity for heavy nuclei and sufficiently large for nuclei of average atomic weight, the number of long-lived levels in the isotopes of the heavy nucleus W is 18, of which 7 are excited in the capture reaction, 11 in the reaction of inelastic scattering. The number of long-lived levels in isotopes of the middle nuclei of Zn is much smaller - 5, of which 3 are excited in the capture reaction, 2 - in the inelastic scattering reaction. The remaining states of the isotopes of both nuclei undergo discharge for times less than 1 ns. We note that it is due to the registration of gamma quanta of the discharge of short-lived compounds with lifetimes τ ~ 1 ns and less that the contribution of resonance capture increases, since long-lived compounds (18.4 ns, 8.3 μs, 19.3 ns, Table 4) in the final nuclei of the capture will not enough to form the necessary multiplicity (ε~574 counts/ neutron for ZWO).

Thus, on the basis of estimates based on the thermodynamic model and the analysis of the experimental data obtained in this work, it is possible to conclude about the possibility of increasing the efficiency of recording fast neutrons by oxide scintillators containing nuclei of high atomic weight and having a large cross section of resonant capture, due to the registration of multiple gamma quanta of final nuclear from resonance capture reactions, if it is possible to register delayed gamma quanta due to the transport delay of secondary neutrons in the crystal provided by the necessary dimensions of the scintillator crystal, especially in the presence of a high-speed, highly sensitive recording path operating in the single-electron mode and allowing to register gamma quanta of delayed nuclear states with a resolution no worse than 0.5-1.0 ns.

For the GSO scintillator, which has practically the same cross-section value in the resonance region as that of the ZWO (σ(Gd) = 398.3 b, σ(W) = 355.1 b), but a significantly smaller value of the efficiency of fast neutron registration – ε(GSO) = 22.2 counts/neutron compared to ε(ZWO)= 574 counts/neutron, this can be explained by the short time of GSO scintillations (τ ~ 600 ns, long-lived component of scintillation). It should be noted that the scintillation time plays an important role in the formation of the registration efficiency value, and in some cases is the determining value. This is indicated, for example, by a comparison of the neutron registration efficiencies of BGO and GSO scintillators, which have practically comparable values of the registration efficiency, although the values of the resonance cross sections in BGO and GSO differ by hundreds of times.

Table 4. Lifetimes, multiplicity of excited states in reactions of inelastic scattering and resonance capture of Zn and W isotopes included in the ZWO scintillator

$(n, n'\gamma)_{in}$	$(n, \gamma)_{res}$	Abundance, %	Gamma-ray energy, keV	N_γ [12]
Zn-64		48.6	< 1ns	
	Zn-65		0.0539 [1.6 μs]	78
Zn-66		27.9	< 1ns	
	Zn-67		0.093 [9.3 μs], 185 [1ns]	17
Zn-67		4.1	0.093 [9.3 μs], 185 [1ns]	
	Zn-68		< 1ns	175
Zn-68		18.7	< 1ns	
	Zn-69		< 1ns	33
Zn-70		0.62	< 1ns	
	Zn-71		< 1ns	79
W-182		26.5	100.0 [1.27ns], 1289 [1.04 ns], 1374 [2.2 ns], 1488 [1.7 ns], 1553 [1,3 ns], 2230 [1.4 μs]	
	W-183		453 [18.4 ns]	131
W-183		14.3	453 [18.4 ns]	
	W-184		111.2 [1.3 ns], 904 [1.1 ns], 1286 [8.3 μs], 1502 [2.3ns]	211
W-184		30.6	111.2 [1.3 ns], 904 [1.1 ns], 1286 [8.3 μs], 1502 [2.3ns]	
	W-185		244 [19.3 ns]	75
W-186		28.4	122 [1.1 ns]	
	W-187		< 1ns	225

3. MEASUREMENT RESULTS

Selected contributions of responses from reactions with fast neutrons (n, n′γ) in, total contribution of reactions $(n, n'\gamma)_{in} + (n, \gamma)_{res} + (n, n)_{res}$, and total contribution of reactions $(n, n'\gamma)_{in} + (n, \gamma)_{res} + (n, n)_{res} + (n, \gamma)_{cap}$ in the counting efficiency of registration ε_n of neutron, counts/neutron, by oxide single-crystal scintillators ZWO, CWO, BGO, GSO of fast neutrons from the ^{239}Pu Be source are presented in Fig. 10 and in Table 5.

The results of the response of these scintillators to gamma quanta from the ^{137}Cs source are also presented. The measurement modes are as follows: τ_f = 1 ns - single-electron mode, τ_f = 1 mks - spectrometric mode. To increase the contribution of radiation capture of neutrons flying out of the scintillator, a moderator with a thickness of d=1 cm was used, surrounding the scintillator from the outside and a tape Cd-converter.

In Table 5 shows the contribution of products of reactions with fast neutrons of the ^{239}Pu-Be source to the counting efficiency of registration ε of neutron by single-crystal oxide scintillators ZWO, CWO, GSO, BGO.

In Tab. 5 a), the contributions of the inelastic scattering reaction $(n, n'\gamma)_{in}$ are presented, the measurement mode is spectrometric, τ=1 ns +1 us.

Tab. 5 b) contains the total contribution of the reactions of the inelastic and resonance mechanisms $(n, n'\gamma)_{in}$, $(n, \gamma)_{res}$, τ = 1 ns, the measurement mode is single-electron. It is possible to note a significant increase in the efficiency of ZWO and CWO due to the contribution of the resonance capture reaction of neutrons by W tungsten nuclei.

The total contribution of reactions of inelastic scattering, resonance and radiation capture $(n, n'\gamma)_{in}$, $(n, \gamma)_{res}$, $(n, \gamma)_{cap}$ is presented in Tab. 5 c), the measurement mode is single-electron, τ = 1 ns. A further increase in the efficiency of ZWO and CWO can be explained by the use of a neutron moderator with a thickness of 1 cm, which has an entrance window, covered with a scintillator and surrounded by a cadmium converter tape. The low value of the efficiency of the GSO and BGO scintillators can be explained by the short scintillation time of the scintillators – τ = 600 ns (slow component) and τ = 300 ns, respectively.

Tab. 5 d). To evaluate the efficiency of gamma quanta registration by the atomic subsystem of the scintillator, the results of measurements with a Cs-137 source in the τ = 1 ns, single-electron mode are presented. It can be seen that the effectiveness of ZWO for fast neutrons is approximately 2.4 times higher than for gamma quanta.

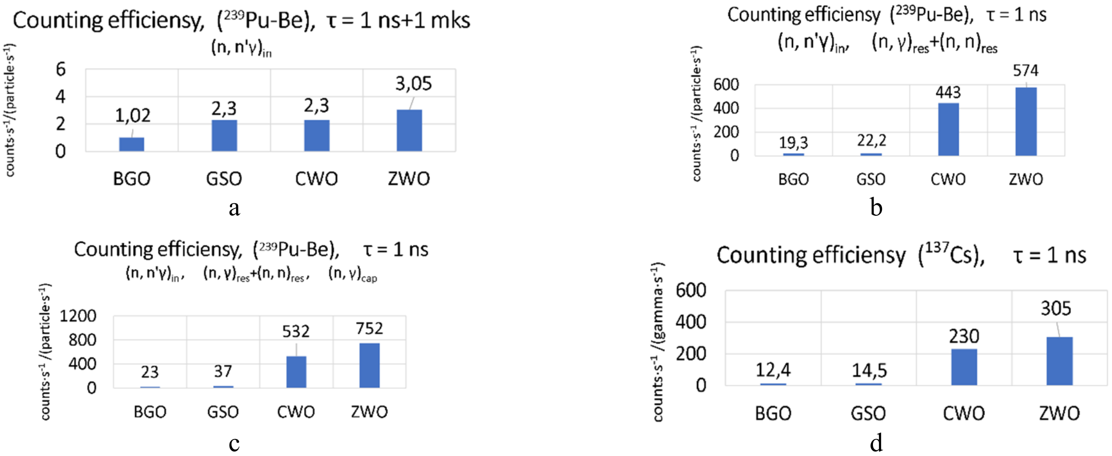


Figure 10. Contributions of reaction products with fast neutrons of the ^{239}PuBe source to the counting efficiency of registration ε_n, counts/neutron, oxide scintillators ZWO, CWO, GSO, BGO. Response of scintillators to gamma quanta of the ^{137}Cs source
a) – contribution of inelastic scattering, $(n, n'\gamma)_{in}$, measurement mode – τ_f =1 ns + 1 us;
b) – is the total contribution of inelastic scattering, resonance capture and scattering reactions $(n, n'\gamma)_{in} + (n, \gamma)_{res} + (n, n)_{res}$, τ_f = 1 ns;
c) – is the total contribution of inelastic scattering, resonance and radiation trapping $(n, n'\gamma)_{in} + (n, \gamma)_{res} + (n, n)_{res} + (n, \gamma)_{cap}$, τ_f = 1 ns – single photoelectron mode;
d) – is the contribution of the scintillator atomic subsystem, τ_f = 1 ns

Table 5. Values of contributions of products of reactions with fast neutrons of the ^{239}Pu-Be source to the counting efficiency of registration ε_n of neutron by oxide scintillators ZWO, CWO, GSO, BGO

Deposits of products in reactions	BiGeO ($\tau_{sc.}$=300 ns)			GdSiO (τ_{sc_slow}=0.6 us)			CdWO (τ_{sc}=18 us)			ZnWO (τ_{sc}=20 us)		
	Bi	Ge	O	Gd	Si	O	Cd	W	O	Zn	W	O
$\sigma(n,n'\gamma)_{in}$	0.392	0.612	0.309	1.487	0.524	0.309	0.366	0.485	0.309	0.491	0.485	0.309
$\sigma(n,n)_{res}$	144.48	120.08	46.13	170.61	25.38	46.13	85.13	1118.48	46.13	97.22	1118.48	46.13
$\sigma(n,\gamma)_{res}$	0.1919	5.935	0.00016	398.3	0.0834	0.00016	71.94	355.1	0.00016	2.495	355.1	0.00016
$\sigma(n,\gamma)_{cap}$	0.0342	2.229	0.00017	48699.8	0.1604	0.00017	2463.3	18.111	0.00017	1.079	18.111	0.00017
	a) $\varepsilon(n,n'\gamma)_{in}$, counts/neutron, τ=1 ns+1 us. V~ 1×1×1 cm											
	1.02			2.3			2.3			3.05		
	b) $\varepsilon((n,n'\gamma)_{in}+(n,n)_{res}+(n,\gamma)_{res})$, counts/neutron, τ=1 ns. V~ ø4×4 cm											
	19.3			22.2			443			574		
	c) $\varepsilon(n,n'\gamma)_{in}+(n,\gamma)_{res}+(n,n)_{res}+(n,\gamma)_{cap}$, counts/neutron, τ=1 ns. V~ ø4×4 cm											
	23			37			532			752		
	d) ε_γ, counts/gamma, Cs-137, τ=1 ns, V ~ ø4×4 cm											
	12.4			14.5			230			305		

3.1. A MODEL OF THE RESPONSE OF A SCINTILLATION DETECTOR TO FAST NEUTRONS

Based on the results of measurements of the counting efficiency of registration of fast neutron, the authors of the paper proposed a phenomenological model of the response of a fast neutron detector based on an oxide scintillator. The following values are used as parameters of the model: cross section of resonance capture, cross section of inelastic scattering, time of scintillation. In this case, the response of the detector can be presented in the form: $R \sim [\sigma_{in} + \sigma_{res}]\cdot\tau_{scint}$, where σ_{in} is the inelastic scattering cross section, σ_{res} is the resonant capture cross section, and τ_{scint} is the scintillation time. It should be noted that for BGO, due to the high sensitivity of the measuring path, correction for afterglow was required.

On Fig. 11 compares the model estimation of the counting efficiency of registration of fast neutrons by oxide scintillators with the experimental results obtained in this work. Calculated values of ZWO efficiency are compared to experimental values. The measurement errors are 5%. The value of the agreement criterion was $\chi^2 = 1.2$.

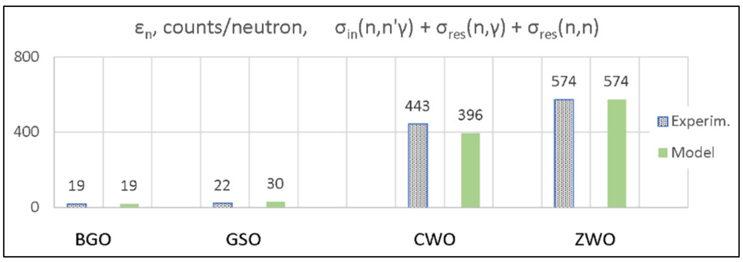


Figure 11. Comparison of the model estimation of the calculated efficiency ε of the registration of fast neutrons (solid columns) by oxide scintillators ZWO, CWO, BGO, GSO and the results of measurements (hatched columns, our experiment)

4. CONCLUSIONS

1. Experimental data on the counting efficiency of registration ε_n of fast neutron of Pu-Be sources with oxide scintillators CWO, ZWO, BGO, and GSO were obtained, which are consistent with the experimental data of other authors on the multiplicity of gamma quanta Nγ from inelastic scattering reactions (Nγ ~ 2-5 counts/neutron) and resonance capture (Nγ ~ 50-500 counts/neutron).

2. Measured contributions of reaction products of the interaction of fast neutrons from a ^{239}PuBe source with the substance of oxide scintillators to the counting efficiency of registration of fast neutron registration ε_n (counts/neutron). At the same time, the technique of efficiency measurements was used using a low-noise, high-sensitivity broadband pre-amplifier as part of a single-electron counter.

3. The following were used to select reaction deposits:

- the results of measuring the counting efficiency in the filtration mode τ ~ 1 ns + 1 us, which allows to distinguish the contribution to the efficiency of the reaction products of inelastic scattering $(n, n'\gamma)_{in}$;

- the results of measuring the counting efficiency of registration in the single-electron mode τ~1 ns, which allows to distinguish the contribution to the efficiency of the resonance capture reaction products $(n, \gamma)_{res}$ on heavy nuclei due to the registration of a significant number of low-energy gamma quanta from transitions between highly excited short-lived (τ ~ 1 ns) states of final nuclei.

- results of comparing the counting efficiency of registration of ZWO (ε ~ 574 counts/neutron, σ_{res}(W)~ 355 b,) and GSO (ε~22 counts/neutron, σ_{res}(Gd)~ 404 b). It is proved that, despite the close and very large values of the resonance capture cross sections for heavy W and Gd nuclei, the counting efficiency also depends on the scintillation time.

- the results of comparing the counting efficiency of BGO (ε ~ 19 counts/neutr., σ_{res}(Bi)~ 0.192 b,) and GSO (ε~22 counts/neutr., σ_{res}(Gd)~ 404 b). The low value of the efficiency for BGO can be explained by the small value of the capture cross section of the heavy Bi nucleus and the short time of scintillator exposure.

3. Registration of low-energy gamma quanta with energies up to Eγ≥0.1 keV required the development and application of a high-speed, highly sensitive path operating in the single-electron mode. This made it possible to separately register genetically related cascade events that occur in the detector, occurring per one input neutron, including secondary neutrons from the resonance scattering reaction, which are subsequently captured in the resonance energy region and increase the useful statistics of events.

4. The results of the measurements of the registration efficiency are consistent with the estimates of the thermodynamic model for medium (Zn) and heavy (W) nuclei for the densities of the ρ levels of final and compound nuclei. The high value of the ZWO efficiency can be explained by the significant value of the cross section of the resonant capture of W nuclei, the possibility of recording in the single-electron mode with maximum efficiency not only some gamma quanta consisting of a final nucleus with lifetimes in the interval from τ > 10 ns, but also shorter-lived cascades, τ ~ 1 ns of low-energy gamma-quanta of W nuclei emitted in highly excited transitions.

5. A phenomenological model of the response of oxide scintillators to fast neutrons is proposed, using the parameters of the inelastic scattering cross section, the resonance capture cross section, and scintillator exposure times τ. The model can be used for the development of new efficient fast neutron scintillators.

6. On the basis of the proposed method of increasing the counting efficiency of registration of fast neutron by the scintillator due to the use of cascade gamma quanta from the resonance capture reaction by heavy W-type nuclei and the developed measurement technique using the single-electron registration mode, a highly efficient fast neutron detector based on the ZWO oxide scintillator was created [3].

In the future, the proposed method will make it possible to create neutron and gamma-quantum detectors that are more compact than the existing ^{3}He counters for monitoring neutron and gamma-neutron fields of low intensity, for use in search gamma-neutron dosimeters and surveillance systems.

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ЛІЧИЛЬНА ЕФЕКТИВНІСТЬ РЕЄСТРАЦІЇ ВКЛАДІВ ПРОДУКТІВ РЕАКЦІЙ ШВИДКИХ НЕЙТРОНІВ СЦИНТИЛЯЦІЙНИМИ ОКСИДНИМИ ДЕТЕКТОРАМИ $ZnWO_4$, $Bi_4Ge_3O_{12}$, $CdWO_4$ та Gd_2SiO_5

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Представлені результати дослідження величини вкладів реакцій взаємодії швидких нейтронів джерел ^{239}Pu-Be та ^{252}Cf в лічильну ефективність реєстрації оксидними сцинтиляторами $CdWO_4$, $ZnWO_4$, $Bi_4Ge_3O_{12}$ і Gd_2SiO_5. Виміряно кількість гамма-квантів, що припадає на один вхідний нейтрон, випущених з кінцевих ядер, збуджених у реакції непружного розсіювання $(n, n'\gamma)_{in}$, резонансного розсіювання $(n, n)_{res}$ і захоплення $(n, \gamma)_{res}$ і радіаційного захоплення $(n, \gamma)_{cap}$. У якості фотоприймача використовується PMT R1307, що працює в одноелектронному режимі, фонове завантаження склало $n \sim 5*10^3$ c^{-1}. Виміряна лічильна ефективність ε для сцинтиляторів ø40x40 мм склала для ZWO – 752, для CWO – 532, для GSO – 37 і для BGO – 23 в одиницях «імпульс/нейтрон», похибка вимірювань ~ 3-5%. На формування відгуку детектора впливають такі параметри ядерного сцинтилятора, як величина взаємодії в резонансній області, щільність ядерних рівнів кінцевого ядра, час життя збуджених ядерних станів, верхня межа резонансної області перерізу, а також час висвічування та геометричні параметри сцинтилятора. Запропонована феноменологічна модель відгуку оксидного сцинтилятора до швидких нейтронів.
Ключові слова: *оксидний сцинтилятор; ZWO; BGO; CWO; GSO; швидкі нейтрони; ^{239}Pu-Be; резонансний захват; лічильна ефективність; густина ядерних рівнів; одноелектронний режим*