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THE THRESHOLD OF DETECTION OF FISSION MATERIALS BY ZnWO4 AND Bi4Ge3O12 SCINTILLATION DETECTORS

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In the present work we found the maximum discovery distance for ²³⁹Pu-Be source using the detectors based on ZWO (ZnWO4) and BGO (Bi₄Ge₃O₁₂) oxide scintillators. Detection distance was defined by using the radiation monitoring system "PORTAL". This research gives us data for estimation of the contribution of low-energy cascade gamma quanta CGQ. The CGQ emitted by excited scintillator nuclei defined the effective discovery distance of the fast neutrons source. The maximum detection distance was obtained with PMT in a single-photon counting mode. The maximum discovery distance for a BGO scintillator of size $Ø40 \times 40$ mm – 38 cm, ZWO scintillator of size $Ø52 \times 40$ mm – 54 cm, with reliability about 0.001. The results of the experiment on the ZWO scintillator can be explained by the registration of additional gamma quanta from the inelastic scattering reaction and the CGQ arising from resonant neutron capture region. This two mechanisms further lead to increase the sensitivity of the detector and increase the detection distance of the monitoring system. The key features of the monitoring system are: ZWO oxide scintillator, wide band measuring path, utilize PMT in single photon mode. The obtained detection distance was about 1.4 times higher in comparison with the spectrometric recording mode and 1.9 times higher in values of efficiency. Our results demonstrate the advantages of the ZWO scintillator compared to the BGO and demonstrate the possibility of using the resonant capture mechanism by ZWO detector nuclei to increase the fast neutrons sensitivity. The resonance capture mechanism increase sensitivity and maximum detection distance of the monitoring system. The low-energy gamma-quanta, which discharge of compound nuclei, are substantially suppressed in comparison with the classic spectrometric recording mode.

KEY WORDS: detector, fast neutrons, excited states, countable efficiency, density of nuclear levels

The compact gamma-neutron radiation detectors based on the oxide scintillators allow the creation of compact, highly sensitive systems for monitoring the unauthorized movement of fissile and radioactive materials. The response of detectors during neutron moderation in oxide scintillators is primarily formed by instantaneous gamma quanta of the inelastic scattering reaction and delayed cascade of gamma quanta from the radiation capture reaction in the resonance region emitted by excited states of the scintillators compound nuclei [1, 2]. Both of these reactions can be realized when neutrons are thermalized in some oxide scintillators of a few centimeters in thick.

Earlier [3-5], the signals of oxide scintillation detectors in order to suppress CGQ were amplified in the spectrometric mode, while the time of formation of the signal from the PMT in this mode was in the range 1 - 10 microseconds. Also, the physical efficiency (impulse×s⁻¹×cm⁻²/neutr⁻¹×cm⁻²) in spectrometric mode does not exceed 1. In [6,7] the first results of counting efficiency studies with ZWO scintillators and BGO. When using the mode of counting single photons in the ZWO scintillator, an increased (up to 60 pulse×s⁻¹×cm⁻²/neutron×s⁻¹×cm⁻²) was detected, compared to the BGO scintillator (2.5 pulse×s⁻¹×cm⁻²/neutron×s⁻¹×cm⁻²), which was explained by the registration of cascade quanta arising in the scintillator nuclei.

The aim of this work is a comparative assessment of the maximum neutron detection distance of a ²³⁹Pu-Be source by a monitoring system using ZWO, BGO detectors in two significantly different modes - spectrometric and single photon counting mode. It was the use of the single photon counting mode in the monitoring system with the highly sensitive wideband preamplifier that made it possible to estimate the contribution of low energy CGQ generated in the compound nuclei of the oxide scintillator to the maximum detection distance.

Since the energy of CGQ emitted by excited states of compound nuclei is small due to the high-level density, a preamplifier with a high gain and baseline low noise level was used. Due to the fact, that the CGQ emitted by the compound nuclei can be superimposed in the measuring path, it was possible to register them separately by using a broadband preamplifier with a differentiating delay line. To ensure the highest possible counting efficiency of cascade gamma-quanta, a single photon counting mode was applied in the PMT, which made it possible to isolate signals of extremely low energies and durations about nanoseconds.

Earlier [6] the obtained data indicated the absence of a noticeable generation of CGQ in the BGO scintillator with size of \emptyset 40×40 mm. The measurement results by using the monitoring system confirm the previous results, since the measured maximum detection distances for the case of counting single photons and the spectrometric mode for the BGO scintillator are practically the same.

RESEARCH & METHODS

The counting efficiency of neutron detection by oxide scintillators in units of impulse×s⁻¹×cm⁻²/neutron×s⁻¹×cm⁻² was previously estimated according to the procedure described in [6]. For effective coupling of scintillator with monitoring system in current work, we use parameter "maximum distance of discovery". This parameter is defined by monitoring system threshold and can be obtained by using equation

$$x_{tr} = x_{avg} + k \times (1/(n-1) \times \Sigma (x_i - x_{avg})^2),$$

where k = 3.5, x_{tr} – threshold level, x_{avg} – average value, x_i – enumeration of all values. The dispersion of the count rate was achieved by using standard deviation low for each data point. It was also found that the background fluctuation is not depend on Poisson distribution. The data reliability of the maximum detection distance was set at 0.001% (error rate), otherwise, no more than 1 false pulse per 1000 pulses. The sensitivity measurements of a radiation monitor system based on ZWO, BGO single crystal detectors under fast neutron irradiation were carried out in a spherical geometry [8]. We use 239 Pu-Be with flux 0.95×10⁵ neutron×s⁻¹, Ø20×30 mm and 52 grams. The source is placed inside a lead ball Ø100 mm with a well Ø20 mm. The lead ball simultaneously attenuates the accompanying gamma radiation from the 239 Pu-Be source [6, 7]. We add an additional lead shield – 5 mm to protect the detector from background gamma radiation. The principle diagram of the monitoring system Fig. 1.



Fig. 1. The principle diagram of the monitoring system

The minimum pulse width of the input signals is ~ 4 ns. The signal accumulation time was defined by software and can be set from 10 ms and more.

In experiment of determination the maximum discovery distance the pulse accumulation time was about 1000 seconds, number of iterations – 1000, time of one sample accumulation 1 sec. Data was obtained in two mode with neutron source and without. Counting rate in single photon counting mode for ZWO $052 \times 40 \text{ mm}^3$ was ~ 3000 sec⁻¹, in spectrometric mode ~ 40 sec⁻¹. Principle of data accumulation was shown on Fig. 2. Where red line is calculated threshold, data below the red line is background fluctuation without source and data above threshold from ²³⁵Pu-Be.



Fig. 2. The data accumulation by using portal software (source ²³⁹Pu-Be). Red line is threshold

Structure diagram of measurement setup contain PMT R1307 Hamamatsu, ultra-low noise amplifier and was discussed previously [6] in our works. The signals from PMT (baseline noise fluctuation + electronic noise ~ 10 mV) was amplified by 6x stage preamplifier (band ~ 200 M Γ u, 60 dBm, output current on 50 Ohm 80 mA). In addition, output signal was trimmed by shorted delay line of 2 m length. The PMT voltage in single photon mode was 1250 – 1350 V, in spectrometric mode – from 650 V to 850 V. Signal slew rate in spectrometric mode was 1 us, in "PORTAL" path ~ 2 ns.

RESULTS

In this work, we obtained the experimental values of the maximum detection distance "source-detector". The counting speed of the recorded signals did not exceed the predefined threshold $x_{tr} = x_{avg} + k \times ((n-1)^{-1} \times \Sigma (x_i - x_{avg})^2)$. The reliability of the measurements is about 1 false alarm per 1000 pulses for ZWO detectors and BGO. The measurements were carried out in two modes – in the spectrometric mode and the mode of counting single photons. The Table shows the results of measurements of the maximum detection distance R (cm) for ZWO, BGO scintillators in the monitor, K1 coefficients — increase in the maximum detection distance, K2 – increase in sensitivity in the monitoring system, K3 – increase in the effective area of the detector compared to the spectrometric mode.

The sensitivity increment and the effective area of the detector were estimated according to the law of inverse squares.

Discovery distance P (cm) for 7WO BGO

Table.

Discovery distance R (cm) for Zwo, BGO					
scintillator	$\begin{array}{c} \text{R, cm} \\ (\tau \sim 2 \text{ ns}) \end{array}$	R, cm $(\tau = 1 \text{ us})$	K1 (increase in detection	K2 (increase in	K3 (increase in detector
711/0		20	distance)	detection efficiency)	window)
ZWO	54	39	1.4	1.9	1.9
BGO	38	38	1	1	1

DISCUSSIONS AND CONCLUSIONS

The obtained values of the maximum detection distance of the "detector – radiation monitor" system during the registration of fast neutrons of a ²³⁹Pu-Be source for a ZWO single crystal detector can be explained as follows.

In the ZWO scintillator [6, 7] arise the additional CGQ which associated with the primary gamma quantum from the inelastic scattering reaction, which results in the case of their efficient isolation and registration (single photon counting mode) to increase the statistics of signals related to one input particle and, as a result, to increase the sensitivity of the system. At the same time, measurements carried out in the spectrometric mode on a ZWO scintillator did not give an increase in sensitivity, since CGQ (with low energies ~ 0.2 -1 keV) are suppressed in this mode. CGQ registration is require high amplifier gain (\sim 60 dBm) and single photon counting mode for PMT.

In the BGO scintillator of the indicated sizes [6, 7, 9, 10], CGQ are practically not observed, which is confirmed by the results of measuring the maximum detection distance of the monitor in different modes — photon counting and spectrometric. Thus, for BGO-type scintillators, an increase in statistics (photon counting mode) practically does not lead to an increase in sensitivity, since the conditions for the extraction of CGQ are not realized in BGO. It should be noted that in monitoring systems, in addition to signal statistics, the dispersion of the signal from neutrons plays an important role, which depends on the mechanism of neutron energy conversion in the scintillator, i.e. on the type of scintillator and reaction.

Thus, the use of the ZWO scintillator as part of a monitor recording signals in the photon-counting mode makes it possible efficiently using of a CGQ from the resonance capture of fast neutrons. That leads to increase the sensitivity of the monitoring system compared to the spectrometric mode by about 1.9 times and increasing the maximum detection distance by about 1.4 times.

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ПОРІГ ВИЯВЛЕННЯ МАТЕРІАЛІВ ПОДІЛУ СЦИНТИЛЯЦІЙНИМИ ДЕТЕКТОРАМИ ZnWO4 ТА Ві4Ge3O12

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²Інститут сцинтиляційних матеріалів, НТЦ "Інститут монокристалів", НАН України, 61001, Харків, Україна У даній роботі виміряна максимальна відстань виявлення ²³⁹Ри-Ве джерела швидких нейтронів детекторами на основі оксидних сцинтиляторів ZWO (ZnWO4) і BGO (Bi4Ge3O12). Виміри відстані виявлення проводилися з використанням розробленої системи радіаційного моніторингу. Це дозволило оцінити величину вкладу низькоенергетичних каскадних гамма-квантів (КГК), що випускаються збудженими ядрами сцинтилятора в відстань виявлення швидких нейтронів. Значення максимальної відстані виявлення детекторів отримані в режимі рахунку одиничних фотонів і склали 38 см для сцинтилятора ВGO розміром Ø40×40 мм, для сцинтилятора ZWO розміром Ø52×40 мм – 54 см з надійністю не гірше 0.001. Результати експерименту для сцинтилятора ZWO можуть бути пояснені реєстрацією, крім гамма-квантів з реакції непружного розсіювання, також КГК, що виникають при резонансному захопленні нейтронів, що призводить до зростання чутливості детектора і збільшенню відстані виявлення системи моніторингу. Низькоенергетичні КГК реєструвалися в режимі рахунку одиничних фотонів. Отже, застосування в системі моніторингу оксидного сцинтилятора ZWO, в складі якого містяться ядра, що випускають КГК з реакцій резонансного захоплення, і широкосмугового вимірювального тракту, що працює в режимі рахунку одиничних фотонів дозволяє збільшити ефективну чутливість детектора приблизно в 1.9 рази, що призводить до збільшення відстані виявлення джерела нейтронів детектором ZWO приблизно в 1.4 рази в порівнянні зі спектральним режимом реєстрації. Отримані результати демонструють переваги сцинтилятора ZWO в порівнянні з BGO і вказують на можливість використання механізму резонансного захоплення нейтронів ядрами детектора ZWO для збільшення чутливості детектора до швидких нейтронах. Використання механізму резонансного захоплення призводить до підвищення чутливості і збільшення максимальної відстані виявлення системи моніторингу в порівнянні зі спектрометричним режимом реєстрації, в якому низькоенергетичні гамма-кванти розрядки компаунд ядер, що утворюються в результаті резонансного захоплення, істотно пригнічені.

КЛЮЧОВІ СЛОВА: детектор, швидкі нейтрони, збуджені стани, лічильна ефективність, щільність ядерних рівнів

ПОРОГ ОБНАРУЖЕНИЯ МАТЕРИАЛОВ ДЕЛЕНИЯ СЦИНТИЛЛЯЦИОННЫМИ ДЕТЕКТОРАМИ ZnWO4 И Bi4Ge3O12

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В настоящей работе измерено максимальное расстояние обнаружения ²³⁹Ри-Ве источника быстрых нейтронов детекторами на основе оксидных сцинтилляторов ZWO (ZnWO4) и BGO (Bi4Ge3O12). Измерение расстояния обнаружения проводились с использованием разработанной системы радиационного мониторинга. Это позволило оценить величину вклада низкоэнергетичных каскалных гамма-квантов (КГК), испускаемых возбужленными ядрами спинтиллятора в расстояние обнаружения быстрых нейтронов. Значения максимального расстояния обнаружения детекторов получены в режиме счета единичных фотонов и составили 38 см для сцинтиллятора ВGO размером Ø40×40 мм, для сцинтиллятора ZWO размером Ø52×40 мм – 54 см с надежностью не хуже 0.001. Результаты эксперимента на сцинтилляторе ZWO могут быть объяснены регистрацией, кроме гамма-квантов из реакции неупругого рассеяния, также КГК, возникающих при резонансном захвате нейтронов, что приводит к росту чувствительности детектора и увеличению расстояния обнаружения системы мониторинга. Низкоэнергетичные КГК регистрировались в режиме счета единичных фотонов. Таким образом, применение в системе мониторинга оксидного сцинтиллятора ZWO, в составе которого содержатся ядра, испускающие КГК из реакций резонансного захвата, и широкополосного измерительного тракта, работающего в режиме счета единичных фотонов позволяет увеличить эффективную чувствительность детектора примерно в 1.9 раза, что приводит к увеличению расстояния обнаружения источника нейтронов детектором ZWO примерно в 1.4 раза по сравнению со спектрометрическим режимом регистрации. Полученные результаты демонстрируют преимущества сцинтиллятора ZWO по сравнению с BGO и указывают на возможность использования механизма резонансного захвата нейтронов ядрами детектора ZWO для увеличения чувствительности детектора к быстрым нейтронам. Использование механизма резонансного захвата приводит к повышению чувствительности и увеличению максимального расстояния обнаружения системы мониторинга по сравнению со спектрометрическим режимом регистрации, в котором низкоэнергетичные гамма-кванты разрядки компаунд ядер, образующихся в результате резонансного захвата, существенно подавлены.

КЛЮЧЕВЫЕ СЛОВА: детектор, быстрые нейтроны, возбужденные состояния, счетная эффективность, плотность ядерных уровней