ABSTRACT

Yakymenko I. I. The research of the mechanisms of interaction of fast neutrons with the substance of single-crystal and composite oxide scintillators. – Qualification scholarly paper: a manuscript.

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The dissertation is devoted to studies of the mechanisms of interaction of fast neutrons with the material of single-crystal and composite oxide scintillators.

The creation of compact high-sensitivity detectors for neutron and gamma-neutron radiation control systems to combat the illegal transportation of radioactive materials is an urgent task. The most common are inspection systems based on ^{3}He-neutron counters and have a low registration efficiency of ~ 10%, due to the need to moderate fast neutrons to thermal energy. The scarcity and high cost of manufacturing ^{3}He stimulates the search for new detectors and new principles for recording fast neutrons. Previous studies have shown that the mechanism of inelastic scattering $(n, n' \gamma)_{in}$ can be used to detect fast neutrons with detectors based on heavy oxide scintillators. The recording efficiency was ~ 0.5 for detectors of small size (~ 10 mm^3). In this case, the signals were registered, the duration of which was formed (constant of the integration time) was in the microsecond range. This was due to the need to reduce the effect of secondary cascade gamma quanta arising in the scintillator substance. The registration of response pulses in the microsecond range allowed to register only high-energy (> 20-30 keV) gamma quanta from the inelastic scattering reaction of fast neutrons (n, n' γ) arising from the discharge of excited single-particle and collective states of medium and heavy scintillator nuclei. Therefore, we proposed to use the gamma-ray cascades generated not only in the

inelastic scattering reaction, but also in the resonant and radiation capture reactions to increase the sensitivity of the fast neutron detector. Fast neutrons ^{239}Pu-Be sources with maximum energy $E \leq 10$ MeV in the process of scattering and deceleration in the substance of the oxide scintillator with linear dimensions of ~ 40-50 mm and more in the reactions of inelastic and resonant scattering, radiation capture pass three energy regions: the region of inelastic scattering in the reaction $(n, n' \gamma)_{in}$ (~ 10 MeV - 100 keV), the region resonant capture $(n, n' \gamma)_{res}$ (100 keV - 100 eV) and the radiation capture region (100 eV - 0.025 keV). In these reactions, the states of compound nuclei (A + 1) are excited with a lifetime of ~ 10^{-14} s - 10^{-12} s, and the states of finite nuclei (A) with lifetimes from picoseconds to tens of microseconds, and delayed gamma can be born -quants of γ_{del} caused by wanderings of secondary neutrons from reactions $(n, n' \gamma)_{in}$ and $(n, n' \gamma)_{res}$ in the scintillator substance. Thus, the response of the detector to one input particle (ie fast neutron) is a mixture of gamma quanta and intermediate neutrons, so the number of registered secondary particles (ie gamma quanta) by the detector may significantly exceed 1. Since the nuclei that are part of the oxide scintillators (W, Gd, Zn) have significant values of the cross-section of the interaction in the resonant region, ~ 50 - 500 bar, while the cross-sectional values in the inelastic region are units of bar (~ 2 - 3 bar) , the registration of gamma quanta associated with these processes can significantly increase the statistics of events per input neutron and, as a consequence, increase the efficiency of neutron registration. Such processes have a small discharge energy in the range from eV units to hundreds of keV and lifetimes in the range $\tau \sim 10^{-14}$ s - 10^{-5} s.

Thus, in the inelastic scattering reaction $(n, n' \gamma)_{in}$, the neutron emitted from the nucleus has a significantly lower energy compared to the initial energy, which significantly increases in the future the probability of the resonant capture reaction $(n, n' \gamma)_{res}$ in the region $\Delta E \sim 1\text{-}100$ keV and the re-formation of a new compound core (A + 1)*. Note that the re-formation of the compound nucleus occurs at times of the order of 0.1 - 10 μs and more for oxide scintillators 4-5 cm thick and due to the time delay of the neutron that slows down on the scintillator nuclei.

The generation of high-energy gamma quanta from the reaction $(n, n'\gamma)_{in}$ is carried out mainly by the final nucleus (A*). The intermediate compound nucleus (A + 1), being highly excited and having a high density of levels, emits mainly rather low-energy photons, the energy of which will lie in the noise region. However, [2] a significant yield of instantaneous cascade gamma quanta of average energies per neutron (~ 50 - 80 and more) of discharge of excited states of compound nuclei (A + 1) formed in reactions $(n, n'\gamma)_{res}$ and $(n, n'\gamma)_{cap}$. The generation of high-energy gamma quanta in the reaction $(n, n'\gamma)_{in}$ is carried out by a finite nucleus (A *). The intermediate compound nucleus (A + 1) receives the average excitation energy ~ $S_n + E_{n\_kin}$, and, after the departure of the neutron, which reduces the energy of the nucleus by the value of S_n, is converted into a final nucleus with excitation energy equal to the kinetic energy of the incident neutron. The final nucleus emits, for the most part, fairly high-energy photons, which are confidently recorded by the path. Instantaneous cascade gamma quanta of very low energies can also be observed experimentally during the discharge of excited states of compound nuclei (A + 1) formed in the reaction $(n, n'\gamma)_{in}$. The energy of these gamma quanta is close to the noise threshold of the path, their number is very small and can be registered only at very low levels of excitation of the final nucleus, when the density of nuclear levels becomes low (increased excitation energy).

In the case of neutron capture in the resonant region in the reaction $(n, n'\gamma)_{res}$, highly excited states (6 - 8 MeV) of compound nuclei (A + 1) emit cascades of instantaneous gamma quanta of very low energies, because at high excitation levels the density of levels compound nuclei are large. Although the excitation energy of the final nucleus during resonant capture is 10-100 keV and less, the density of nuclear levels is significantly lower than for high-energy excitations, the distance between the levels in this case can be ~ 1 - 10 keV or more. Therefore, in the reaction (n, n' γ)$_{res}$ we should expect the manifestation of low-energy gamma quanta of the discharge states of the finite nuclei, which can be registered only in the presence of sensitive appropriate equipment.

The analysis of neutron nuclear reactions on the basis of a thermodynamic model allowed us to estimate the average excitation energy of comparable and finite nuclei that are part of oxide scintillators, to specify the energy of secondary neutrons. The density of levels and distances between levels at low excitation energies of finite nuclei were also estimated. These estimates confirm the experimental results that indicate the existence of low-energy gamma quanta (~ 0.1 - 30 keV) that occur in the transitions of light compound nuclei and finite nuclei. In addition, based on the analysis of experimental data from the reactions (n, n' γ)$_{in}$, (n, n' γ)$_{res}$ and (n, γ)$_{cap}$. On the nuclei of the scintillators, the amounts of yield of secondary gamma quanta produced in these reactions were specified in response to one primary input neutron.

Thus, the inelastic scattering reaction is the starting point, which starts the cascade process of formation and decay of excited states of nuclei in the studied crystals. In the process of deceleration of the released inelastic neutron from the reaction (n, n' γ), in a fairly long oxide scintillator, in addition to instantaneous gamma quanta from the composite nucleus (A + 1, Z), there are delayed gamma quanta from the daughter nucleus (A, Z) from the reaction (n, n' γ) (for 1 input neutron - several hundred and more gamma quanta) with energy from units of MeV to ten keV, respectively. In fact, a chain of time-genetically related processes generates excited nuclear states, and the delay in the occurrence of secondary gamma quanta is determined both by the time of secondary neutrons passing through the scintillator and the time of existence of the excited nuclear states themselves. Delaying the response of the detector from wandering in the crystal of secondary neutrons and neutrons from secondary reactions of resonant capture can contribute to a quantitative increase in the number of signals recorded by the detector per input neutron.

In comparison with gamma quanta produced by inelastic scattering from the reaction $(n, n' \gamma)_{in}$ cascade gamma quanta from the reaction of resonant scattering $(n, n' \gamma)_{res}$ and radiation $(n, \gamma)_{cap}$ capture have an energy of up to ten keV (~ 0.1 - 30 keV), so their registration requires appropriate experimental equipment (especially preamplifier). These gamma quanta form genetically linked signal chains that can increase the statistics of events generated by the primary fast neutron in the scintillator. Practically the times of existence of excited states are in the range from units of nanoseconds to tens of microseconds. In the presence of a suitable low-noise broadband path with a high gain, which allows you to effectively record both low-energy gamma quanta for the discharge of nuclear states with short lifetimes and gamma quanta born in retarding neutron capture, you should expect an increasing the counting efficiency, accordingly, the sensitivity of the neutron detector.

Objectives and purpose of the work were defined as: development of a scintillator response model for search and fabrication of new fast neutron detectors, study of the relationship of fast neutron interaction mechanisms with scintillator nuclei to identify genetic links of reaction products to improve registration efficiency, identify effective pathways reduction of fast neutron energy in the detector, development of accompanying electronics of the measuring path in order to register low-energy gamma quanta from transitions excited in fast neutron interaction reactions, detection of mechanisms of gamma quanta cascade generation in reactions $(n, n' \gamma)_{in}$, $(n, n' \gamma)_{res}$, $(n, \gamma)_{res}$ and secondary neutrons.

Among the research methods used, the following approaches should be emphasized: measuring the efficiency of scintillation detectors using different types of sources (^{252}Cf, ^{239}Pu-Be, ^{137}Cs); numerical calculations of detector responses based on the developed model; variation in both the isotopic composition of the scintillator and the volume of the detector; application of time filtering of the pulse response of the detector (7 ns - 1 μs); the use of a radiation monitor to experimentally determine the threshold for detecting fast neutron sources online.

The novelty of the work lies in the following provisions: the genetic relationship of the mechanisms of interaction of fast neutrons with the nuclei of scintillators with the hardware response of the scintillator; the most productive mechanisms of fast neutron energy loss in the substance of scintillators and ways of generating cascade gamma quanta are revealed; the connection of mechanisms of gamma-ray cascade generation in reactions $(n, n' \gamma)_{in}$, $(n, n' \gamma)_{res}$, $(n, \gamma)_{res}$ and secondary neutrons, compound nuclei and finite nuclei is revealed; the genetic connection of cascades of products of inelastic, resonant scattering and fast neutron capture reactions on the nuclei of the ZWO ($ZnWO_4$) scintillator was used for the first time, confirmed by the patent of Ukraine; for the first time a new high-efficiency fast neutron detector ZWO was created using three mechanisms $(n, n' \gamma)_{in}$, $(n, n' \gamma)_{res}$, $(n, \gamma)_{res}$. for the first time a new KDP (K_2PO_4) detector with high selectivity for fast neutrons in comparison with gamma quanta was created; developed and created a new high-speed broadband preamplifier for recording the response of the scintillator and allocating the contributions of the interaction mechanisms in single-photon registration mode; developed and created a high-speed radiation monitor to detect in a continuous mode the threshold of detection of a source of fast neutrons for the studied scintillator.

Summarizing the results of measurements of counting efficiency in units of imp. * s^{-1} / neutron * s^{-1} taking into account the three mechanisms $(n, n' \gamma)_{in}$, $(n, \gamma)_{res}$ + $(n, n' \gamma)_{res}$ and $(n, \gamma)_{cap}$ was 752 for ZWO, 532 for CWO ($CdWO_4$), 37 for GSO (Gd2SiO5) and 23 for BGO ($Bi_4Ge_3O_{12}$), measurement error is 3 - 5 %. The Hamamatsu R1307 photodetector used in experiments was operating in a single-photon mode (voltage 1250 V) and a low-noise broadband amplifier with a gain of ~ 70 dB and a band-width speed of ~ 300 MHz was created. The formation of the detector signal is influenced by such parameters of nuclei and atoms that are part of the scintillators as the cross-sectional sizes of inelastic and resonant scattering, the density of nuclear levels of constituent and terminal nuclei in the energy interval, the width of the resonant region.

Use of one-photon technique of registration of gamma quanta from excited in reactions $(n, n' \gamma)_{in}$ one-particle states, low-energy gamma quanta from reaction $(n, \gamma)_{res}$ + $(n, n' \gamma)_{res}$ and reaction $(n, \gamma)_{cap}$ significantly increases the number of genetically related events in the detector per incident neutron and, as a consequence, increases the computational efficiency and sensitivity to neutron detection. Experimental results are fully consistent with the proposed phenomenological model of the response of the oxide scintillator to fast neutrons.

The results of the work will be used in the development of new fast neutron radiation detectors. The basis for the study of the mechanisms of interaction of fast neutrons with the substance of single-crystal and composite oxide scintillators has been created. Developed software and hardware complex for estimating the distance of detection of radiation sources with a given reliability type "Portal", working on the developed method of registration of fast neutrons is the basis for experimental development of monitoring systems for checkpoints, radiation control monitors, quality control of detectors.

Keywords: fast neutron detector, ZWO, BGO, CWO, GSO, KDP, mechanisms of fast neutron interaction, detector registration efficiency, radiation monitor, fast preamplifier, nuclear levels, scintillator response model, photon counting mode, excited levels, gamma multiplicity.