SIMULATION OF CHARACTERISTICS OF GAMMA-RADIATION DETECTORS BASED ON MERCURY COMPOUNDS

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We used Monte-Carlo method to compare characteristics of HgI_2- and HgS-detectors which work in the mode of pulse amplitude analysis. The GEANT4 Simulation Toolkit version 4.9.4p04 and EGSnrc version 4.r2.3.2 were used for modeling gamma-quantum trajectories and calculating response functions in the gamma-quantum energy range between 25 keV and 3 MeV. We supposed that simulated HgI_2- and HgS-detectors were equipped with planar contacts and their geometric sizes were 3.14 $mm^2 \times 1$ mm. Basing on calculated detector's response functions, we obtained the efficiency of gamma-quantum registration and detector sensitivity dependence on energy for energy range from 0.025 to 3 MeV. Statistical characteristics of theoretical response functions and amplitude distributions of pulses are studied taking into account noise and incomplete charge collection distortions of theoretical response functions. Results of simulation show that in the high-energy region the efficiency of energy conversion of gamma-quanta in charge signal for HgS detectors can exceed the same characteristic for HgI_2-detectors up to 10…12 percents.

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1. INTRODUCTION

Mercuric iodide (HgI_2) is a well-examined material for registering gamma-radiation at room temperatures. Theoretical detection capability of HgI_2 exceeds the characteristics of most materials produced for gamma-radiation spectroscopy [1]. Present technologies of HgI_2 growth allow to obtain specific resistance ρ of this semiconductor compound in the range between 10^{13} Ohm×cm and 10^{14} Ohm×cm [2]. Using the techniques of single-carrier collection (for wide band-gap semiconductor detectors it is only electron collection) [3], the resolution of HgI_2-detectors can exceed the resolution of the same CdZnTe-detectors.

However, detectors based on HgI_2 did not become commercially available because crystals from this semiconductor compound have essential defects. In particular, the higher sublimation rate of iodine leads to the sample surface enrichment with iodine (stoichiometric impurity), and the transition from tetragonal α-HgI_2 phase to the orthorhombic β-HgI_2 phase destroys order of the material structure [4]. As a result, grown material is an excessively soft. Furthermore, it easy delaminates along iodine planes.

Although other wide band-gap semiconductor compounds which include iodine (for example, PbI_2) do not undergo phase changes, but they are also not suitable for mechanical treatment (cutting the ingot, polishing the detector's surface) because of high fragility of the grown crystals [1].

The disturbance of the crystalline structure leads to formation of the electrically active defects in HgI_2 which essentially deteriorate charge collection efficiency (CCE) in the planar detectors [4, 5]. Therefore, in the majority of published investigations the thickness of the HgI_2-detector does not exceed 1 mm. Such detectors can be used for registering X-rays and low-energy gamma-quanta (E_γ is below 100 keV). Since ratio of the lateral side value (or diameter for cylindrical detectors) to the thickness of HgI_2-detector is usually not less than 10 to 1, that the value of anisotropy of detector's sensitivity is specified as tens or even hundreds percents (at 90° angle). Field of application of anisotropic detectors is mainly confined to stationary devices wherein detector orientation toward axis of gamma-radiation beam is not changed.

Mentioned disadvantages of iodine-containing semiconductor compound cause the search of other materials which have the same detection characteristics as HgI_2. Moreover, new materials must be more suitable for mechanical treatment and they must provide the reproduction of good detection characteristics of detector. Mercuric sulphide (HgS) is one of the new promising materials for detecting gamma-radiation [6]. It is the most widely distributed stable mercuric mineral (cinnabar). As shown in Ref. [6], even impure natural HgS-crystals have sufficiently

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good characteristics of charge carrier transport for creation of gamma-radiation detectors which could work in the energy range up to 100 keV.

In this work, Monte-Carlo method is used for comparison between detection characteristics of planar HgS gamma-radiation detectors working in the mode of registration of single gamma-quanta [7, p. 110] and characteristics of the same HgI_2-detectors. Statistical characteristics of theoretical response functions and amplitude distributions of pulses have been investigated taking into account the distortions of theoretical response functions due to noises and incomplete charge collection. Basing on the calculated detector's response functions we obtained the dependencies of the efficiency of gamma-quantum registration, $\epsilon(E_\gamma)$, and detector's sensitivity $\delta(E_\gamma)$ on energy E_γ for energy range from 0.025 to 3 MeV.

2. SIMULATION OF RESPONSE FUNCTION OF ROOM-TEMPERATURE SEMICONDUCTOR DETECTORS

The computational model of response functions of room-temperature semiconductor detector (SCD) was originally developed and tested for CdTe- and CdZnTe-detectors with planar contacts [8]. Also, this model has been applied for analysis of characteristics of other wide band-gap semiconductor materials [9]. For computations, we used the universal EGSnrc and Geant4 codes for the Monte-Carlo simulating the passage of photons, electrons and positrons through matter [10, 11]. Besides of interaction of gamma-quanta and charged particles with detector matter the model [8] allows us to take into account the influence of noises and losses of non-equilibrium charge on the amplitude of the output signal of detector.

Fig. 1 and Fig. 2 show the calculated response function of the HgI_2-detector with planar contacts on gamma-quanta with energy $E_\gamma = 662$ keV (^{137}Cs source) and its transformation in the measuring channel. The plot ordinate is defined as the ratio of pulse counts N_i in i–th channel of the simulated analog-digital converter (ADC) to total pulse counts $N_{tot} = \sum N_i$. Detector parameters correspond to data Ref. [12]: the sizes of 1×1×1 cm^3, the charge transport characteristics (mobility-lifetime product) – $(\mu\tau)_e = 5\times10^{-3}$ cm^2/V, $(\mu\tau)_h = 3\times10^{-5}$ cm^2/V, the SCD's bias U = 2.5 kV. Fig. 1 demonstrates the processes of absorption of gamma-quantum energy and production of non-equilibrium charge in the detector. The investigated detector has sufficiently high probability of the complete absorption (photo-effect) of gamma-quanta with energy $E_\gamma = 662$ keV. The simulation shows that about fifty percents of the interacted quanta are fully absorbed. The efficiency of gamma-ray registration for energy $E_\gamma = 662$ keV is about 42% and so absolute theoretical intrinsic probability of scattering in the photopeak is about 21%. Therefore, less than 25% of input gamma-quantum flow is fully absorbed in HgI_2 at the length 1 cm. It coincides with results of simulations by MCNP code [1]. Escape peaks corresponded to characteristic gamma-quanta of the mercury and iodine which have left the detector volume are clearly visible on the Compton valley background ($0.45 < E < 0.65$ MeV). The ratio of the photopeak amplitude E_γ to the average-pulse amplitude in the Compton continuum region ($0 < E < 0.45$ MeV) exceeds two orders of magnitude. Small peak spreading comes from fast electron energy losses due to the generation of lattice vibrations (losses up to 5%).

Fig. 2 shows the changes of response function of HgI_2-detector (Fig. 1) after the output from the measuring channel. A main part of pulse amplitudes is shifted to the Compton continuum region. The escape peaks are almost completely spreaded. The ratio of the photopeak amplitude E_γ to the average amplitude in the Compton continuum region (i.e. where $0 < E < 0.45$ MeV) is less than 1.3. Overall, the response function (see Fig. 2) is in satisfactory agreement with the calculation in Ref. [12].

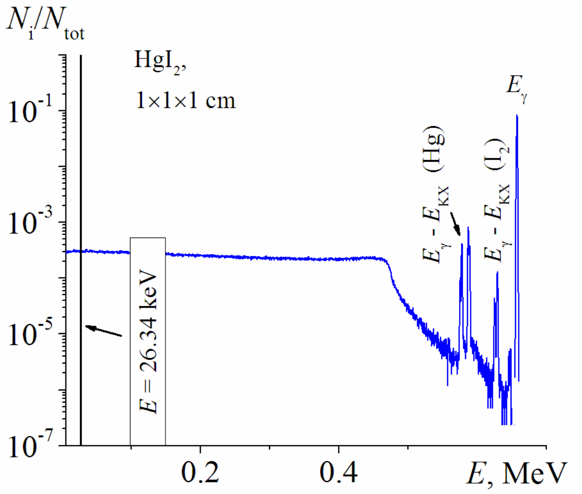

Fig. 1. *The response function of HgI_2-detector on gamma-quanta with 662-keV energy without taking into consideration the charge collection and noises*

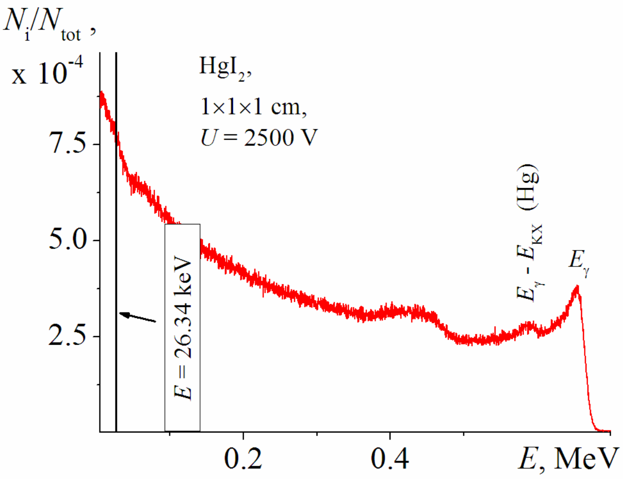

Fig. 2. *The response of HgI_2-detector after the output of the measuring channel corresponded to Ref. [12]*

The results of simulation (see Fig. 2) are satisfactorily conformed to calculations presented in the Ref. [12] but the shape of the experimental ^{137}Cs spectrum from this Ref. differs from the shape of the calculated spectrum. The most important distinctions are the photopeak lack and higher value of experimental ratio N_i/N_{tot} at the initial energy range compared with simulation. The variations in the model's parameters showed that we can achieve such spectral distribution if the intensity of internal electric field inside the detector is much less than expected value, that is $U/d = 2500$ V/cm for planar detectors. It may result from the distortion of the internal electric fields near numerous growth defects of semiconductors as in Ref. [13, 14]. This case can be reproduced via the used model at lower bias U (Fig. 3). From Fig. 3, it is evident that the photopeak disappears as well as it occurs for the real spectrum of a ^{137}Cs source presented in Ref. [12]. The pulse-number in the initial channels exceeds the average value of pulse-number in the Compton continuum up to one order of magnitude.

Simulation shows the change in $CCE(x)$ corresponding to reduction of the intensity of internal electric field inside the detector. At the same time the detector thickness-averaged collection efficiency CCE is decreased from 57.6% to 42.9%. It leads to complete degradation of the 662-keV photopeak (see Fig. 3).

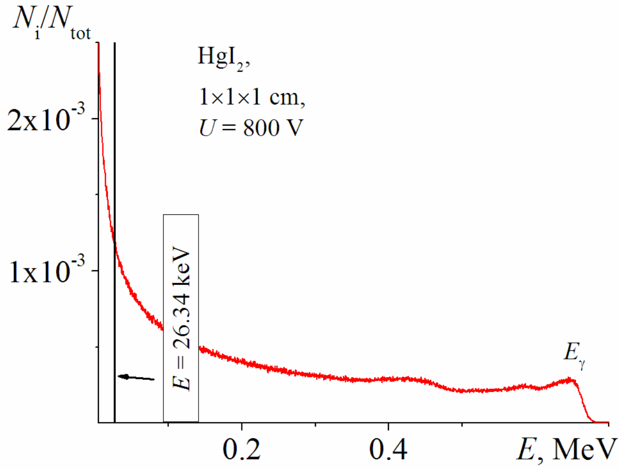

Fig. 3. *The change in the HgI_2-detector's response at the bias reduction*

Thus, the developed model of room-temperature semiconductor gamma-radiation detector allows us to obtain the response function of HgI_2-detector which is in satisfactory agreement with known experimental- and simulation-data. Consequently, we can use the described model for comparison between detection characteristics of HgI_2- and HgS-detectors.

3. CHARACTERISTICS OF HgI_2- AND HgS-DETECTORS

We calculated the response functions for the planar ø2 mm × 1 mm HgI_2- and HgS-detectors with ohmic contacts. Also, the spectrometric mode of detector operation was simulated. In the both cases, the bias voltage was 100 V. The equivalent noise charge was 400 e$^-$ (electron charge units). Mobility-lifetime products were chosen $(\mu\tau)_e = 1{\times}10^{-4}$ cm^2/V for electrons and $(\mu\tau)_h = 4{\times}10^{-5}$ cm^2/V for holes in both detectors. At the first stage, we carried out HgS-detector simulation with electron and holes mobilities $\mu_e = 15$ cm^2/(V×s) and $\mu_h = 2$ cm^2/(V×s) respectively (detector no. 1). HgI_2-detector mobilities were $\mu_e = 67$ cm^2/(V×s) and $\mu_h = 8$ cm^2/(V×s). At the second stage, we explored HgS-detector simulation with the same electron and hole mobilities as for HgI_2-detector (detector no. 2).

As the calculation shows (Fig. 4), the sensitivity δ of HgS-detector no. 1 is essentially lower than the sensitivity of HgI_2-detector. The detector thickness-averaged charge collection efficiency CCE for HgS-detector no. 1 was changed in the range from about 42% for 26.34-keV energy of gamma-quanta to about 4% for 3 MeV. At the same time for HgI_2-detector CCE change was observed in the range between about 65% and about 36%. We suppose that the main reason of comparatively low sensitivity of HgS-detectors is a ballistic deficiency due to low mobility of charge carriers [15].

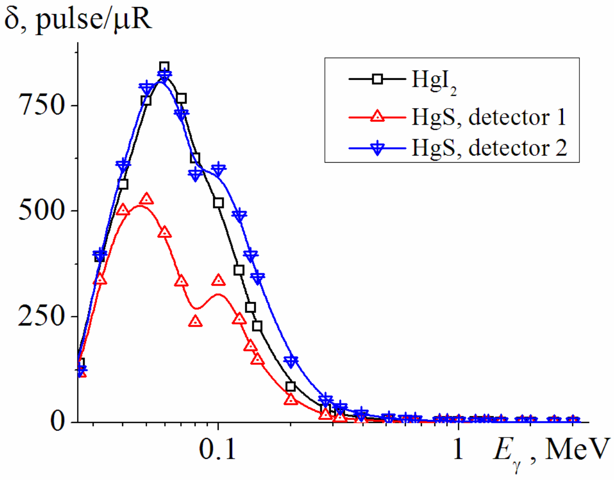

Fig. 4. *The dependence of sensitivity δ versus gamma-quantum energy for HgI_2- and HgS-detectors*

In order to check the last assumption we computed the theoretical response of HgS-detector no. 2 with the same mobility of charge carriers as for HgI_2-detector. As follows from data of Fig. 4, the sensitivity δ of HgI_2-detector is practically equal to sensitivity δ of HgS-detector no. 2 in the range of gamma-quantum energy up to about 80 keV. Sharp change in the sensitivity of HgS-detector no. 2 is observed in the energy region above 80 keV. This jump corresponds to transition over K-edge of absorption of mercury (83.1 keV). Interestingly, that this jump is almost invisible for HgI_2 (Fig. 5 and Fig. 6). The sharp changes in the sensitivity have to be taken into account at calculating the parameters of correction of energy dependence of sensitivity of HgS-detectors in low-energy dosimetry applications.

Fig. 6 demonstrates that the value of jump of mass energy coefficient in the HgI_2-detectors for about 83.5-keV energy compared with the same jump in the HgS-detectors is lower in 2 times. Taking into consideration the distinctions in the specific densities of HgI_2 and HgS, the changes in the probability of interaction of gammas with detector's material in the energy range of 82...85 keV for HgS is higher almost in 3 times than for HgI_2. It leads to more visible variations in the sensitivity of HgS-detectors compared with HgI_2-detectors in the range of about 83.5-keV energy.

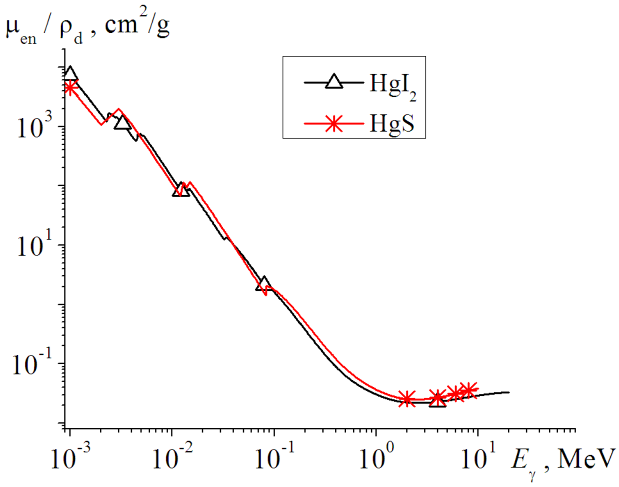

Fig.5. *The energy dependence of mass energy coefficient in the HgI_2- and HgS-detectors*

The comparison between dependencies of investigated detector's sensitivity on energy in the range above 300 keV can be most easily made for inverse function $1/\delta = f(E_\gamma)$ (Fig. 7). The calculation data show (plot – HgS, detector no. 1) that HgS-detectors which can be made from currently available material can not compete with detectors based on mercuric iodide. In the high-energy region the sensitivity of HgS gamma-radiation detectors compared with the same HgI_2-detectors is lower in 2 or more times.

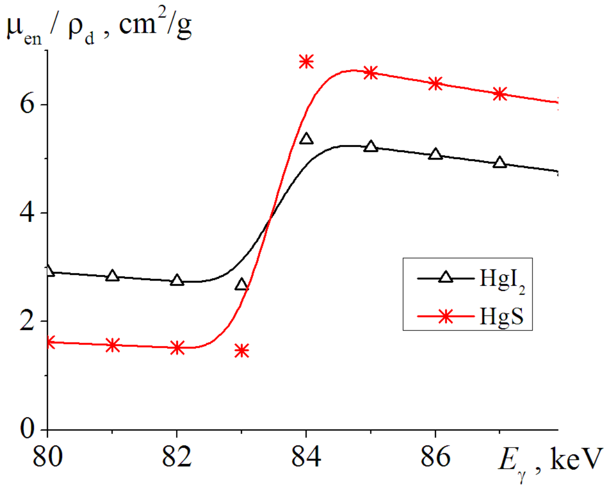

Fig.6. *The energy dependence of mass energy coefficient in the HgI_2- and HgS-detectors in low-energy range*

In order to use advantages of HgS semiconductor compound (chemical stability, mechanical strength) it is necessary to improve the growth technologies of mercuric sulfide. The principle problem consists in increasing values of charge carrier transport in HgS up to level of averaged values which are typical for currently available mercuric iodide. In this case, as the simulation shows (see Fig. 7, plot – HgS, detector no. 2), the sensitivity of HgS-detector can exceed the sensitivity of the same HgI_2-detector in the high-energy gamma-quantum range at 10...12%. The thickness of serial HgS-detectors can be increased up to 3 mm. In this case, the theoretical value of their sensitivity in the high-energy gamma-quantum range can exceed the sensitivity of serial HgI_2-detectors up to 15%. For further improvement of characteristics of HgS-detectors the grown single-crystals of mercuric sulfide have to allow to use higher bias.

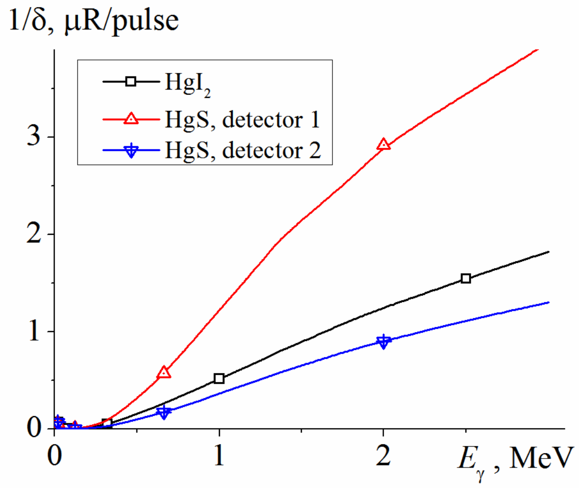

Fig.7. *The dependence of 1/δ value versus gamma-quantum energy for HgI_2- and HgS-detectors*

4. CONCLUSIONS

Mercuric sulfide (HgS) is a new promising material for production of room-temperature gamma-radiation detectors. The our calculation showed that for using advantages of HgS (chemical stability, mechanical strength) it is necessary to improve the growth technology of mercuric sulfide in order to increase mobility of charge carriers. Achieving mobility values of electrons and holes which are typical for currently available mercuric iodide the sensitivity of HgS-detectors in the high-energy gamma-quantum range (above 1 MeV) can exceed the sensitivity of the same HgI_2-detectors up to 15%. For further increasing sensitivity it is necessary to obtain the mercuric sulfide crystals with higher values of electrons and holes mobility.

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МОДЕЛИРОВАНИЕ ДЕТЕКТОРОВ ГАММА-ИЗЛУЧЕНИЯ НА ОСНОВЕ СОЕДИНЕНИЙ РТУТИ

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Метод Монте-Карло использован для сравнения характеристик HgI_2- и HgS-детекторов, которые работают в режиме анализа амплитуд импульсов. Для моделирования траекторий гамма-квантов и расчета функций отклика детекторов в области энергий от 25 кэВ до 3 МэВ использованы пакеты GEANT4 v4.9.4p04 и EGSnrc v4.r2.3.2. Исследуемые детекторы были оснащены планарными омическими контактами, а их размеры были 3,14 мм^2 × 1 мм. В диапазоне энергий 0,025...3 МэВ для детекторов были рассчитаны эффективность регистрации гамма-квантов и зависимость чувствительности от энергии. Показано, что в области сравнительно высоких энергий гамма-квантов эффективность преобразования энергии квантов в зарядовый сигнал для HgS-детекторов может превышать эффективность HgI_2-детекторов на 10...12 процентов.

МОДЕЛЮВАННЯ ДЕТЕКТОРІВ ГАММА-ВИПРОМІНЮВАННЯ НА ОСНОВІ СПОЛУК РТУТІ

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Метод Монте-Карло використаний для порівняння характеристик HgI_2- і HgS-детекторів, які працюють у режимі аналізу амплітуд імпульсів. Для моделювання траєкторій гамма-квантів та розрахунку функцій відгуку детекторів в області енергій від 25 кеВ до 3 МеВ використані пакети GEANT4 v4.9.4p04 і EGSnrc v4.r2.3.2. Досліджувані детектори були оснащені планарними омічними контактами, а їх розміри були 3,14 мм^2 × 1 мм. У діапазоні енергій 0,025...3 МеВ для детекторів були розраховані ефективність реєстрації гамма-квантів і залежність чутливості від енергії. Показано, що в області порівняно високих енергій гамма-квантів ефективність перетворення енергії квантів у зарядовий сигнал для HgS-детекторів може перевищувати ефективність HgI_2-детекторів на 10...12 відсотків.