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DESIGN AND MANUFACTURE OF SCINTILLATOR-PMT TYPE DETECTORS BASED ON ZnSe(Al) AND UPS-923A PLASTIC SCINTILLATOR FOR α-, β- AND MIXED α-β-RADIATION DEVICES

Introduction. *The search for technical solutions for the creation of effective α- and β- radiation detectors is a global trend in the field of radiation monitoring.*

Problem Statement. *The characteristics of α- and β-particle detectors can be improved by using materials with optimized parameters, original designs and technology.*

Purpose. *The purpose of this research is to develop and create a technology framework for manufacturing scintillation detectors based on activated zinc selenide ZnSe(Al) and plastic scintillator (PS) of UPS-923A type polystyrene for registration of α-, β- and α-β-radiations.*

Material and Methods. *ZnSe(Al) crystal, PS based on UPS-923A polystyrene, polymethyl methacrylate (PMMA) have been used; the hot pressing method; the detector parameters under irradiation with α- and β-particles have been tested with the use of the spectrometry and spectrophotometry methods.*

Results. *The following process techniques have been elaborated: for manufacturing the α-detector based on a thin layer of ZnSe(Al) fine-crystalline scintillator applied to a PMMA plate, which operates in the counting mode of registration (sensitivity > 0.15 pulse per sec/Bq (pps/Bq) (^{239}Pu)); for manufacturing the PS plates of UPS-923A polystyrene with a given thickness and area by the hot pressing method and the β-detectors based on them, which operate in the counting mode of registration (sensitivity > 0.28 imp · s^{-1}/Bq (^{90}Sr-^{90}Y)); for manufacturing*

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the α-β-detector based on a thin layer of fine-crystalline ZnSe(Al) applied to PS plate of UPS-923A polystyrene, which operate in the counting mode of registration (sensitivity > 0.15 pps/Bq (^{239}Pu) and > 0.28 pps/Bq (^{90}Sr-^{90}Y)); for manufacturing the α-β-detector with the use of a thin monocrystalline plate of ZnSe(Al) scintillator optically connected to a PMMA plate, which operates in the spectrometric mode of registration (the detector simultaneously registers α- and β-particles with spectrum separation, α/β ratio > 0.85, sensitivity > 0.3 pps/Bq (^{239}Pu), and > 0.28 pps/Bq (^{90}Sr-^{90}Y)).

Conclusions. *The developed detectors are as good as the world analogs and provide signal registration in both counting and spectrometric modes.*

Keywords: alpha-beta radiation, detector, scintillator, zinc selenide, polystyrene, and polymethyl methacrylate.

One of the tasks of radiation control is to prevent the harmful effects of radiation on the humans and animals, as well as on various objects of the natural environment (soil, water, air, plants, etc.). At present, for meeting radiation safety standards, the most important problem of radiation control is detecting the short-range alpha- (α) and beta- (β) particles, which are the biggest factor of natural radiation effect on humans. To date, mass α-radiation recording devices are manufactured with the use of luminophore ZnS(Ag) [1—4]. It has an alpha-beta ratio (α/β) close to 1 and a high luminescence brightness with a luminescence maximum of 450 nm [5], however, possesses a significant phosphorescence [6].

The characteristics of α-particle detectors can be improved by using other materials, in particular, scintillators based on zinc selenide (ZnSe). ZnSe-based scintillators belong to the class of semiconductor materials of the A^2B^6 group and are widely used in X-ray detectors of the scintillator-photodiode type for advanced multi-channel low-energy radiation means for visualizing the hidden image (nondestructive control systems, medical tomography, radiography, etc.) [7—10]. Scintillating zinc selenide crystals are effectively grown from the melt by the Bridgman-Stockbarger method [11], both nominally pure and doped. Such crystals have a high reproducibility of scintillation parameters, due to which they are serially manufactured for the use in X-ray detectors [12]. As compared with ZnS(Ag) [6, 13], the advantages of scintillators based on activated zinc selenide ZnSe(Al) are light output of 80,000 photons/MeV, afterglow < 0.01% after 3 ms, decay time 1÷3 μs [11], α/β ratio from 0.8 to 4.4 [14—16], and the absence of phosphorescence, which is an additional factor for increasing the detector sensitivity. Importantly, it is possible to manufacturing detectors having a large area, due to the use of composite scintillators with a high uniformity of parameters over the area. Depending on the design of the detectors, it is possible to create modifications that operate in the spectrometric mode or in the pulse counting mode.

Ionization chambers, scintillation counters with organic crystals (anthracene, stilbene, scintillation plastic based on polystyrene) [17—20], Geiger-Muller counters are used as detectors for registration of β-particles. Today, the plastic scintillators are the most promising for β-particle detectors due to their low density and low effective atomic number, which reduces sensitivity to gamma (γ) radiation.

The purpose of this research is to develop and create the technology framework for manufacturing scintillation detectors based on activated zinc selenide ZnSe(Al) and UPS-923A type polystyrene scintillator for registration of α-, β- and mixed α-β-radiations.

To achieve this, it is necessary to create and modify the technological process of development and manufacture of detectors of various designs with given parameters of sensitivity and efficiency of registration of α- and β-particles in both counting and spectrometric modes, which match the known world analogs. As compared with the existing samples, this can be achieved by using a combination of scintillation crystals of zinc selenide doped with aluminum ZnSe(Al) and transparent plastics obtained at the Institute for Scintillation Materials of the National Academy of Sciences of Ukraine. The results of the research allow expanding the range of objects with the use of new scintillation detectors.

Single crystals of zinc selenide doped with a controlled admixture of aluminum, ZnSe(Al), have been used as raw materials for the manufacture of alpha and alpha-beta detectors. The crystals are grown from the melt by the Bridgman-Stockbarger method under the pressure of an inert gas (argon) of 1—2 MPa at a temperature of 1850 K; the growth rate varies within 0.5—2 mm/h. This growing method ensures a high level of crystal purity (5N) and a slight deviation from the stoichiometric composition, which does not exceed 0.004%. The dopant concentration in the crystal is 0.001 wt.%.

The finely dispersed ZnSe(Al) crystalline powder is obtained by mechanical grinding of the grown crystals. This method allows the use of crystals with mechanical defects or substandard crystal remnants. Such use of the material reduces the amount of waste from machining the crystals and increases the environmental friendliness and cost-effectiveness of the production.

The UPS-923A type plastic scintillator based on polystyrene is used as a raw material for the manufacture of beta detectors. The UPS-923A plastic scintillator is based on linear polystyrene that contains two luminescent additives (2% paraterphenyl and 0.1% 1,4-bis-2-(5-phenyloxazolyl)-benzene (POROP)) [21].

CO-120 optically transparent polymethyl methacrylate (PMMA) is used as a light guide, due to the absence of impurities in its spectrum that absorb in the wavelength range of 400—800 nm.

The transmittance spectra in the visible region are measured with the use of a Shimadzu UV mini-1240 spectrophotometer operating in the range of 200—1100 nm with a resolution of 1 nm.

The parameters of the sensitivity and non-uniformity of the sensitivity of the detectors over the area are calculated on the basis of measurements made by *Canberra Multiport II* spectrometric complex. ^{239}Pu isotope (E_α = 5.156 MeV) is used as a source of α-particles, while ^{90}Sr-^{90}Y (E_β = 2.2 MeV) and ^{207}Bi (E_β = 976 keV) isotopes are used as a source of β-particles. The R1307 (maximum sensitivity at 420 nm) and R669 (maximum sensitivity at 600 nm) *Hamamatsu* PMTs are employed as photodetectors. The detector sensitivity is calculated as the ratio of the difference between the detector count rate and the background count rate to the source activity. The non-uniformity of the detector parameters is defined as the ratio of the deviations of the smallest and largest measured values to the average.

1. MANUFACTURE OF α-DETECTORS THAT OPERATE AS RADIATION COUNTERS

There are known research works on the creation of alpha detectors based on composite scintillators [22] that consist of powdered scintillation material ZnSe with a granule size of 200—240 μm and an immersion medium. However, these detectors have not been widely employed because of sufficiently large size of the granules and, as a result, the non-uniformity of the sensitivity of the detector over the area, as well as the difficulty of ensuring a low sensitivity to the γ-background.

The idea of our development is to create an original design of a combined detector using a thin scintillator layer based on fine-crystalline ZnSe(Al) deposited on a polymethyl methacrylate plate. This type of alpha detector operates as a counter. Transparent polymethyl methacrylate is used in the detector as a light guide material. It is important that PMMA has no absorption bands in the luminescence region of the scintillator. The presence of a light guide ensures the mechanical strength of the scintillation layer and ease of mounting the photoreceiver. If the area of the photoreceiver is smaller than the area of the scintillator, the use of a light guide in the form of a truncated cone also improves the conditions for collecting light from a large area of the ZnSe(Al) scintillation layer.

In order to develop a zinc selenide based alpha detector of a higher efficiency, as compared with serially manufactured detectors based on ZnS(Ag) luminophor [23—25], we proceed from the fact that the thickness of the detecting layer should

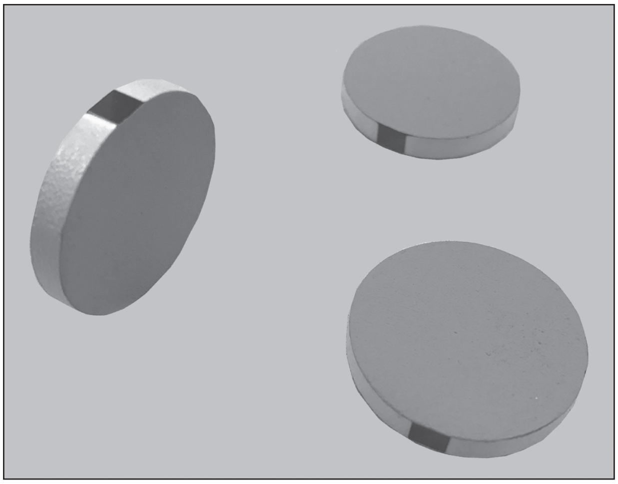

Fig. 1. Alpha-detectors, size ∅ 60 mm × 5.1 mm, operating as counters

be of the order of the path of α-particles in this substance, i.e. about 100 μm, at the energy of α-particles E_α = 5.1 MeV.

At the first stage of detector manufacture, a fine crystal powder is obtained by mechanical grinding of scintillating single crystal ZnSe(Al). The necessary fraction of powder granules is selected by sieving through a system of calibrated sieves. At the second stage, a scintillation coating is prepared: the calculated amount of ZnSe(Al) powder of the selected fraction is mixed with water-soluble acrylic varnish in a ratio of 3 : 1. The use of a water-soluble acrylic varnish as a binder component allows us to effectively absorb α-particles directly in the scintillation coating layer due to the fact that during polymerization, the aqueous solvent of the acrylic varnish evaporates and a thin layer of varnish is formed on the surface of the chalcogenide luminophor, which does not significantly affect the sensitivity of alpha-detector. Also, it should be noted that water-soluble acrylic varnish does not contain additional absorption bands in the transmission spectrum. At the third stage, we spread a scintillation coating, layer by layer, on an optically transparent PMMA substrate with a size of ∅ 60 mm × 5 mm, while controlling the coating thickness that is 100 μm. To minimize light loss, a reflective coating based on titanium dioxide and acrylic varnish is applied on the side surface of the PMMA substrate plate.

While working out the modes for alpha detectors, we have determined the composition and conditions for obtaining a thin layer of scintillation coating based on fine-crystalline ZnSe(Al) [26]. The experimentally determined optimal conditions for obtaining a scintillation coating based on ZnSe(Al) are as follows:

- the size of ZnSe(Al) luminophor particles is 10 ± 5 μm. In this case, a high level of selectivity is achieved when registering α-particles relative to β-particles and γ-quanta due to the fact that α-particles absorbed in a thin layer of the scintillation coating, and β-particles and γ-quanta penetrate the scintillation coating almost without absorption;
- the mass fraction of fine-crystalline ZnSe(Al) in the composition of the scintillation coating is 75 ± 5 wt.%. Reducing the content of finely dispersed scintillator to less than 70 wt.% is impractical because the fact that the thickness of the varnish layer on the ZnSe(Al) surface increases, which leads to a deterioration of the penetration of α-particles into the phosphor and a decrease in the sensitivity of the alpha detector. Increasing the content of finely dispersed luminophor up to more than 80 wt.% leads to a decrease in the mechanical strength of the scintillation coating and, possibly, to its separation from the optical substrate;
- the surface density of the scintillation coating is 11.0 ± 0.25 mg/cm^2. In this case, effective absorption of short-range α-particles in the scintillation coating and conversion of ionized α-particle energy into a scintillation signal are ensured due to the presence of a thin layer of varnish and a developed surface of ZnSe(Al) luminophor with a high concentration of surface point defects.

Figure 1 shows a photograph of test samples of the alpha detectors.

The amplitude spectra of the prototype of the alpha detector have been measured under irradiation by α-source ^{239}Pu, the spectra are shown in

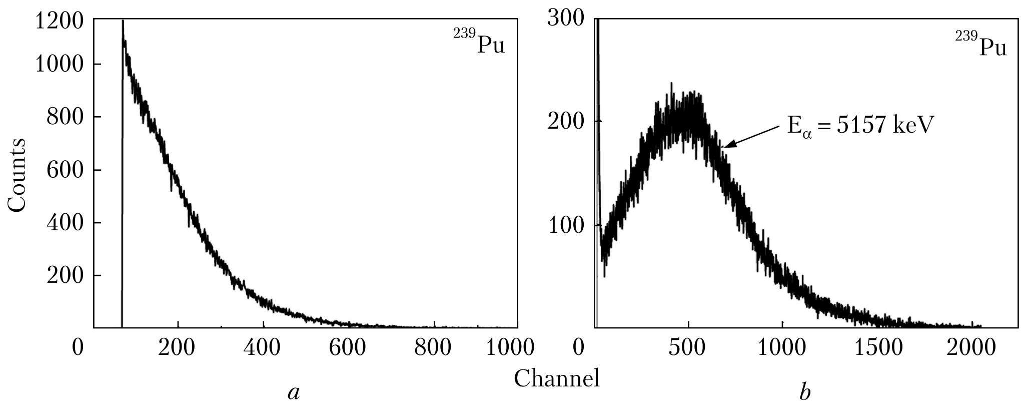

Fig. 2. Amplitude spectra of α-detector with scintillation coating based on ZnSe(Al) in the case of irradiation by ^{239}Pu: *a* — PMT R1307; *b* — PMT R669

Fig. 2 for different types of PMTs: R1307 (*a*) and R669 (*b*). A distinctive feature of the amplitude spectrum in Fig. 2, *b* is that there is observed a clear alpha peak that corresponds to the spectrometric detectors, in contrast to the spectrum in Fig. 2, *a*, which is typical for the counting detectors. This result demonstrates the advantage of using PMTs of the R669 type in the manufacture of α-radiation detection units.

The parameters of the obtained test samples of alpha detectors have been tested in the counting mode. The test results are shown in Table 1.

The specified manufacturing conditions of the combined alpha detector with a scintillation coating based on ZnSe(Al) ensure the sensitivity of the detector to α-particles of ^{239}Pu isotope higher than 0.15 imp·s^{-1}/Bq and the non-uniformity of sensitivity over the area is, at least, ±1.5 %. The sensitivity of the alpha detector with an active surface diameter of 60 mm with the use of R699 photodetector is 0.36 imp · s^{-1}/Bq. It is 1.5 times higher as compared with that of the well-known analog based on ZnS(Ag) luminophor, which is equal to 0.25 imp · s^{-1}/Bq [27].

Thus, the materials, design, and manufacturing technology proposed by us have made it possible to obtain test samples of alpha detectors that have a much larger area and the parameters that are as good as those of the existing analogs of alpha detectors. The detectors have the following characteristics:

- the scintillator: ZnSe(Al) powder with a particle size of 10 ± 5 μm (the layer thickness is up to 0.1 mm);
- the optical substrate (light guide): polymethyl methacrylate;
- Hamamatsu PMTs: R1307, R669;
- the operating voltage of the detector: 500—1200 V;
- the operating temperature: –20 °C ... +60 °C;
- the range of registered energies: for α-radionuclides, from 2000 to 10000 keV.

2. MANUFACTURE OF β-DETECTORS THAT OPERATE AS RADIATION COUNTERS

To solve the problem of registration of β-radiation in the γ-background field, the most suitable device is a detector based on a thin plastic scintillator [19]. It has been shown in [28] that the opti-

Table 1. **The Main Parameters of α-Radiation Detectors**

Parameter and measuring unit	Value
Active surface diameter, mm	Up to 200
Impulse rise time (0—100%), ^{239}Pu, ns	55—60
Impulse fall time (1/e), ^{239}Pu, ms	1.7—1.8
Sensitivity, ^{239}Pu, imp · s^{-1}/Bq	>0.15
Non-uniformity of sensitivity over the area, ^{239}Pu, %	±1.5

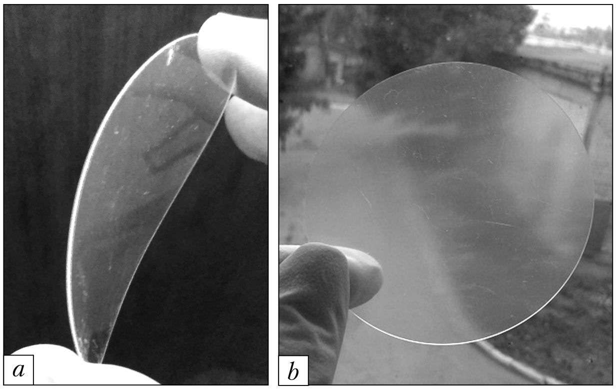

Fig. 3. Samples of plastic scintillator, size ∅ 80 mm × 0.2 mm, obtained by the hot pressing method (*a*) and surface machining (*b*)

mal thickness of the scintillator for effective registration of β-particles is 0.2 mm. The authors of [29] have also established that the theoretical maximum efficiency of recording fast neutrons by hydrogen-containing scintillators with a thickness of 25 mm, 51 mm, 102 mm, and 203 mm is 27%, 55%, 82%, and 90%, respectively. The thickness of the plastic scintillator of 0.2 mm ensures a satisfactory registration sensitivity and a high selectivity of the detector to β-particles against the background of neutron-gamma radiation. The existing methods of obtaining transparent plastic scintillators have certain disadvantages, the main of which is the mechanical processing of the scintillation element, which leads to surface degradation over time, which affects the initial parameters of the detector.

The idea of our development was to manufacture detection units using a thin optically transparent plastic scintillator plate based on UPS-923A polystyrene. This type of detector works as β-radiation counter. We have developed a technology for manufacturing plastic scintillator plates of the required thickness and size by the hot pressing method that has a number of advantages, namely: long service life, absence of mechanical stress, optically polished surface, manufacture of products of the required thickness and area.

A plastic scintillator of the UPS-923A type with a size of 10 mm × 10 mm × 10 mm is used as a raw material for the production of a prototype beta detector. The pressing is carried out with the use of special equipment in a vacuum cabinet in an atmosphere of inert argon. The sample is heated to a temperature of 423 K at a rate of 20 deg/h, after which it is kept at this temperature for a day. Then the temperature is raised up to 433 K and the sample is kept in such conditions for one more day. After that, it is cooled at a rate of 10 deg/h.

The temperature mode of pressing, the equipment elements and the mass of raw materials are selected in such a way as to obtain a transparent plastic scintillator plate of a given size (∅ 80 mm × × 0.2 mm) and the required class of surface treatment. A photo of the sample obtained by the hot-pressing method is shown in Fig. 3, *a*. For comparison, Fig. 3, *b* features a photograph of the sample obtained by the method of mechanical cutting and polishing, which shows the deterioration of the sample transparency as a result of damages of the surface layer during mechanical treatment.

We have measured the optical transmission spectra in the visible range of comparative samples of the plastic scintillator shown in Fig. 4, *a*. The spectra show that in the wavelength range of 340—500 nm, the sample pressed according to our method (curve *1*) has no absorption bands, with the optical transmittance being by approximately 7% higher, which indicates the presence of a deeper defective layer on the surface of the samples treated by other methods (curves *2* and *3*).

The amplitude spectra of the comparative samples of the plastic scintillator under irradiation with a ^{90}Sr-^{90}Y source of β-particles have been measured as well and are shown in Fig. 4, *b*. The spectra demonstrate a satisfactory sensitivity of the samples to the registration of β-particles, with the sample obtained by the method proposed herein (curve *1*) having a higher sensitivity as compared with other samples (curves *2* and *3*). Therefore, the application of the hot-pressing method for making a plastic scintillator based on polystyrene is a promising technique for manufacturing beta detectors, which ensures their satisfactory sensitivity to β-radiation.

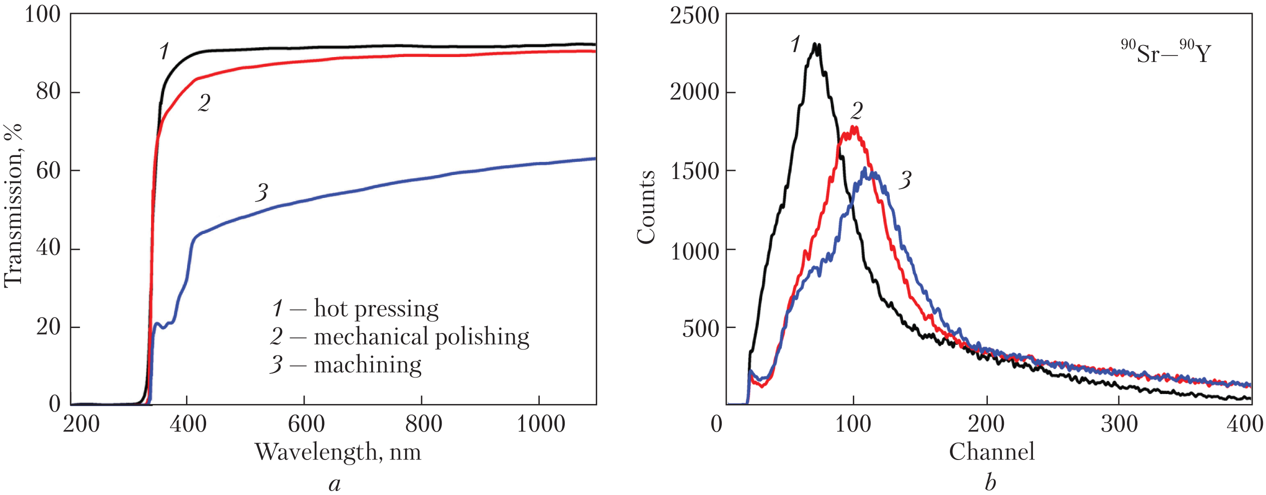

Fig. 4. Optical transmission spectra in the visible region (*a*) and amplitude spectra of β-detectors in the case of irradiation by ^{90}Sr-^{90}Y (*b*) of plastic scintillator samples obtained by the hot pressing method (*1*), mechanical polishing (*2*) and surface machining (*3*)

The β-radiation detectors are made on the basis of the obtained plastic scintillator samples. Figure 5 shows a photograph of the obtained beta detector.

The parameters of the obtained prototypes of beta detectors have been tested in the counting mode. The test results are shown in Table 2.

So, the material, design, and process technology proposed by us has made it possible to obtain prototypes of beta detectors of a large area, a given thickness and a higher quality scintillator surface with parameters that are as good as the existing world analogs [18]. The detectors can register β-radiation in the counting mode. They have the following characteristics:

- the scintillator: UPS-923A-type polystyrene (the thickness is up to 4 mm);
- Hamamatsu PMT: R1307;
- the operating voltage of the detector: 500—1200 V;
- the operating temperature: –20 °C...+60 °C;
- the range of registered energies: for β-radionuclides, from 65 to 4000 keV.

3. MANUFACTURE OF α–β-DETECTORS THAT OPERATE AS RADIATION COUNTERS

There are known research works [30—32] on the creation of alpha-beta detectors, which are a combination of two scintillation materials, namely

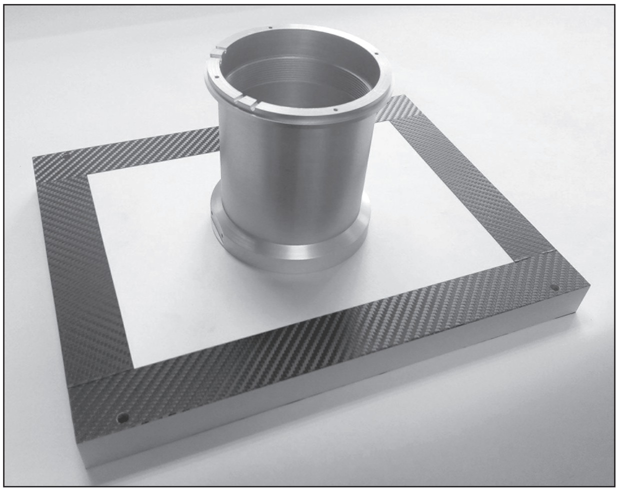

Fig. 5. Beta-detector based on plastic scintillator of UPS-923A polystyrene, which operates as counter

Table 2. **The Main Parameters of the β-Detectors that Operate as Counters**

Parameter and measuring unit	Value
Active surface diameter, mm	Up to 200
Impulse rise time (0—100%), ^{90}Sr-^{90}Y, ns	60—65
Impulse fall time (1/e), ^{90}Sr-^{90}Y, ms	270—280
Sensitivity, ^{90}Sr-^{90}Y, imp · s^{-1}/Bq	>0.28
Minimum detecting activity, ^{90}Sr-^{90}Y, Bq	0.15

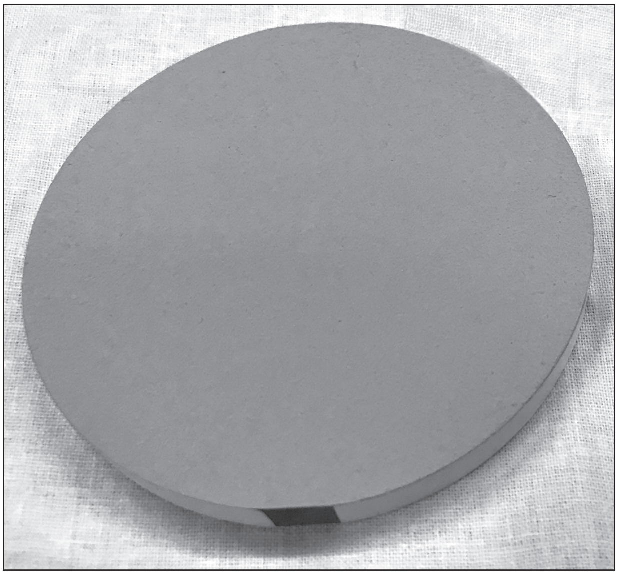

Fig. 6. Alpha-beta detector, size ∅ 85 mm × 0.6 mm, operating as counter

ZnS(Ag) and a plastic scintillator, each being responsible for a certain channel of radiation registration. However, as mentioned before, there are certain disadvantages of the ZnS(Ag) luminophor, for example, the kinetics of luminescence that depend on the afterglow parameter, which do not allow increasing the load of the detector for measuring highly active samples.

The idea of our development is to create an original design of a combined detector using a thin scintillator layer based on fine crystal ZnSe(Al) applied on an optically transparent plate of a plastic scintillator based on UPS-923A polystyrene. This type of alpha-beta detector works in the counting mode of signal registration. A transparent plate of a plastic scintillator is used in this type of detectors, as a detector of β-particles and as a light guide to improve the conditions for collecting light from a large area of the scintillator and for mounting the photodetector.

The test alpha-beta detectors operating in the counting mode have been manufactured by a process technique that is similar to that for e production of alpha detectors, which is described in Chapter 1. A distinctive feature is the use of an optical substrate made of a different material: we have used a plastic scintillator of the UPS-923A type based on polystyrene, manufactured by the hot-pressing method, which is described in Chapter 2, instead of PMMA. Similarly to the described in Chapter 1, we have used the composition of a thin layer of scintillation coating based on fine-crystalline ZnSe(Al) and elaborated the technology for its application on an optical substrate that is a plastic scintillator.

Figure 6 shows a photograph of the obtained alpha-beta detector.

We have measured the amplitude spectra of the prototype of the alpha-beta detector while irradiating them with α-particles ^{239}Pu (Fig. 7, *a*) and β-particles ^{90}Sr-^{90}Y (Fig. 7, *b*). The use of a scintillation coating based on fine-crystalline ZnSe(Al) applied on an optical substrate made of a plastic scintillator provides a satisfactory sensitivity to α-radiation without loss of sensitivity to β-radiation.

The parameters of the obtained test samples of alpha-beta detectors have been tested in the counting mode. The test results are shown in Table 3. The developed detectors have a high uniformity of the scintillation signal and the sensitivity throughout the area for irradiation by a collimated ^{239}Pu source, which is ensured by the use of the composite scintillation layer and makes it possible to manufacture detectors that have a significant area.

Table 3. **The Main Parameters of α-β-Radiation Detectors**

Parameter and measuring unit	Value
Active surface diameter, mm	Up to 200
Impulse rise time (0—100%), ^{239}Pu, ns	55—60
Impulse fall time (1/e), ^{239}Pu, ms	1.7—1.8
Sensitivity, ^{239}Pu, imp · s^{-1}/Bq	>0.15
Non-uniformity of sensitivity over the area, ^{239}Pu, %	±1.5
Impulse rise time (0—100%), ^{90}Sr-^{90}Y, ns	60—65
Impulse fall time (1/e), ^{90}Sr-^{90}Y, ms	270—280
Sensitivity, ^{90}Sr-^{90}Y, imp · s^{-1}/Bq	>0.28

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The materials, design, and manufacturing technology offered by us has made it possible to obtain prototypes of alpha-beta detectors with having a much larger area and the parameters that are as good as the existing world analogs of detectors based on ZnS(Ag). The detectors can operate as counters. For the detector with an active surface diameter of 85 mm, the sensitivity of α-particle registration is 0.36 imp · s^{-1}/Bq, while that of β-particles is 0.28 imp · s^{-1}/Bq, which exceeds the parameters of the detector based on ZnS(Ag) applied on scintillation plastic [32].

The detectors have the following characteristics:

- the scintillator: ZnSe(Al) powder with a particle size of 10 ± 5 μm (the layer thickness is up to 0.1 mm) and UPS-923A type polystyrene (the thickness is up to 0.5 mm);
- optical substrate (light guide): UPS-923A-type polystyrene;
- Hamamatsu PMTs: of R1307, R669;
- the operating voltage of the detector: 500—1200 V;
- the operating temperature: –20 °C ... +60 °C;
- the range of registered energies: for α-radionuclides, from 2000 to 10000 keV; for β-radionuclides, from 65 to 4000 keV.

4. MANUFACTURE OF α-β-DETECTOR THAT OPERATE AS RADIATION SPECTROMETERS

Known research works [33] on the creation of detectors for separate registration of α-, β-, and γ-radiations, which are a combination of two scintillation materials, namely ZnSe(Te) and CsI(Tl), each being responsible for a certain channel of radiation registration. However, these detectors have some disadvantages: the limited size of the detectors, as well as the use of CsI(Tl) that is hygroscopic and quickly degrades in terms of functional parameters.

The idea of our development is to create an original design of a combined detector using a thin polished plate of scintillation single crystal ZnSe(Al) that is optically connected to a transparent plate of polymethyl methacrylate. This type of alpha-beta detector works in the spectrometric mode of signal registration. A transparent PMMA plate is used in this type of detector as a light guide to improve the conditions for collecting light from a large area of the scintillator and mounting the photodetector.

In this case, the ZnSe(Al) monocrystalline layer serves as both an α- and a β-channel for recording radiation. The calculated path of β-particles with

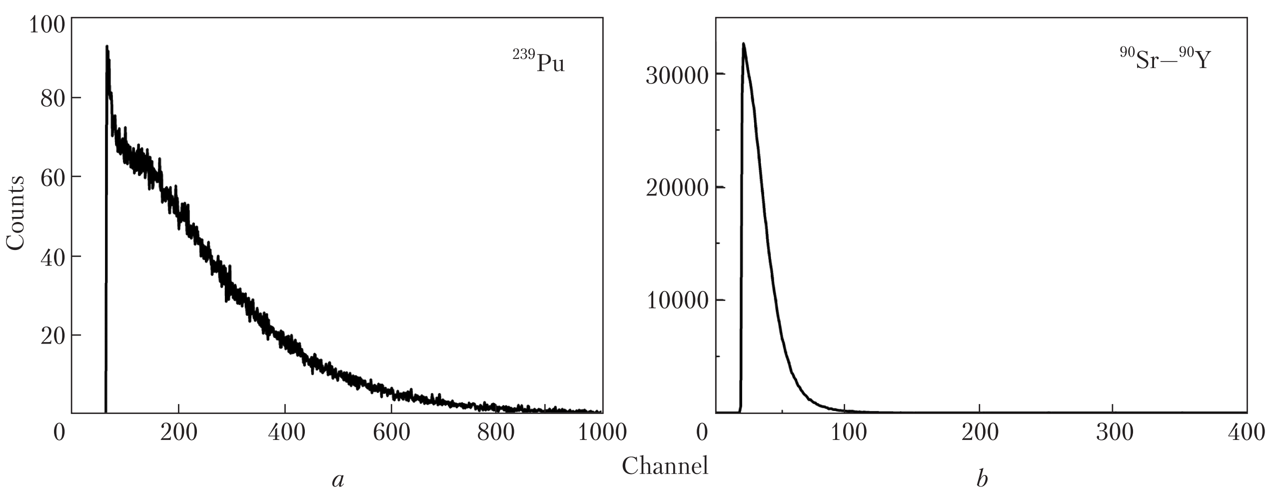

Fig. 7. Amplitude spectra of α–β-detector with scintillation coating based on ZnSe(Al) on plastic scintillator plate in the case of irradiation by: *a* — α-particles of ^{239}Pu; *b* — β-particles of ^{90}Sr-^{90}Y

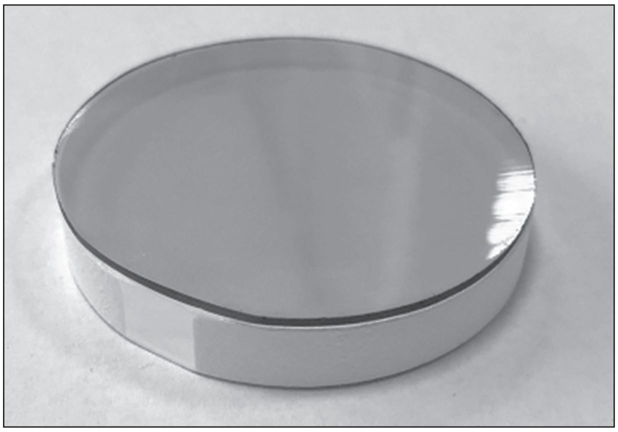

Fig. 8. Alpha-beta detector, size ∅ 50 mm × 9 mm, operating as radiation spectrometer

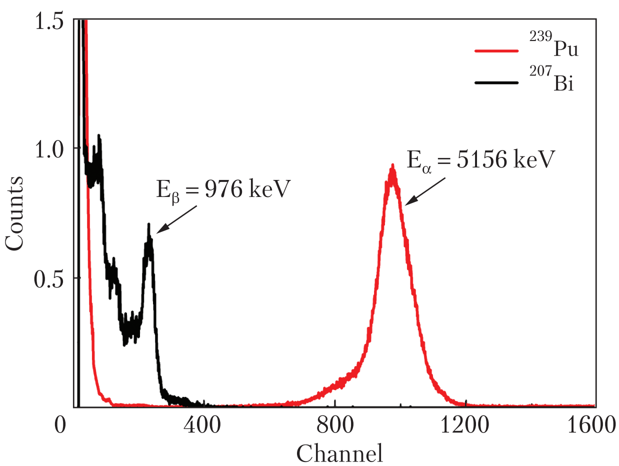

Fig. 9. Normalized amplitude spectra of α-β-detector based on a single-crystal ZnSe(Al) plate optically connected to a PMMA plate in the case of irradiation by ^{239}Pu and ^{207}Bi

an energy of 2.2 MeV in a ZnSe(Al) single crystal is approximately 1.6 mm. It should be noted that the ZnSe(Al) crystal with a thickness of 1.6 mm, in addition to α- and β-particles, also registers photons of γ-quanta. That is, it is necessary to determine the optimal thickness of the β-sensitive scintillation layer, which ensures the maximum absorption of β-particles, is transparent enough to output the scintillation light to the photodetector, and has the minimum efficiency of recording photons of γ-radiation. It was experimentally established that the thickness of the single crystal layer, which selectively registers only α- and β-particles, is approximately 1 mm.

The alpha-beta detector prototypes operating in the spectrometric registration mode have been manufactured by connecting a polished ZnSe(Al) scintillator crystal plate having a size of ∅ 50 mm × × 1 mm with the use of optical organosilicon glue with a polished PMMA plate having a size of ∅ 50 mm × 8 mm. The cleanliness class of the polished surface of the plates shall meet the requirements for optical crystals with a roughness level of R_a = 0.1—0.4 μm. It should be noted that the optical glue did not contain additional absorption bands in the transmission spectrum. To minimize light loss, a reflective coating based on titanium dioxide and acrylic varnish was applied on the side surface of the PMMA substrate plate.

Figure 8 shows a photograph of the obtained alpha-beta detector.

We have measured the amplitude spectra of the alpha-beta detector prototype for irradiation by ^{239}Pu α-particles and ^{207}Bi β-particles, which are shown in Fig. 9.

The application of a thin monocrystalline layer based on a ZnSe(Al) scintillator, which is optically connected to a layer of transparent polymethyl methacrylate, provides simultaneous registration of α- and β-particles and separation of spectra with an α/β ratio of 0.85. Testing the efficiency of registration of α- and β-radiations over the entire area of the scintillation detector has shown a high uniformity of the parameter.

The parameters of the obtained test samples of alpha-beta detectors have been tested in the spectrometric mode. The test results are shown in Table 4.

The materials, design, and manufacturing technology offered by us has made it possible to obtain the prototypes of combined alpha-beta detectors, which have a much larger area and the parameters that are as good as those of the existing world analogs based on ZnS(Ag). The detectors can register radiation in the spectrometric mode. For the detector with a diameter of an active surface of 50 mm, the sensitivity of registration of α-particles is 0.31 imp · s^{-1}/Bq while that of β-parti-

Table 4. **The Main Parameters of the α-β-Detectors**

Detector parameter and unit of measurement	Value
Diameter of the active surface, mm	Up to 200
Energy resolution (5156 kV), %	At least, 7.0 (∅ 50 mm) At least, 15 (∅ 200 mm)
α/β ratio (^{239}Pu, ^{207}Bi)	0.85
Registration efficiency (4ϖ), ^{239}Pu, %	> 35
Sensitivity, ^{239}Pu, impulse · s^{-1}/Bq	> 0.3
Non-uniformity of sensitivity over the area, ^{239}Pu, %	±1.5
Sensitivity, ^{90}Sr-^{90}Y, impulse · s^{-1}/Bq	> 0.28
Non-uniformity of sensitivity over the area, ^{90}Sr-^{90}Y, %	±1.5

cles is 0.34 imp · s^{-1}/Bq, which is by 26% and 14% higher, respectively, than in the case of detectors based on ZnS(Ag) deposited on scintillation plastic [27].

The detectors have the following characteristics:

- the scintillator: single crystal ZnSe(Al) (the plate thickness is up to 1 mm);
- the optical substrate (light guide): polymethyl methacrylate;
- Hamamatsu PMTs: of R1307, R669;
- the operating voltage of the detector: 500—1200 V;
- the operating temperature: –20 °C ... +60 °C;
- the range of registered energies: for α-radionuclides, from 2000 to 10000 keV; for β-radionuclides, from 65 to 4000 keV.

CONCLUSIONS

1. Combined alpha detectors based on a thin layer of fine-crystalline ZnSe(Al), which operate as counters have been designed and manufactured. It has been determined that the alpha detectors have a sensitivity of > 0.15 imp · s^{-1}/Bq for a ^{239}Pu radiation, with a uniformity over the area of ±1.5%. It has been established that the use of a 100 μm thick fine-crystalline scintillation layer ZnSe(Al) with a grain size of 10 ± 5 μm, a grain content of 75 ± 5 wt.% and a layer surface density of 11.0 ± 0.25 mg/cm^2 as part of the detector provides a high rejection of the alpha particles relative to the beta particles and the gamma quanta, due to the complete absorption of alpha particles in the scintillation layer.

2. A method of obtaining UPS-923A type polystyrene plastic scintillator plates of a given thickness and area by hot pressing has been developed and the beta detectors that operate as counters have been manufactured based on them. It has been shown that, due to the absence of machining of the plates, their optical transparency increases by approximately 7%, which provides the required counting rate and satisfactory sensitivity to beta radiation. It has been determined that the beta detectors have a sensitivity of > 0.28 imp · s^{-1}/Bq for a ^{90}Sr-^{90}Y radiation source.

3. The combined alpha-beta detectors based on a thin layer of fine-crystalline ZnSe(Al) applied on a transparent plate of a polystyrene plastic scintillator of the UPS-923A type, which operate as counters have been designed and manufactured. It has been determined that the alpha-beta detectors have a sensitivity of > 0.15 imp · s^{-1}/Bq for a ^{239}Pu radiation source with a uniformity over the area of ±1.5% and a sensitivity of > 0.28 imp × × s^{-1}/ Bq for a ^{90}Sr-^{90}Y radiation source. It has been shown that the use of a scintillation coating based on fine-crystalline ZnSe(Al) in the detector provides a satisfactory sensitivity to alpha radiation without loss of sensitivity to beta radiation.

4. Combined alpha-beta detectors with a thin monocrystalline ZnSe(Al) scintillator plate, which operate in the spectrometric mode have been designed and manufactured. This type of detector allows simultaneous registration of alpha and beta particles and spectrum separation with α/β ratio of > 0.85. It has been determined that the alpha-beta detectors have a registration efficiency (4π) of > 35% and a sensitivity of > 0.3 imp · s^{-1}/Bq, in the case of irradiation with ^{239}Pu source, and a sensitivity of > 0.28 imp · s^{-1}/Bq, in the case of irradiation with ^{90}Sr-^{90}Y source.

5. The research results have been implemented at the research and experimental site of the Insti-

tute for Scintillation Materials for the National Academy of Sciences of Ukraine. With the use of them we may introduce to the market a series of new scintillation detectors based on zinc selenide and a plastic scintillator for detecting alpha and alpha-beta radiation. It is expected that the creation of technology framework for the manufacture of these detectors will become the basis for the development of other types of radiation detectors.

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REFERENCES

1. ZnS(Ag) Zinc Sulfide Scintillation Material. © 2002 Saint-Gobain Ceramics and Plastics, Inc. URL: http://www.hep.ph.ic.ac.uk/fets/pepperpot/docs+papers/zns_602.pdf (Last accessed: 01.11.2022).
2. *Patent of USA № US7679064B2*. Particle detector and neutron detector that use zinc sulfide phosphors. Katagiri M. URL: https://patents.google.com/patent/US7679064 (Last accessed: 01.11.2022).
3. Lee, S. K., Kang, S. Ya., Jang, D. Yu., Lee, Ch. H., Kang, S. M., ..., Kim, Y. K. (2011). Comparison of new simple methods in fabricating ZnS(Ag) scintillators for detecting alpha particles. *Progress in Nuclear Science and Technology*, 1, 194—197. https://doi.org/10.15669/pnst.1.194.
4. Morozova, N. K., Kuznetsov, V. A. (1987). *Zinc sulfide: preparation and optical properties* (Eds. M. V. Fok). Moscow [in Russian].
5. Foster, J. (2006). *A comparison of the ZnS(Ag) scintillation detector to the silicon semiconductor detector for quantification of alpha radioactivity in aqueous solutions*. All Theses. Clemson University: TigerPrints. URL: https://tigerprints.clemson.edu/all_theses/10 (Last accessed: 01.11.2022).
6. Nikl, M. (2006). Scintillation detectors for X-rays. *Meas. Sci. Technol.*, 17(4), 37—54. https://doi.org/10.1088/0957-0233/17/4/R01.
7. Ryzhikov, V., Starzhinskiy, N., Galchinetskii, L., Gashin, P., Kozin, D., Danshin, E. (2001). New semiconductor scintillators ZnSe(Te,O) and integrated radiation detectors based thereon. *IEEE T. Nucl. Sci.*, 48(3), 356—359. https://doi.org/10.1109/23.940080.
8. Ryzhikov, V., Starzhinskiy, N. (2005). Properties and peculiar features of application of isoelectronically doped A^2B^6 compound-based scintillators. *Journal of Radiation Protection and Research*, 30(2), 77—84. URL: https://www.jrpr.org/upload/pdf/BSBOB5_2005_v30n2_77.pdf (Last accessed: 01.11.2022).
9. Lee, W. G., Kim, Y. K., Kim, J. K., Seo, H. J., Ryzhikov, V., Starzhinskiy, N., ..., Zelenskaya, O. (2006). Particularities of ZnSe-based scintillators for a spectrometry of charged particles and gamma quanta. *Journal of the Korean Physical Society*, 48(1), 47—50. URL: https://www.jkps.or.kr/journal/view.html?uid=7413&vmd=Full (Last accessed: 01.11.2022).
10. Ryzhikov, V. D., Galchinetskii, L. P., Starzhinskiy, N. G., Danshin, E. A., Katrunov, K. A., Chernikov, V. V. (2001). Combined detectors of charged particles based on zinc selenide scintillators and silicon photodiodes. *Problems of Atomic Science and Technology. Nuclear Physics Investigations*, 5, 174—176. URL: https://vant.kipt.kharkov.ua/ARTICLE/VANT_2001_5/article_2001_5_174.pdf (Last accessed: 01.11.2022).
11. Ryzhikov, V., Grinyov, B., Galkin, S., Starzhinskiy, N., Rybalka, I. (2013). Growing technology and luminescent characteristics of ZnSe doped crystals. *J. Cryst. Growth*, 364, 111—117. https://doi.org/10.1016/j.jcrysgro.2012.11.034.
12. Galkin, S. M., Rybalka, I. A., Tupitsyna, I. A., Zvereva, V. S., Litichevskiy, V. A. (2016). The development of flexible scintillation panels based on chalcogenide and oxide phosphors for advanced X-ray scanners and tomographs. *Sci. Innov.*, 12(6), 37—45. https://doi.org/10.15407/scine12.06.037.
13. van Eijk, Carel W. E. (2002). Neutron PSDs for the next generation of spallation neutron sources. *Nuclear Instruments and Methods in Physics Research A*, 477, 383—390. https://doi.org/10.1016/S0168-9002(01)01836-8.
14. Beeman, J. W., Bellini, F., Cardani, L., Casali, N., Dafinei, I., Di Domizio, S., ..., Vignati, M. (2013). Performances of a large mass ZnSe bolometer to search for rare events. *Journal of Instrumentation*, 8, P05021. https://doi.org/10.1088/1748-0221/8/05/P05021.
15. Nagorny, S., Cardani, L., Casali, N., Dafinei, I., Pagnanini, L., Pattavina, L., ..., Schaeffner, K. (2017). Quenching factor for alpha particles in ZnSe scintillating bolometers. *IOP Conf. Series: Materials Science and Engineering*, 169, 012011. https://doi.org/10.1088/1757-899X/169/1/012011.

16. Arnaboldi, C., Capelli, S., Cremonesi, O., Gironi, L., Pavan, M., Pessina, G., Pirro, S. (2011). Characterization of ZnSe scintillating bolometers for Double Beta Decay. *Astroparticle Physics*, 34(6), 344—353. https://doi.org/10.1016/j.astropartphys.2010.09.004.
17. Maekawa, T., Sumita, A., Makino, Sh. (1998). Thin beta-ray detectors using plastic scintillator combined with wavelength-shifting fibers for surface contamination monitoring. *Journal of Nuclear Science and Technology*, 35(12), 886—894. https://doi.org/10.1080/18811248.1998.9733961.
18. Miramonti, L. (2002). A plastic scintillator detector for beta particles. *Radiation Measurements*, 35(4), 347—354. https://doi.org/10.1016/S1350-4487(02)00051-3.
19. Pourtangestani, K., Machrafi, R. (2012). Optimization of plastic scintillator thicknesses for online beta/gamma detection. *EPJ Web of Conferences*, 24, 07010. https://doi.org/10.1051/epjconf/20122407010.
20. Bae, J. W., Kim, H. R. (2020). Plastic scintillator beta ray scanner for in-situ discrimination of beta ray and gamma ray radioactivity in soil. *Nuclear Engineering and Technology*, 52(6), 1259—1265. https://doi.org/10.1016/j.net.2019.11.013.
21. Grinyov, B. V., Senchishin, V. G. (2003). *Plastic scintillators*. Kharkov [in Russian].
22. *Patent of Ukraine № UA103711C2*. Voronkin, Ye. F., Galkin, S. M., Lalaiants, O. I., Litichevskyi, V. O., Tarasov, V. O. Method for production of a scintillation element for registration of alpha-radiation [in Ukrainian]. URL: https://patents.google.com/patent/UA125108C2/en (Last accessed: 01.11.2022).
23. Alpha Detection EJ-440, EJ-442. © 2021 Eljen Technology. URL: https://eljentechnology.com/products/zinc-sulfide-coated/ej-440-ej-442 (Last accessed: 01.11.2022).
24. Alpha Detector Model 43-1. © 2022 Ludlum Measurements, Inc. URL: https://ludlums.com/products/health-physics/product/model-43-1 (Last accessed: 01.11.2022).
25. *Patent of USA № US7375336B2*. Hasegawa, I., Izaki, K., Kobayashi, H., Ino, K., Kanazawa, N. ZnS(Ag) scintillation detector. URL: https://patents.google.com/patent/US7375336B2/en (Last accessed: 01.11.2022).
26. *Patent of Ukraine № UA125108C2*. Boiaryntsev, A. Yu., Nepokupna, T. A., Galkin, S. M., Sibilieva, T. H. Scintillation coating for alpha detector [in Ukrainian]. URL: https://patents.google.com/patent/UA125108C2/en (Last accessed: 01.11.2022).
27. AT1329 sample counter. © 2023 ATOMTEX. URL: https://atomtex.com/en/at1329-sample-counter (Last accessed: 14.04.2023).
28. Kumar, A., Waker, A. J. (2012). An experimental study of the relative response of plastic scintillators to photons and beta particles. *Radiation Measurements*, 47(10), 930—935. https://doi.org/10.1016/j.radmeas.2012.08.003.
29. Reeder, P. L., Peurrung, A. J., Hansen, R. R., Stromswold, D. C., Hensley, W. K., Hubbard, C. W. (1999). Detection of fast neutrons in a plastic scintillator using digital pulse processing to reject gammas. *Nuclear Instruments and Methods in Physics Research A*, 422, 84—88. https://doi.org/10.1016/S0168-9002(98)01068-7.
30. Alpha/Beta Detection EJ-444. © 2021 Eljen Technology. URL: https://eljentechnology.com/products/zinc-sulfide-coated/ej-444 (Last accessed: 01.11.2022).
31. Alpha-Beta Detector Model 43-1-1. © 2022 Ludlum Measurements, Inc. URL: https://ludlums.com/products/all-products/product/model-43-1-1 (Last accessed: 01.11.2022).
32. Alpha and beta radiation detector BDPS-96. © 1995-2022 «SPE «TETRA» Ltd. Devices of radiation and technological control. URL: http://tetra.ua/en/production/blocks_and_devices/96/bdps-96.pdf (Last accessed: 01.11.2022).
33. Ryzhikov, V., Galchinetski, L., Galkin, S., Danshin, E., ..., Chernikov, V. (2000). Combined detectors based on ZnSe(Te), CsI(Tl) and Si-PIN-PD for separate detection of alpha, beta and gamma radiation. *IEEE Transactions on Nuclear Science*, 47(6), 1979—1981. https://doi.org/10.1109/23.903832.

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РОЗРОБКА ТА ВИГОТОВЛЕННЯ ДЕТЕКТОРІВ ТИПУ СЦИНТИЛЯТОР-ФЕП НА ОСНОВІ ZnSe(Al) ТА ПЛАСТМАСОВОГО СЦИНТИЛЯТОРУ UPS-923A ДЛЯ ПРИЛАДІВ РЕЄСТРАЦІЇ α-, β- ТА ЗМІШАНИХ α-β-ВИПРОМІНЕНЬ

Вступ. Пошук технічних рішень зі створення ефективних детекторів α- та β-випромінень є світовою тенденцією в галузі радіаційного контролю.

Проблематика. Поліпшити характеристики детекторів α- та β-частинок можна через використання матеріалів з оптимізованими параметрами, оригінальні конструкції та технологічні прийоми.

Мета. Розробка та створення технологічних основ виготовлення сцинтиляційних детекторів на основі активованого селеніду цинку ZnSe(Al) та пластмасового сцинтилятору (ПС) полістиролу типу UPS-923A для реєстрації α-, β- й α-β-випромінень.

Матеріали й методи. Використано кристал ZnSe(Al), ПС типу UPS-923A на основі полістиролу, поліметилметакрілат (ПММА); застосовано гаряче пресування, тестування параметрів детекторів при опроміненні α- та β-частками методами спектрометрії та спектрофотометрії.

Результати. Відпрацьовано технологічні режими виготовлення: α-детектору на основі тонкого шару дрібнокристалічного сцинтилятору ZnSe(Al), нанесеного на пластину ПММА, що працює у лічильному режимі реєстрації (чутливість > 0,15 імпульс · $с^{-1}$/Бк (імп · $с^{-1}$/Бк) (^{239}Pu)); пластин ПС полістиролу типу UPS-923A заданої товщини й площі методом гарячого пресування та β-детекторів на їх основі, що працюють у лічильному режимі реєстрації (чутливість > 0,28 імп · $с^{-1}$/Бк (^{90}Sr-^{90}Y)); α-β-детектору на основі тонкого шару дрібнокристалічного ZnSe(Al), нанесеного на пластину ПС полістиролу типу UPS-923A, що працює у лічильному режимі реєстрації (чутливість >0,15 імп · $с^{-1}$/Бк (^{239}Pu) та >0,28 імп · $с^{-1}$/Бк (^{90}Sr-^{90}Y)); α-β-детектору з використанням тонкої монокристалічної пластини сцинтилятору ZnSe(Al) оптично з'єднаної з пластиною ПММА, що працює в спектрометричному режимі реєстрації (детектор одночасно реєструє α- та β- частки з розділенням спектрів з α/β > 0,85, чутливість > 0,3 імп · $с^{-1}$/Бк (^{239}Pu) та > 0,28 імп · $с^{-1}$/Бк (^{90}Sr-^{90}Y)).

Висновки. Розробки детекторів відповідають рівню світових аналогів та забезпечують реєстрацію сигналу як в лічильному, так і у спектрометричному режимі.

Ключові слова: альфа-бета-випромінення, детектор, сцинтилятор, селенід цинку, полістирол, поліметилметакрилат.