COMPOSITE SCINTILLATORS AS NEW TYPE OF A SCINTILLATION MATERIAL

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(Received May 16, 2014)

For radioecological tasks, we have developed a new type of scintillation materials – composite scintillators, consisting of dielectric gel as a base into which granules of scintillating substances were introduced. It has been shown that such material can be created both on the basis of organic and inorganic granules. In the first case, efficient fast neutron detectors can be created, with neutron discrimination on the background of gamma-radiation close to organic single crystals. In the second case, efficient detectors of thermal neutrons could be developed, with variation of the granule size allowing substantial reduction of the effects of background radiation. Separate fragments of the scintillator can be linked together, creating endless planes. A possibility of using bases with higher radiation hardness as compared with standard scintillator bases, as well as using any scintillating substance for granules allows thinking about possible application of such technological approach not only for radioecological tasks (ultra-low fluxes), but also in high energy physics.

PACS: 29.40.Mc, 81.05.Zx

1. INTRODUCTION

Earlier, we proposed a technology for preparation of new type of organic scintillation materials for efficient detection of fast and thermal neutrons. This allowed creation of detectors without limitations imposed on area and shape of the input window [1, 2]. The obtained scintillation materials and detectors on their base are characterized by high universality, simplicity of their use and possibility of their application for wide spectrum of problems related to detection and identification of ionizing radiations. However, not all possibilities of this technological approach have been used. Thus, with a new generation of accelerators, irradiation of detectors in these installations became much stronger. Unique possibilities of composite scintillators make it promising to create scintillation systems with high radiation stability. In this paper, we present our analysis of possible applications of composite scintillators.

2. APPLICATION OF COMPOSITE SCINTILLATORS

2.1. Fast neutron detectors

In a hydrogen-containing organic material, fast neutrons generate recoil protons, with their maximum energy equal the energy of neutrons. Thus, organic scintillators and detectors on their base can be used for spectroscopy of fast neutrons [3].

We have developed composite scintillators that, like organic single crystals, were able to discern particles by the shape of scintillation pulse. The schemes used for separation between the radiation to be detected and the background were presented in our previous works [4, 5, 6, 7]. These schemes allow determining the spectra of recoil protons. Appropriate processing by numerical differentiation allows obtaining, after reconstruction, of the Pu-Be source neutron spectrum. As an example, Fig.1 shows the obtained neutron spectrum of Pu-Be source for a composite scintillator based on stilbene granules with linear dimensions of granules L from 1.7 to 2.0 mm. The scintillator dimensions: diameter 30 mm, height 20 mm.

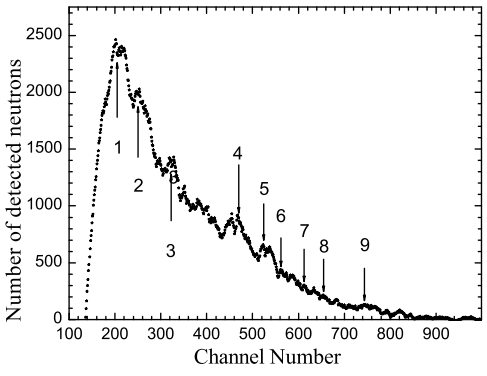

Fig.1. *Reconstructed neutron spectrum of ^{239}Pu-Be source for composite scintillator on the basis of stilbene granules with linear dimensions of granules L from 1.7 to 2.0 mm*

In Fig.1, peaks 1 – 9 correspond to energies 3.1; 4.2; 4.9; 6.4; 6.7; 7.3; 7.9; 8.6 and 9.7 MeV of neutrons emitted by ^{239}Pu-Be source [8].

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When the granule size is optimal, we can obtain spectra identical to those with a single crystal; with grain size smaller than or comparable to the path of recoil protons, the spectrum will be smeared.

One should note that detection selectivity with our samples was confirmed by studies of our colleagues in Poland [9], as well as in China and South Korea [10].

2.2. Thermal neutron detectors

The next step in the use of broad possibilities offered by our technological approach is creation of composite scintillators for detection of thermal neutrons on the basis of Ce:GSO and Ce:GPS.

These substances were chosen because of their uniquely high cross-section of thermal neutron radiation capture by gadolinium nuclides ^{155}Gd, ^{157}Gd, alongside with high content of these nuclides in the natural raw material. This allows using a natural mixture of gadolinium nuclides without its additional enrichment. As a result of thermal neutron capture, gadolinium emits conversion electrons, as well as photons of characteristic X-ray and gamma radiation. When the edge effect is small, gadolinium-based scintillators should exhibit a characteristic peak of 33 keV energy, alongside with another peak at 77 keV, which is the sum of the 33 keV peak of conversion electrons and 44 keV – of X-ray radiation [11].

To obtain thermal neutrons, we used a calibrated paraffin sphere with a ^{239}Pu-Be source inside. The yield of thermal neutrons was 9% of the total fast neutron flux (10^5 neutrons/s). The thermal neutron peak was identified by the "cadmium difference" method. To reduce the number of gamma-radiation photon detection events, a lead shield of 2.5 cm thickness was used.

The efficiency of thermal neutron detection was estimated as follows:

$$\varepsilon_{th} = \frac{N_\Sigma}{t \cdot F_{fast} \cdot \eta_{th} \cdot \frac{S}{4\pi R^2}} \times 100\%, \qquad (1)$$

where N_Σ is the number of events of thermal neutron detection, t is the time of accumulation of the events (spectrum); F_{fast} is the number of fast neutron emitted by the source per second, η_{th}=0,09 is the number of neutrons moderated in the paraffin sphere to thermal energy per one fast neutron, S is the thermal neutron detector area, R is the distance between the source and the detector.

The value ε_γ (Fig.6) was calculated as

$$\varepsilon_\gamma = 1 - exp(-\mu_\rho \rho \xi d), \qquad (2)$$

where μ_ρ is the mass attenuation coefficient of X-ray radiation [12], ρ is the density, ξ is the volume fraction of crystalline granules for Ce:GSO and Ce:GPS (see Chapter 3.3)), d is the scintillator thickness. It was assumed that d was equal to the average grain size in the given fraction. The coefficient ξ accounted for the fact that in the composite scintillator (as distinct from single crystals) the space between the scintillating substance (granules) was filled by non-scintillating gel. The calculations were carried out for gamma-radiation energies of 30 keV, 60 keV, 80 keV, 150 keV, 600 keV, and 8 MeV.

Fig.2 shows, as an example, the calculation results for thermal neutron detection efficiency ε_{th} for a set of composite scintillators Ce:GSO. Calculations were carried out separately for each of three energy ranges. Unfilled signs in Fig.3 show ε_{th} values for Ce:GSO single crystals of 0.39 mm thickness. Data for the conversion electron detection range are noted by squares, for the 77 keV peak – triangles, and for their total signal – circles. The calculated curves of gamma-radiation detection efficiency ε_γ are presented as solid lines.

We have compared the values of thermal neutron detection efficiency calculated by formula (1) using the obtained experimental data (internal counting) and detection efficiency of external gamma-radiation photons ε_γ, which were calculated by formula (2).

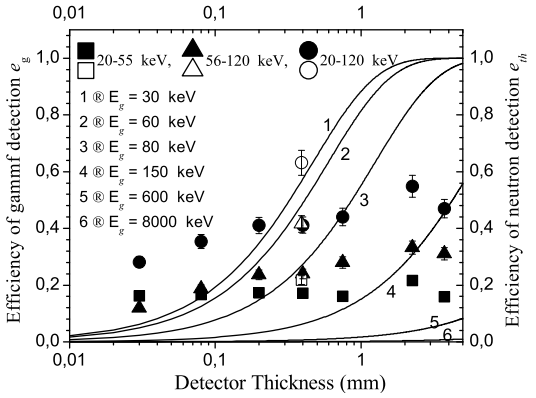

Fig.2. *Thermal neutron detection efficiency ε_{th} for a set of composite scintillators Ce:GSO (signs) and calculated detection efficiency ε_γ of gamma-radiation of energy E_γ (lines) as function of detector thickness d*

As it can be seen from Fig.2, the detection efficiency of 33 keV conversion electrons (squares) is weakly dependent upon the scintillator thickness. This results becomes clear if we take into account that the free path of 33 keV electrons in a given scintillation material is approximately equal to 0.002 mm [13]. Therefore, a single-layer composite gadolinium-containing scintillator with average Ce:GSO or Ce:GPS granule size above 0.002 mm is already an efficient selective detector of conversion electrons of 33 keV energy. The detection efficiency of external gamma-radiation photons in this case becomes lower as compared with detection efficiency of secondary radiations emerging inside the granules, and substantial passive protection from external gamma-radiation photons becomes redundant.

2.3. Combined neutron detectors

Using the broad possibilities of the proposed technological approach, we have developed new combined

composite detectors for separate detection of fast and thermal neutrons on the background of gamma-radiation. These detectors are composed of an organic composite scintillator that detects fast neutrons and a single-layer inorganic composite scintillator that detects thermal neutrons [5].

2.4. Detectors of alpha particles

The proposed technological approach to preparation of composite scintillation materials can also be used for those materials which cannot be grown from the melt as bulky crystals. Fig.3 shows amplitude scintillation spectra of a single-layer composite scintillator on the basis of 1,4-diphenyl-1,3-butadiene excited by alpha-particles of different energies.

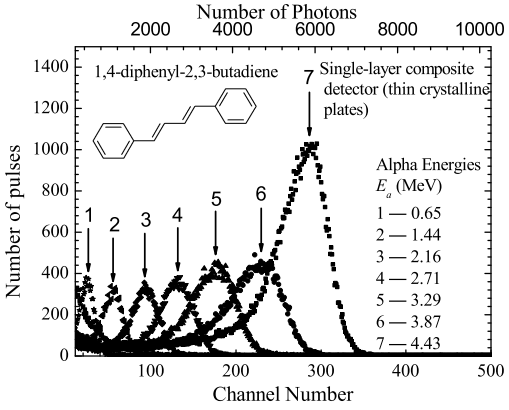

Fig.3. *Amplitude scintillation spectra of single-layer composite scintillator on the basis of 1,4-diphenyl-1,3-butadiene in detection of alpha-particles of energies 0.65 MeV, 1.44 MeV, 2.16 MeV, 2.71 MeV, 3.29 MeV, 3.87 MeV and 4.43 MeV (^{239}Pu)*

Alpha-particles of different energies were obtained by their moderation in air. The energy of alpha-particles passed through the air layer of thickness h was determined in the following way. Knowing the alpha-particle path in air r, we find the residual path $\Delta = (r–h)$, and then, using literature data [14], we determine the energy corresponding to the residual path Δ.

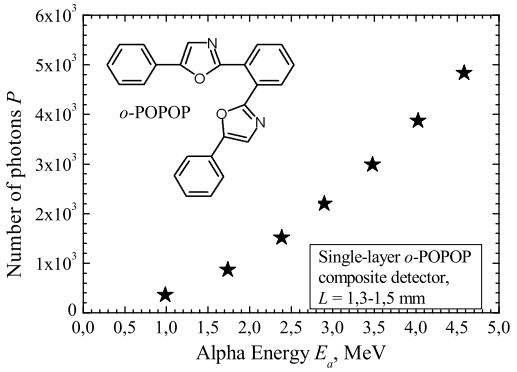

Fig.4. *small Scintillation signal as function of excitation energy (alpha-excitation, scintillators based on o-POPOP)*

Amplitude scintillation spectra of scintillators based on *o*-POPOP, stilbene and 1,4-diphenyl-1,3-butadiene obtained under irradiation by alpha-particles of different energies were rather similar and showed no peculiar features.

Figs.4 and 5 show scintillation signals from composite scintillators based on *o*-POPOP and 1,4-diphenyl-1,3-butadien as function of excitation energy for the case of alpha particles.

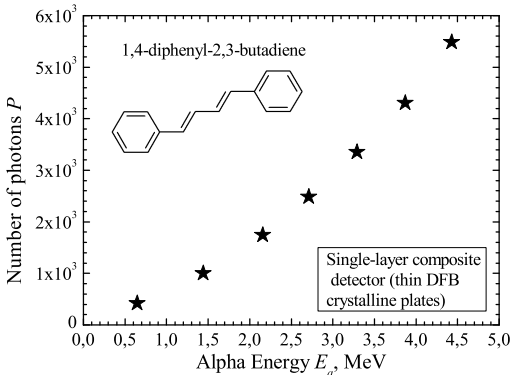

Fig.5. *Scintillation signal as function of excitation energy (alpha-excitation, scintillators based on 1,4-diphenyl-1,3-buradiene)*

3. SAMPLE PREPARATION TECHNOLOGY FEATURES

3.1. Light transmission in the systems studied

Composite scintillators are distinguished, as compared to conventional systems, by their universality and much broader application possibilities. Properties of known scintillators are generally characterized by two main groups of parameters: 1–volume (accounting for shape); 2–chemical composition. In the case of composite scintillators, the third group of parameters becomes important, related to the size of scintillation granules. Therefore for short-range radiations, accounting for transparence features of these systems, thin-layered samples shouls be developed. For penetrating radiations, such as fast neutrons, optimal number of layers should be determined. This number should be sufficiently high to ensure efficient interaction and sufficiently low to ensure light output from the scintillator. Thus, for detection of fast neutrons the optimum thickness was determined as 20 mm (see Chapter 2.1), while for thermal neutrons – single-layered systems with granule size of 30-100 microns. In this respect, it is important to study the transparence characteristics of composite scintillators as function of thickness.

Since light propagation in such systems is of diffuse character, it is reasonable to study transparence as function of average granule size at different wavelengths. This is very important for development of layered systems, where radiation detection efficiency is directly dependent on light transmis-

sion through the scintillator. Measurements of optical transmission for single-layered and multi-layered stilbene-based samples were carried out using a Shimadzu 2450 spectrophotometer with an integrating sphere. The spectral measurement range was from 300 to 700 nm. The results obtained show that for composite scintillators based on crystalline granules of stilbene there is strong light absorption at 360 nm, while the material was practically transparent at 700 nm. The intensity of light transmitted through the composite scintillator is decreased with higher thickness, which is related to an increase in the number of layers of light-scattering granules.

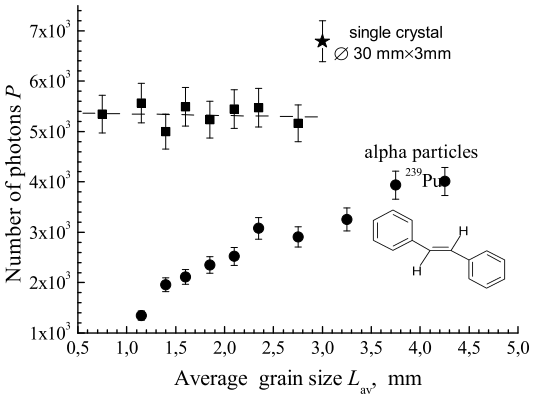

Fig.6. *Number of light photons as function of the average size of crystalline stilbene granules for multi-layered composite scintillators ∅ 30 mm × 20 mm (circles) and single-layered composite scintillators ∅ 30 mm (squares)*

The transparence dependence on the average granule size is of the same character as the similar dependence of the number of photons. For single-layer systems practically no effect of the grain size is observed, while for multi-layered systems smaller granule size, which means larger contribution from the scattering boundaries, both scintillation signal and transmission are decreased, which should be accounted for in development of multi-layered scintillators [8, 15, 16].

3.2. Detectors with unlimited size of the input window

The main advantage of composite scintillators, as compared with structurally perfect organic single crystals, is the possibility to create radiation detectors with no limitations on the input window size and shape. For scintillators of large area, a problem emerges due to possible non-uniformity of the light signal obtained in irradiation of different points of the scintillator surface. For our studies, we chose, as an example, composition detectors of diameter 200 mm. This size was considered as sufficient for check-up of uniformity of scintillation characteristics [17]. The scatter of relative light output values measured in different regions of composite scintillator of 200 mm diameter did not exceed 1%, i.e., less than the standard 5% error of light output measurements. Thus, it can be concluded that our proposed chain of technological procedures allows preparation of uniform composite detectors with large output windows. Technological preparation procedures of composite scintillators impose no limitations on the area and shape of the input window; however, such limitations can be due to other factors, e.g., conditions of transportation and assembling of the detector.

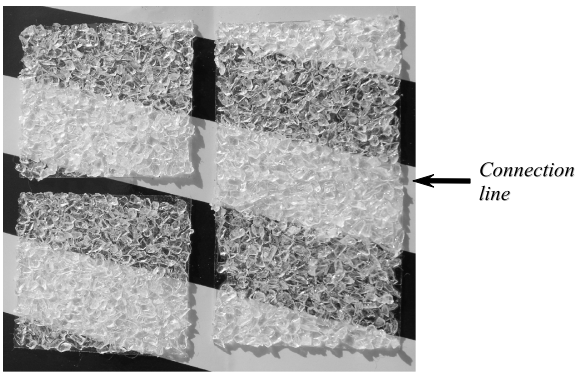

Fig.7. *Connection of separate parts of the composite scintillator*

Fig.7 shows an approach to this problem. First, samples of smaller size are fabricated, which are then glued together by a non-scintillating dielectric gel. The connection line is indicated by arrow [16].

3.3. Technological features of preparation of composite scintillators for different applications.

Our technology allows introduction of granules of any nature into a binding base. Thus, application possibilities of composite scintillators for different tasks are probably the highest as compared with other scintillation systems.

The technological chain for preparation of composite scintillation materials on the basis of crystalline granules of stilbene and *p*-terphenyl (Fig.8, path I) was the first step in creation of new scintillation materials [1, 18, 19]. We have noticed that properties of the obtained composite scintillators are weakly dependent upon structural perfectness of the initial organic crystals. We made an attempt to make the fabrication process of organic scintillators cheaper and simpler, taking stilbene single crystals as example (see Fig.8, path II).

The most energy-consuming and expensive stage of preparation of composite scintillators is the process of growth of structurally perfect single crystals. For single crystal growth, preliminary purification of the raw material is carried out by method of directional crystallization. The primary crystal obtained has rather large number of defects, and such sample cannot be directly used as a bulky single crystalline scintillator. In preparation of crystalline granules by cryogenic fragmentation, the primary crystal is fragmented over the defects into many micro-single crystals.

Calculated values of the local field strength E_{loc} created by the pair of polaron states M_p^- and M_p^+ in anthracene crystal

Technological operations	Sample based on granules of grown single crystal	Sample based on granules of primary crystal
Raw material purification by directional crystallization, hours	168	168
Preparation of ampoule with raw material for growth, hours	7	–
Single crystal growth*, hours	240	–
Fragmentation at low temperature, hours	1.5	1.5
Preparation of scintillator (selection of fractions, introduction into binder and binder polymerization), hours	61	61
Total, hours	477,5	230,5

* For single crystal sample of 20 mm diameter and 10 mm height. With larger volume of required raw material, the difference between the required rime by two methods increases.

The obtained granules were separated into fractions of different sizes, and composite scintillators were fabricated using technological procedures worked out in our studies. I.e., with our approach, we can obtain composite scintillators omitting the stage of growing single crystals of high structural perfectness. Duration of technological operations in preparation of organic scintillators is shown in Table.

Our studies have shown that composite scintillators prepared from crystalline granules of stilbene obtained from the primary crystal (after raw material purification by directional crystallization) have as good scintillation characteristics as composite scintillators prepared from granules obtained from a pregrown single crystal of high structural perfection [18].

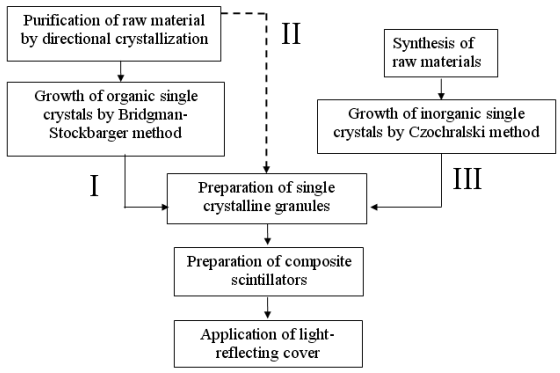

Fig.8. *Flow chart of technological chain for preparation of composite scintillators: I, II – on the basis of crystalline granules of stilbene and p-terphenyl using traditional and modified (i.e., without stage of growing structurally perfect single crystal) technology, respectively; III – on the basis of crystalline granules of gadolinium silicate (Ce:GSO) or pyrosilicate (Ce:GPS) activated by Ce*

Path III in Fig.8 demonstrates technological operations of preparation of composite scintillators on the basis of crystalline granules of gadolinium silicate (Ce:GSO) or pyrosilicate (Ce:GPS) activated by Ce [19]. Such scintillators are efficient detectors of thermal neutrons (see Chapter 2.2).

3.4. Possibility to use composite scintillators under high radiation loads. Prospects of increasing radiation stability of composite scintillators.

With a new generation of accelerators, such as, e.g., LHC CERN (Switzerland), irradiation intensity of detectors used in such installations has substantially increased. Thus, for the end-cap hadronic calorimeter (Hadron Endcap (He)) of CMS detector CMS (LHC CERN) they use scintillation detecting plates (tiles), with some of them located near the beam axis, where particle flux intensity is high, and correspondingly high are the dose loads. After 10 years of operation of CMS-detector, the total dose reaches 10 Mrad at average dose rate 1 Mrad/year or 0.0001 Mrad/hour.

Therefore, there is a constantly growing interest in increasing the radiation stability of scintillators. We have made a first step in this direction. We obtained information on radiation stability of materials that can be used as a base for composite scintillators. These materials are industrially produced gels required for creation of radiation-resistant detectors on their base [20]. This justifies our search in this direction.

4. CONCLUSIONS

Analysis of the results obtained for scintillation characteristics of composite scintillators allows us to make the following conclusions:

1. We have developed composite scintillators composed of crystalline granules introduced into an organosilicon base. The obtained scintillation materials are not hygroscopic, non-combustible, have no technological limitations on the size and shape of the input window. These materials show selectivity to detected signals, good scintillation characteristics, as well as high spatial uniformity of the scintillation signal.

2. A procedure has been developed for connection of separate parts of composite scintillators, which allows fabrication of scintillators with really unlimited area of the input window. This is achieved by connecting separate parts in assembling the scintillator at location of its application.

3. It has been shown that the developed technology can be used not only for traditional organic scintillation

materials, but also for materials which cannot be grown from the melt as bulky crystals.

4. The obtained information on radiation stability of gel dielectric bases allows us to state that the use of such materials for preparation of composite scintillators is one of the promising directions in improvement of characteristics of detecting devices.

ACKNOWLEDGEMENTS

This work was supported by the State Fund for Fundamental Research of Ukraine (project No. F58/06, "The effect of large radiation doses on scintillation and optical properties of novel types of organic detectors").

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КОМПОЗИЦИОННЫЕ СЦИНТИЛЛЯТОРЫ КАК НОВЫЙ ВИД СЦИНТИЛЛЯЦИОННОГО МАТЕРИАЛА

Н. Л. Караваева

Для радиоэкологических задач нами был разработан новый вид сцинтилляционного материала — композиционные сцинтилляторы, состоящие из диэлектрического геля в качестве основы, в которую внедрены гранулы сцинтиллирующего вещества. Показано, что данный материал может создаваться как на основе органических, так и неорганических гранул. Первые позволяют создавать эффективные детекторы быстрых нейтронов со степенью разделения нейтронов на фоне гамма-излучения, близкой к органическим монокристаллам. Вторые явились эффективными детекторами тепловых нейтронов, а варьирование размеров их гранул позволило существенно уменьшить влияние фоновых излучений. Отдельные фрагменты сцинтиллятора можно соединять вместе, создавая бесконечные плоскости. Возможность выбора более радиационно-стойких основ, чем стандартные сцинтилляционные основы, и любых сцинтилляционных веществ для создания гранул позволяет задуматься о возможности использования такого технологического подхода не только для задач радиоэкологии (сверхмалые потоки), но и для задач физики высоких энергий.

КОМПОЗИЦІЙНІ СЦИНТИЛЯТОРИ ЯК НОВИЙ ВИД СЦИНТИЛЯЦІЙНОГО МАТЕРІАЛУ

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Для радіоекологічних задач нами був розроблений новий вид сцинтиляційного матеріалу – композиційні сцинтилятори, що складаються з діелектричного гелю в якості основи, в яку введені гранули сцинтилюючої речовини. Показано, що даний матеріал може створюватися як на основі органічних, так і неорганічних гранул. Перші дозволяють створювати ефективні детектори швидких нейтронів з ступенем поділу нейтронів на фоні гамма- випромінювання, близькою до органічних монокристалів. Другі є ефективними детекторами теплових нейтронів, а варіювання розмірів їх гранул дозволило істотно зменшити вплив фонових випромінювань. Окремі частини сцинтилятора можна з'єднувати разом, створюючи нескінченні площі. Можливість вибору більш радіаційностійких основ, ніж стандартні сцинтиляційні основи, та будь-яких сцинтиляційних речовин для створення гранул дозволяє замислитися про можливість використання такого технологічного підходу не тільки для задач радіоекології (надслабкі потоки), а й для задач фізики високих енергій.