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Large size composite scintillators

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Discussed are the main technological approaches to composite scintillators comprising grains of stilbene and *p*-terphenyl organic single crystals as the fast neutron detectors and Ce:GSO and Ce:GPS inorganic single crystals as thermal neutron detectors. The scintillation characteristics of the composite materials obtained have been studied and compared to those of reference single crystals. The proposed composite scintillation materials are non-hygroscopic efficient detectors of neutrons that, in contrast to the reference single crystal detectors are free from technological restrictions on the input window shape and size.

Обсуждаются основные технологические приемы создания композиционных сцинтилляторов на основе кристаллических зерен органических монокристаллов стильбена и *n*-терфенила как детекторов быстрых нейтронов и неорганических монокристаллов Ce:GSO и Ce:GPS — детекторов тепловых нейтронов. Проведены исследования и сравнительный анализ сцинтилляционных характеристик полученных композиционных материалов и эталонных монокристаллов. Предложенные композиционные сцинтилляционные материалы — это негигроскопичные, эффективные детекторы нейтронов, которым, в отличие от эталонных монокристаллических детекторов, не свойственны технологические ограничения формы и размеров входного окна.

1. Introduction

The problem of spectrometry of a low intensity fluxes of fast neutrons and α-particles is of a great importance in modern ecological, geological, biological, medical fields, custom survey, etc. Such radiations are among the most hazardous for human organism. The hazard is characterized by the radiation-weighting factor. The radiation-weighting factor w_R is the value expressing the long-term risk (primarily cancer, leukemia, heavy pulmonary allergies, etc.) of a low-level chronic exposure. It depends upon the radiation type and other factors. For example, for γ-radiation photons $w_R = 1$, while for fast neutrons with energy $E_n \leq 2$ MeV, the $w_R = 20$. Similar high w_R values characterize alpha particles. For thermal neutrons, $w_R = 5$ [1]. In natural conditions there are numerous hydrogen-contain media where the fast neutrons are slowed down and transformed into thermal neutrons, the detection of the latter allowing to estimate the flux of primary neutrons. In detection of very low activity ionizing radiation, large size detectors are necessary where an increased detection efficiency is attained due to increased spatial angle of detection. In addition, such detectors should provide the spectral composition of luminescence making it possible to use the commercial (and thus inexpensive) photodetectors. The materials should provide the separate detection of neutrons and background γ-radiation, and show a sufficiently high light yield.

In this work, described are two preparation procedures of composite scintillators for (i) fast neutrons basing on grains of stilbene and *p*-terphenyl crystals and (ii) thermal neutrons basing on grains of gadolinium silicate $Ce:Gd_2SiO_5$ (Ce:GSO) and pyrosilicate $Ce:Gd_2Si_2O_7$ (Ce:GPS). This work is actual, because the maximum size of the above organic single crystals grown

from the melt is 150 mm and that of gadolinium pyrosilicate single crystals, ten millimeters. The Ce:GSO single crystals were first proposed for detection thermal neutrons in [2], and Ce:GPS single crystals, in [3].

2. Preparation of composite scintillators

In general, the single crystals were crushed. The grains of different size were obtained by passing the crushed crystals through sieves with different cell sizes. The linear size L of organic grains varied from 0.5 up to 4.5 mm and inorganic grains, from 0.06 up to 1.0 mm. The crystalline grains were incorporated into a two-component polymer matrix. The prepared composite material was placed into an optically transparent organic glass container.

Composite scintillations for fast neutron detection. At first, the size of crystalline grains for composite scintillators intended for the fast neutron detection was pre-estimated. The fast neutrons generate in the organic material the recoil protons with maximum energy equal to that of neutrons. The grain size should be at least equal to the recoil proton path in our material. Therefore, depending on the energy of neutrons to be detected, the scintillators are to be prepared basing on fractions of various sizes (from submillimeter range to several millimeters) [4].

The technology proposed by us included the following stages:

1. Growing of single crystals with high structural perfection using the Bridgman-Stokbarger technique.
2. Preparation of grains and separation thereof into fractions of various sizes.
3. Preparation of organosilicon polymeric matrix.
4. Introduction of the obtained grain fractions into the matrix.
5. The matrix introduction into the shaping container.
6. Evacuation and holding of the sample during 48 h.
7. Application of the reflective coating onto the container.

The crystalline grains of stilbene and *p*-terphenyl were obtained by crushing of a structurally perfect single crystal grown from the melt by the technique described in [5]. The grown single crystals were crushed in the liquid nitrogen medium. That crushing technique reduces the loadings influencing negatively the structural perfection of grains and their scintillation properties. The grains so obtained were dried up in darkness at a room temperature for at least 12 h. Thereafter, the crushed material was sieved through calibrated sieves separating the grains into fractions of different size [6, 7].

During the researches [6, 7], the light yield value of stilbene/*p*-terphenyl composite scintillators was established to increase with the grain size L attaining a maximum at $L \sim 1.5$ mm, and then remains unchanged as L increases. Therefore, the grains of 1.0 to 2.0 linear dimension were used in the further study of organic composite scintillators. This provided the optimum values of the light yield and efficiency of fast neutron detection. As the polymer matrix, the two-component adhesive composition Sylgard-527 (Dow Corning Corporation [USA]) was used due to its inertness, non-hygroscopicity, and maximum transparency in the stilbene luminescence range.

To determine the optimum ratio of stilbene grains and organosilicon matrix, a series of composite scintillators was prepared and examined. The experimental data show that the optimum amount of stilbene grains in the immersion medium is at least 70 % of the matrix mass [6, 7]. To shape the scintillator, it is necessary to polymerize the matrix in a shaping case. While preparing the container, the following requirements were taken into account. First, the scintillator height should provide an efficient detection of fast neutrons of several MeV energy and the scintillator must remain sufficiently transparent to its intrinsic emission. The container was been made of organic glass. The container surface (except a target window of scintillator detector) was coated with the Tetratek diffuse reflector. These processing methods have been used to prepare large-diameter composite detectors, (∅200 mm×20 mm) composite detector based on stilbene grains and on activated *p*-terphenyl grains. Those were used in particular to research the light signal inhomogeneity in various points of the scintillator surface.

Composite scintillators for thermal neutron detection. The free path of thermal neutrons in a substance is much shorter that of moderated energy γ photons. Therefore, on the one hand, the inorganic detector thickness should be sufficient to record thermal neutrons and the secondary radiations generated by those. On the other hand, the same detector should be thin enough to reduce probability of detecting background γ radiation. For such thin scin-

tillator (but not limited in diameter), the efficiency of thermal neutron detection will exceed that of background radiations.

To prepare the composite material, the grains of Ce doped inorganic crystals Gd_2SiO_5 and $Gd_2Si_2O_7$ were introduced into a polymer matrix [8]. To prepare the crystal grains, Ce:GSO and Ce:GPS crystals were mechanically ground. Thus, the Ce concentration in the grains was the same as in the reference single crystals. The obtained grains were sieved through calibrated sieves to obtain the grains with necessary size L. The grains of the chosen fractions were introduced into non-luminescent Sylgard-527 silicone material. Then the composition was applied onto an optically transparent non-luminescent organic glass plate. The polymerization process for Sylgard-527 takes 48 h [9].

To study further the single-layer composite scintillators on the base of Ce:GSO and Ce:GPS grains, 5 grain size ranges with L values, namely, <0.06 mm, 0.06–0.1, 0.1–0.3, 0.3–0.5, 0.5–1.0 mm. Additionally, 0.5–1.0 mm grain fraction was used to prepare multi-layer (3 or 5 layers) Ce:GSO composite scintillators. To determine the optimum composition of the inorganic scintillator, samples of 514 mm^2 area were used. To study the light signal inhomogeneity in various points of the scintillator surface, large diameter detectors (∅200 mm) were made.

3. Results

In what follows, presented are the experimental results showing scintillator properties of composite detectors obtained using the stilbene crystals grains as well as Ce:GSO and Ce:GPS ones.

The stilbene based composite scintillators as fast neutron detectors. To estimate the relative recording efficiency of fast neutrons, the recoil proton spectra were obtained for the stilbene based composite detector (∅50mm×12 mm) and for a reference stilbene single crystal (∅50mm×9 mm). For the chosen detectors, the scintillator volume was essentially the same, taken into account the container dimensions of the composite detector. The technique of particle identification basing on the scintillation pulse shape [10] has allowed to discriminate the fast neutron scintillations from a ^{239}Pu–Be source against the background γ radiation. Fig. 1 shows the spectra of recoil protons for a reference stilbene single crystal (curve *1*) and for stilbene composite scintillator (curve *2*) irradiated with a ^{239}Pu–Be source. Both spectra were collected for an identical time interval. In this case, the ratio of the collected pulse number in the recoil proton spectrum for the composite scintillator (curve *2*) to the pulse number in a similar spectrum for stilbene single crystal (curve *1*) characterizes the relative detection efficiency of fast neutrons from the ^{239}Pu–Be source for the composite scintillator. It has been found that for composite stilbene scintillators it makes 50 to 55 % of the fast neutron detection efficiency for organic single crystals of similar size.

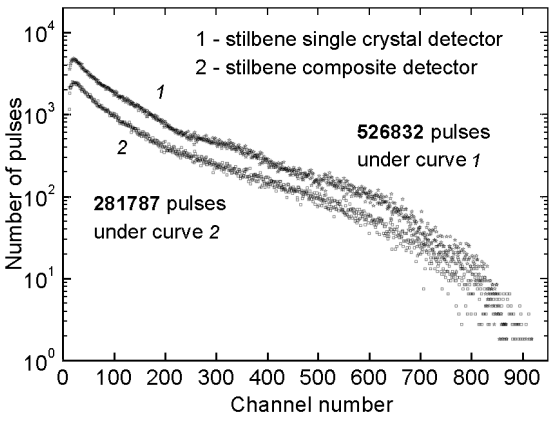

Fig. 1. Spectra of recoil protons for a reference stilbene single crystal (*1*) and for stilbene composite scintillator (*2*) under irradiation with a ^{239}Pu–Be source. Both spectra collected for an identical time interval.

The light yield inhomogeneity was estimated for the obtained large diameter composite scintillators. The light yield values were measured according to the standard procedure using a ^{137}Cs source. To estimate the light yield inhomogeneity, the ΔLY parameter was used:

$$\Delta LY_i = \left| \frac{I_0 - I_i}{I_0 + I_i} \right| \times 100\%, \qquad (1)$$

where I_0 is the of light yield value in the scintillator center; and I_i, the values for measuring point i at the scintillator surface [7]. The analysis has shown that the maximum yield inhomogeneity calculated for different sites of the large-diameter organic composite scintillators is considerably lower than the light yield determination that makes 5 %.

The Ce:GSO and Ce:GPS based composite scintillators. As a result of a thermal neutron capture, gadolinium radiates conver-

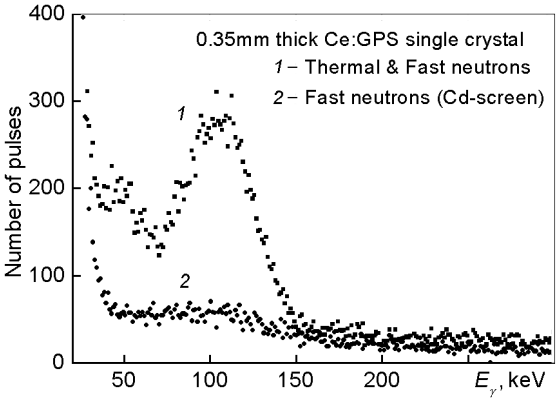

Fig. 2. Amplitude spectra of 0.35 mm thick Cs:GPS single crystal irradiated with fast and thermal neutrons (*1*) and with fast neutrons only (*2*).

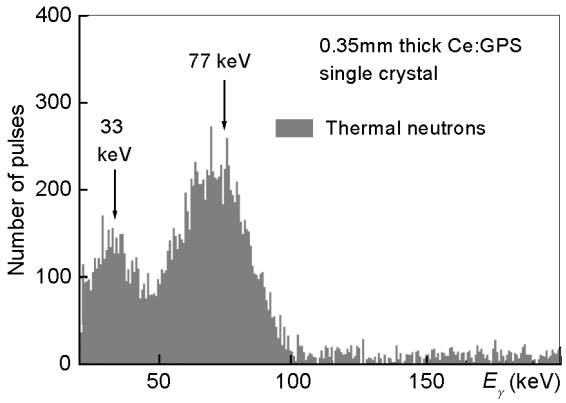

Fig. 3. Amplitude spectrum of 0.35 mm thick Ce:GPS single crystal excited by thermal neutrons.

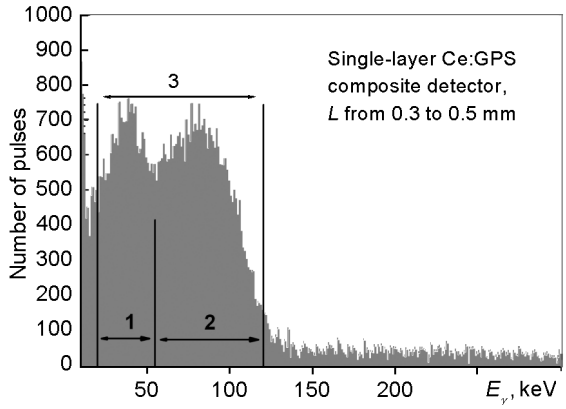

Fig. 4. Amplitude spectrum of single-layer Ce:GPS composite detector with grain size L 0.3–0.5 mm excited by thermal neutrons.

sion electrons, characteristic X-ray and γ radiation. When the edge effect influence is low, for the gadolinium based scintillators must show a characteristic peak with 33 keV energy and along with it, a 77 keV peak being the sum of the conversion electron peak (33 keV) and X-ray radiation (44 keV) [3]. The thickness of single-layer composite scintillators was determined as the average size of the corresponding fraction of grains, and for three-layer and five-layer Ce:GSO composite scintillators, as triple and fivefold value of the average fraction size with L 0.5 to 1 mm, that is, 0.75 mm. Obtained were the thermal neutron scintillation spectra for reference detectors on the basis of Ce:GSO and Ce:GPS single crystals and for composite detectors containing the grains of Ce:GSO and Ce:GPS crystals.

Figs. 2–3 demonstrate the measurement procedure of the thermal neutron scintillation spectra for Gd-based detectors taking a Cs:GPS single crystal as an example. Curve *1* in Fig. 2 is the total scintillation spectrum of a Cs:GPS single crystal irradiated simultaneously with thermal and fast neutrons from a ^{239}Pu–Be source placed inside a paraffin sphere. Curve *2* represents the scintillation spectrum measured for the same experimental geometry but with a cadmium screen placed between the source and the scintillator. The accumulation time was the same for both spectra. The spectrum presented in Fig. 3 is obtained by subtraction of the fast neutron scintillation spectrum (curve *2* in Fig. 2) from the total scintillation spectrum (curve 1 in Fig. 2). Fig. 4 presents as an example the thermal neutron peak amplitude spectrum for a single-layer Ce:GPS composite detector with grain size L 0.3–0.5 mm.

The obtained results hav shown that the intensity ratio of 33 keV and 77 keV scintillation peaks increases as the grain size of a composite scintillator is diminished. The best neutron-to-gamma ratio was obtained for the single-layer composite scintillators. For Ce:GSO composite scintillators with 3 or 5 layers of 0.5–1 mm grains, the peak of 33 keV was not observed. For composite scintillators, the resolution of both lines is worse than for the reference single crystal. To characterize the properties of the scintillators, we have introduced three arbitrary energy ranges (see Fig. 4), namely, 20 to 55 keV (the amplitude range of the scintillation peak generated by 33 keV conversion electrons), 55 to 120 keV (the range of total 77 keV peak), and 20 to 120 keV (the whole scintillation spectrum).

Figs. 5 and 6 present the calculation results of thermal neutron detection effi-

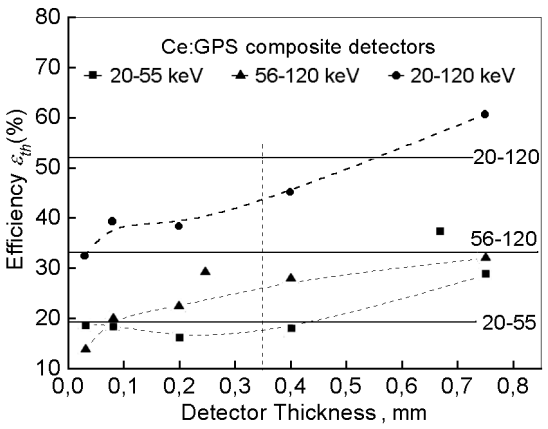

Fig. 5. Thermal neutron detection efficiency ε_{th} for a series of Ce:GPS composite scintillators calculated for different energy ranges of thermal neutron detection. Horizontal solid lines present the ε_{th} values for 0.35 mm thick Cs:GPS single crystal, the vertical line indicates the thickness of the reference single crystal.

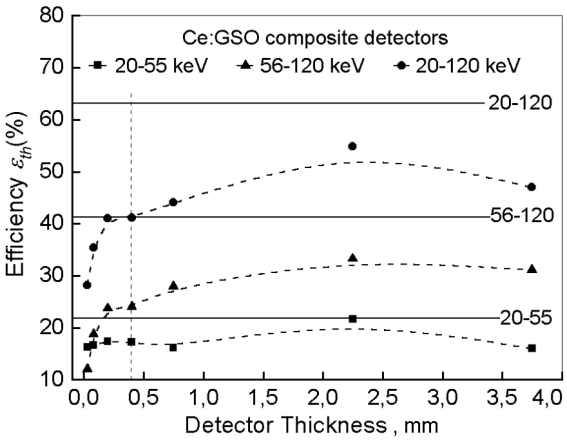

Fig. 6. Thermal neutron detection efficiency ε_{th} for a series of Ce:GSO composite scintillators calculated for different energy ranges of thermal neutron detection. Horizontal solid lines present the ε_{th} values for 0.39 mm thick Cs:GSO single crystal, the vertical line indicates the thickness of the reference single crystal.

ciency ε_{th} for a series of Ce:GSO composite scintillators and for a series of Ce:GPS ones. Direct horizontal solid lines are the thermal neutron detection efficiency for Cs:GPS single crystals (0.35 mm thick) and 0.39 mm thick Cs:GSO ones. Vertical lines in Figs. 5 and 6 indicates the thickness of the reference single crystals.

The efficiency of thermal neutron detection ε_{th} we estimated as follows:

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

where N_Σ is the number of thermal neutron detection events; t, the accumulation time of the events; F_{fast}, the flux of fast neutrons from the source; $\eta_{th} = 0.09$, the number of thermal neutrons obtained due to deceleration of fast neutrons in the paraffin sphere per one fast neutron; S, the detector area; R, the source-to-detector distance.

It is seen from Figs. 5 and 6 that the detection efficiency of 33 keV conversion electrons depends weakly on the detector thickness. The very thin scintillation layer (L less than 0.06 mm) is enough to detect this radiation effectively. Such a result becomes clear when taking into account that the free path of 33 keV electrons in such scintillation materials is about 0.002 mm [10]. Therefore, a single layer of the Gd-containing composite scintillator with grains of the average size not exceeding 0.1 mm is sufficient for selective detection of 33 keV conversion electrons. The detection efficiency of thermal neutrons in the energy range 20–55 keV makes ~20 %. A Ce:GPS containing composite scintillator has a higher light yield than Ce:GSO one, therefore, the former selects the scintillation signal from 33 keV conversion electrons better than the latter. A single-layer composite scintillator on the base of Ce:GSO crystal grains (or on the base of Ce:GPS grains of larger size) is good to detect the total scintillation signal from 44 keV X-ray radiation and 33 keV conversion electrons. The background signal increases with increasing grain size L (see Fig. 4). Therefore, the average size of the grains has not to be large. Figs. 5 and 6 show that for the 20 to 120 keV energy range, the thermal neutron detection efficiency makes 40 to 50 %. Our researches also have shown that detection efficiency of background γ radiation inreases sharply with increasing grain size, or a thickness of a Cd-containing inorganic scintillator. Note that the detection efficiency of γ photons with energies $E_\gamma > 120$ keV for a scintillator containing grains with the average size $L \leq 0.5$ mm is negligibly small. Therefore, by our estimations, the average grain size of the single-layer Ce:GSO or Ce:GPS detector should be no more than 0.5 mm.

Let $\varepsilon_{th}{}^{S}$ and $\varepsilon_{th}{}^{C}$ denote the thermal neutron detection efficiency values obtained for a singe crystal and a composite scintillator with the input window area S_S and S_C, respectively. The ratio between the numbers of thermal neutrons detected by the composite scintillator to those detected by the single crystal, at the same neutron flux is equal to:

$$\zeta_{th} = \frac{\varepsilon_{th}^{C} S_C}{\varepsilon_{th}^{S} S_S} = \alpha \frac{S_C}{S_S}. \quad (3)$$

The α value in (3) is a constant defined by the properties of the specific single crystal sample and the composite scintillator that is made from the grains of this crystal. In contrast to S_S, the S_C value has no technological limitations. Therefore, if $S_S{}^{max}$ is the largest possible input window area for the single crystal, then a composite scintillator with $S_C{}^{0}$ can be chosen for which

$$\zeta_{th}^{0} = \alpha \frac{S_C^0}{S_S^{\max}} = 1. \quad (4)$$

It means that all composite scintillators having the same $\varepsilon_{th}{}^{C}$ and $S_C > S_C{}^{0}$ are more effective detectors of thermal neutrons than the single crystal used to prepare the composites. Figs. 5 and 6 show that for single-layer composite scintillators with grain size L similar to the thickness of the single crystal show in the 20–55 keV energy range the $\varepsilon_{th}{}^{S}$ and $\varepsilon_{th}{}^{C}$ values very close together (i.e. α is about 0.7 and 0.8 for composite scintillators based on the grains of a Cs:GSO single crystal and a Cs:GPS single crystal, respectively). For the energy range 20–120 keV, α is about 0.8 and 0.9 for composite scintillators based on the grains of a Cs:GSO single crystal and a Cs:GPS single crystal, respectively. In accordance to (1), the ε_{th} values are independent of S_C/S_S ratio. So, even for composite scintillators with $S_C > 2S_S{}^{max}$ (where e.g. for Ce:GPS crystals $S_S{}^{max}$ is about 1 cm^2) the number of detected events in time unit will be larger than for single crystal. This value grows with S_C increase. The measurement results for the composite detector of 200 mm diameter prepared on the basis of Cs:GSO crystal grains have shown that the maximum light yield inhomogeneity ΔLY (1) calculated for different sites of such detector is comparable the light yield measurement error (about 5 %).

4. Conclusion

Considering the study results of scintillation characteristics for organic and inorganic composite scintillators, the following conclusions can be drawn. The preparation technology of organic composite scintillators as fast neutron detectors as well as of new inorganic composite scintillators as detectors of thermal neutrons. For stilbene composite scintillators, the fast neutron detection efficiency makes 50 to 55 % with respect to organic single crystals of the similar size. The single-layer composite scintillators with the Ce:GSO or Ce:GPS crystal grain size $L \leq 0.1$ mm provides detection of thermal neutrons by registration of scintillation pulses of conversion electrons with 33 keV energy (the energy range 20 to 55 keV) at efficiency ε_{th} ~ 20 %. At simultaneous registration of scintillation pulses of conversion electrons (33 keV) and the total signal (77 keV), the detection efficiency of thermal neutrons ε_{th} makes 40 to 50 % in the 20 to 120 keV energy range. The composite scintillations are non-hygroscopic efficient detectors of neutrons free of technological restrictions for the input window shape and area, that allows to obtain more effective neutron detectors in comparison with single crystals scintillators.

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Композиційні сцинтилятори великих розмірів

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Обговорюються основні технологічні прийоми виготовлення композиційних сцинтиляторів на основі кристалічних зерен органічних монокристалів стильбену та *n*-терфенілу як детекторів швидких нейтронів і неорганічних монокристалів Ce:GSO та Ce:GPS як детекторів теплових нейтронів. Проведено дослідження та порівняльний аналіз сцинтиляційних характеристик отриманих композиційних матеріалів та еталонних монокристалів. Запропоновані композиційні сцинтиляційні матеріали — це негігроскопічні ефективні детектори нейтронів, які, на відміну від еталонних монокристалічних детекторів, не мають технологічних обмежень щодо форми та розмірів вхідного вікна.