RADIATION-RESISTANT COMPOSITE SCINTILLATORS BASED ON INORGANIC CRYSTALS GSO:Ce, GPS:Ce and Al_2O_3:Ti

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Radiation-resistant composite scintillators based on grains of inorganic single crystals Gd_2SiO_5:Ce, $Gd_2Si_2O_7$:Ce and Al_2O_3:Ti^{3+} were studied. To obtain the composite scintillator the grains were introduced into a dielectric silicone gel composition Sylgard-184. The samples were irradiated at the KIPT 10 MeV electron Linac by 9.2 MeV beam electrons (at the dose rate of 1500 Mrad/hr) and by bremsstrahlung photons (at the rate of 230 krad/hr). The values of luminous transmittance and scintillation light outputs of a scintillator were measured before and after irradiation. The paper presents and analyzes the results obtained for scintillators exposed to doses about 170 Mrad and 300 Mrad.

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1. INTRODUCTION

Experiments which are planned or being carried out at the new-generation high luminosity particle and heavy-ion accelerators (such as the LHC at CERN) are featured by the exposure of the detectors and subdetectors (trackers, calorimeters, etc.) to a high level of radiation doses. One of the examples is the CMS experiment at the LHC. For the first 10 years of LHC operation, the most "critical" (closest to the interaction point) zones of the CMS endcap hadron calorimeter are expected to get the dose D up to 10 Mrad at the rate of about 0.1 krad/hr. Furthermore, there are plans of gradual increase of the LHC luminosity by the order of the magnitude in the future. The maximum dose accumulated in the CMS HE "critical" zones upon the LHC ultimate shutdown is estimated as 30 Mrad [1]. Therefore, a development of materials for radiation detectors with high radiation resistance becomes an important issue.

A luminescent material is regarded as radiation-resistant up to the dose of radiation $D = D_F$, for which its luminescence characteristic changes on a half of it initial value (i.e. for value that is true for $D = 0$) (see p.205 of the monograph [2]). On the first step of the above-mentioned problem solution (i.e. the problem of the development of a luminescent radiation-resistant material) it is necessity to find a radiation-resistant material for the basis component of such a composition (or according to Birks terminology (see book [2], p.55) we have to use not basis component but "main constituent or solvent"). Our previous paper [3] answered this question. We have investigated the luminous transmittance T as a function of an integrated radiation dose D for commercially available silicone dielectric gels. It was namely SKTN-med (20 Pa), SKTN-med (100 Pa), Sylgard-184, Sylgard-186, Silgard-527, and SUREL-SL-1 [3], [4]. The samples were irradiated by 9.2 MeV beam electrons (at the dose rate of 1500 Mrad/hr) and by bremsstrahlung photons (at the rate of 230 krad/hr) at the 10 MeV electron Linac of the NSC Kharkov Institute of Physics and Technology (KIPT). The study was performed up to integrated radiation doses about 90 Mrad. It showed that the T-values, practically, did not change with increase in the radiation dose D. For the wavelengths of the light λ longer than 400 nm the reduction of the T-values did not exceed measurement error (namely 5%). Some insignificant increase in luminous transmittance T for some gels (SKTN-med (20 Pa), SKTN-med (100 Pa), Silgard-527, and

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SUREL-SL-1) was obtained. It can be caused by the effect of a partial solid-state recrystallization arising from the action of ionizing radiation. On the other side, the improvement of structure perfection can only lead to reduction of light photons scattering and as the result to increase in a transparency of a material [5]. It should be also stated that the dielectric gels can be used in the wide range of working temperatures, they do not chemically react with the materials used in a scintillation technique, they are non-hygroscopic, and they possess high transparency in the range of a luminescence of majority of scintillation materials [3].

2. EXPERIMENTAL DETAILS

2.1. Preparation of composite scintillators

In this work we used the dielectric polydimethylsiloxane gel Sylgard-184 as the base material for composite scintillators [4].

To obtain a composite scintillator we mechanically grind up a single crystal boule and after that we use a set of sieves with the sized meshes to select of the grains with required sizes. The grains were introduced in dielectric gel according to the following technique. Firstly, we introduced them in the first component of the gel. After adjunction of the second component the gel composition is carefully mixed, and after that, it is introduced into a forming container, in which it left up to it complete polymerization. As the result, the scintillator is obtained and can be taken from the forming container. We investigated single-layer and multi-layer composite scintillators. For the former case the grains are posed in one layer inside the gel, and the thickness of such a scintillator was determined by the average grain size. In the latter case the grains are posed in more than one layer inside the gel, and some of them spontaneously dispose between the layers.

2.2. Irradiation of the samples

As in our earlier studies [3], we irradiated the samples at the KIPT 10 MeV electron Linac. During the irradiation runs, which took place at the room temperature, the dose rate was practically uniform over the sample surfaces. Inhomogeneity of irradiation of the samples did not exceed 5%. The highest dose rate, (1500±5) Mrad/hr, was provided, when the samples were irradiated directly by 9.2 MeV beam electrons. Other samples were subjected to irradiation by bremsstrahlung photons at the considerably lower rate of (0.23±0.01) Mrad/hr. The samples were, consistently (by one sample of each type), exposed to radiation until they accumulated the necessary integrated radiation dose. The dose was measured by Harwell Red 4034 plastic dosimeters to an accuracy of ±10%. The details are outlined in [3]. One of the samples in each series was taken as a benchmark scintillator and was not exposed to radiation.

2.3. Measurements of scintillation light yield

The set of gamma sources allowed us to calibrate the energy scale of the measuring setup. We used a single crystal of Gd_2SiO_5:Ce (GSO:Ce), $Gd_2Si_2O_7$:Ce (GPS:Ce) and Al_2O_3:Ti^{3+} as the reference for corresponding composite scintillators. For all the scintillators the relative light output was obtained as the result of measurements of scintillation amplitude spectra. For GSO:Ce and GPS:Ce the scintillators were excited by photons of gamma-radiation from radionuclide sources ^{137}Cs (33 keV) and ^{251}Am (59.6 keV) (see details in [6]). For single crystal and composite scintillators based on the grains of Al_2O_3:Ti^{3+} the scintillators were excited by alpha-particles from ^{239}Pu (5.15 MeV). Measurements were run before and after irradiation.

2.4. Measurements of transmittance

The measurements of the luminous transmittance T in the range from 300 to 700 nm were performed by Shimadzu-2450 spectrophotometer with the integrating sphere. The comparison channel remained blank and the light flux inside it was calibrated to be the same as the light flux falling on a sample in measuring channel. The inaccuracy of the calibration was limited by 0.5%. The value of T was calculated as follows:

$$T = (I/I_0) \cdot 100\%, \qquad (1)$$

where I_0 is the light flux in comparison channel, I is the light flux, which has passed through a sample in measuring channel. Actually, the T-value (1) is a relative luminous transmittance, where $T = 100\%$ it is the luminous transmittance of air.

3. RESULTS AND DISCUSSION

3.1. Relative light transmittance

GSO and GPS scintillators

Grain Size of GPS and GSO grains was 0.5...2.0 mm. The luminous transmittances T were measured in the wavelength range from 300 to 700 nm.

Figs. 1 and 2 demonstrate the luminous transmittance T of the composite scintillators based on grains of single crystals GSO as a function of light wavelength λ. Results were obtained for different doses of irradiation. The dose rate was 0.23 Mrad/hour.

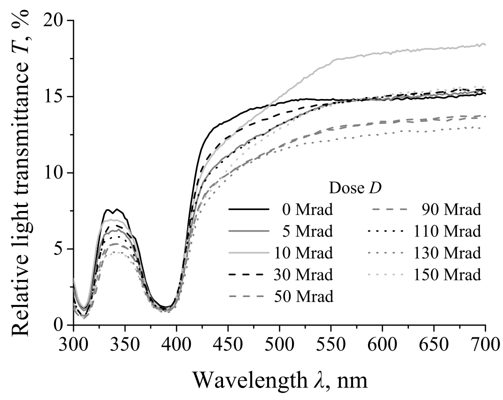

Fig.1. *T versus λ for 4 mm thick composite scintillator based on the grains of a GSO single crystal. The dose rate was 0.23 Mrad/hour*

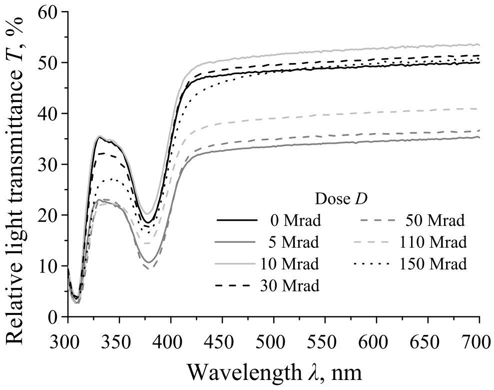

Fig.2. *T versus λ for single-layer composite scintillator based on the grains of a GSO single crystal. The dose rate was 0.23 Mrad/hour*

Figs. 1 and 2 show that the composite scintillators based on GSO at least for $D = 150$ Mrad is radiation resistant, because the T-value for the band of luminescence ($\lambda > 420$ nm [6]) reduce less than to half.

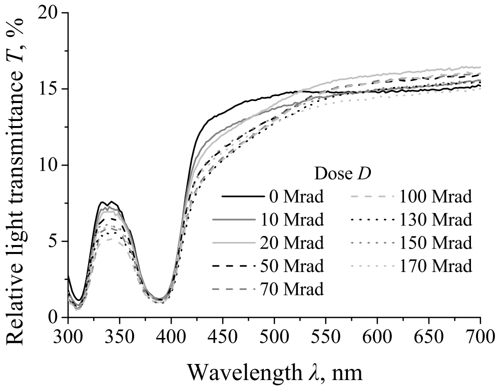

Fig.3. *T versus λ for 4 mm thick composite scintillator based on the grains of a GSO single crystal. The dose rate was 1500 Mrad/hour*

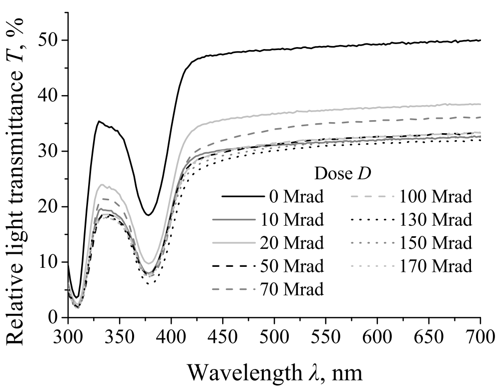

Fig.4. *T versus λ for single-layer composite scintillator based on the grains of a GSO single crystal. The dose rate was 1500 Mrad/hour*

To estimate the influence of dose rate on the luminous transmittance, the composite scintillators were exposure with the dose rate 1500 Mrad/hour. Figs. 3 and 4 demonstrate that for such a case the composite scintillators based on the grains of a GSO single-crystal are radiation-resistant up to $D = 170$ Mrad.

Figs. 5 and 6 shows the T-values obtained for composite scintillators based on the grains of a GPS single crystal with different thickness after irradiation with the dose rate of 0.2 Mrad/hour.

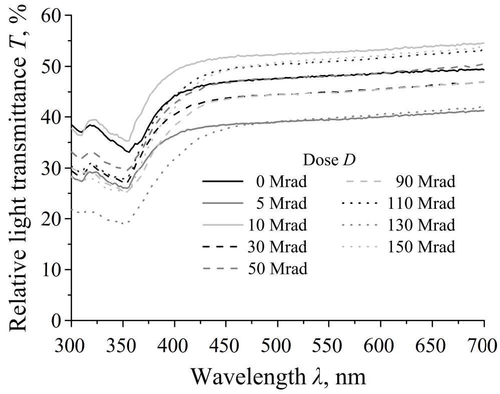

Fig.5. *T versus λ for single-layer composite scintillator based on the grains of a GPS single crystal. The dose rate was 0.23 Mrad/hour*

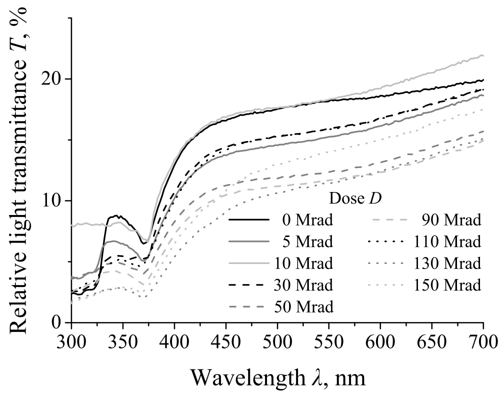

Fig.6. *T versus λ for 4 mm thick composite scintillator based on the grains of a GPS single crystal. The dose rate was 0.23 Mrad/hour*

Figs. 5 and 6 demonstrate that the composite scintillators based on the grains of a GPS single crystal are radiation-resistant up to doses 150 Mrad.

$Al_2O_3:Ti^{3+}$ scintillators

The luminous transmittances T were measured in the wavelength range from 300 to 700 nm.

Figs. 7, 8 and 9 shows the luminous transmission T versus wavelength λ for the composite scintillators based on grains $Al_2O_3:Ti^{3+}$.

Figs. 7, 8 and 9 demonstrate that the composite scintillators based on $Al_2O_3:Ti^{3+}$ up to doses about 100 Mrad (dose rate was 0.23 Mrad/hour) are radiation-resistant because in the spectral band of their luminescence ($\lambda > 600$ nm) the T-values decrease less than 2 times.

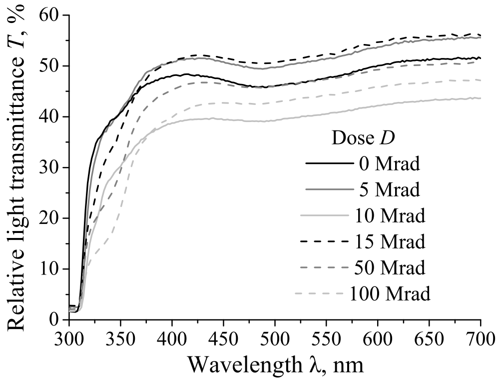

Fig.7. *Luminous transmittance T of the single-layer composite scintillator based on the grains of an Al_2O_3:Ti^{3+} single crystal (grain size is 0.3...0.5 mm). Dose rate was 0.23 Mrad/hour*

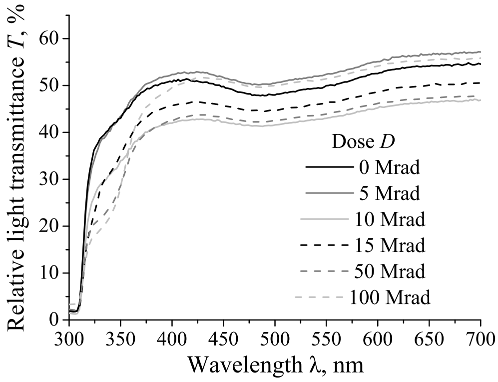

Fig.8. *Luminous transmittance T of the single-layer composite scintillator based on the grains of an Al_2O_3:Ti^{3+} single crystal. Grain size was 0.5...1.0 mm. Dose rate was 0.23 Mrad/hour*

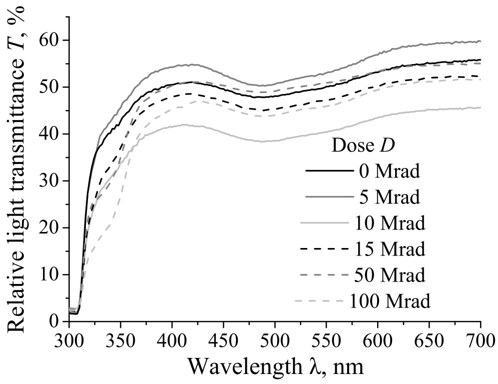

Fig.9. *Luminous transmittance T of the single-layer composite scintillator based on the grains of an Al_2O_3:Ti^{3+} single. Grain size was 1.5...2.0 mm. Dose rate was 0.23 Mrad/hour*

To estimate the influence of dose rate on the luminous transmittance, the composite scintillators were exposure with the dose rate 1500 Mrad/hour.

Figs. 10, 11 and 12 demonstrate that the composite scintillators based on the grains of an Al_2O_3:Ti^{3+} single crystal are radiation-resistant up to the dose of 300 Mrad (dose rate 1500 Mrad/hour) because the transmittance decreases less than 2 times in the spectral band of their luminescence ($\lambda > 600$ nm).

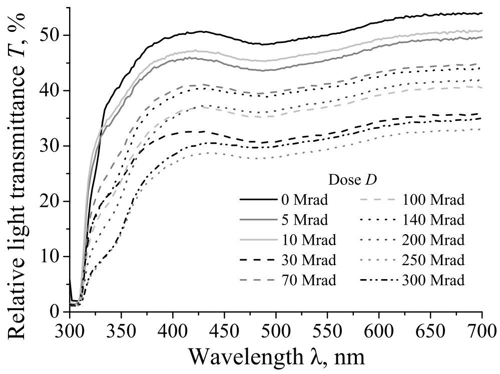

Fig.10. *Luminous transmittance T of the single-layer composite scintillator based on the grains of an Al_2O_3:Ti^{3+} single crystal. Grain size was 0.3...0.5 mm. Dose rate was 1500 Mrad/hour*

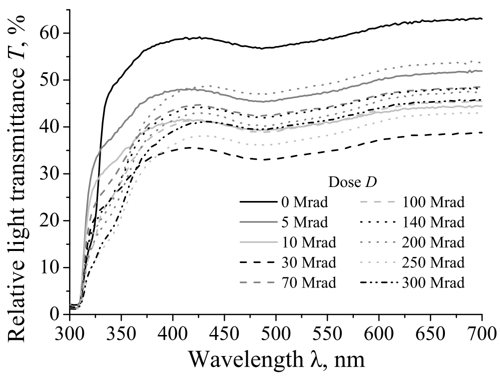

Fig.11. *Luminous transmittance T of the single-layer composite scintillator based on the grains of an Al_2O_3:Ti^{3+} single crystal Grain size was 0.5...1.0 mm. Dose rate was 1500 Mrad/hour*

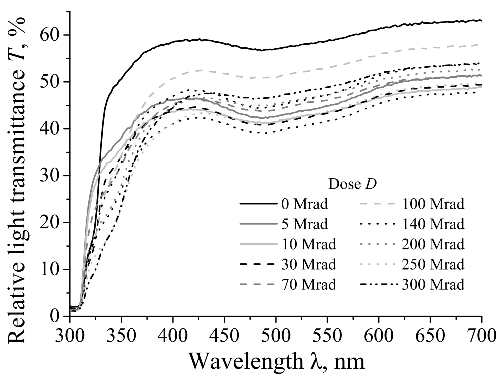

Fig.12. *Luminous transmittance T of the single-layer composite scintillator based on the grains of an Al_2O_3:Ti^{3+} single crystal. Grain size was 1.5...2.0 mm. Dose rate was 1500 Mrad/hour*

3.2. Relative light output

Al_2O_3:Ti^{3+} scintillators

Fig. 13 shows the results of measuring the relative light output L_{rel} for composite scintillators based on the grains of an Al_2O_3 single crystal activated by Ti^{3+}.

Fig.13 demonstrates that L_{rel}-value does not change more than 2 times with the doses increase. It gives the evidence of the high radiation resistance of composite scintillators based on the grains of an Al_2O_3:Ti^{3+} single crystal.

To understand the possible influence of dose rate on the light output of composite scintillators based on Al_2O_3:Ti^{3+}, irradiation was conducted at dose rate of 1500 Mrad/hour as well. Fig. 14 shows the results of measurements of the relative light output L_{rel} for composite scintillators based on the grains of an Al_2O_3:Ti^{3+} single crystal, irradiated at the dose rate of 1500 Mrad/hour.

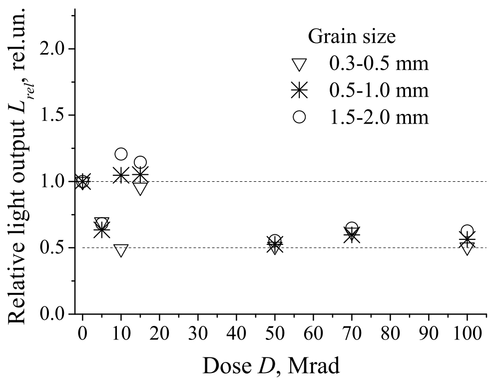

Fig.13. *Relative light output L_{rel} of the single-layer scintillators based on Al_2O_3:Ti^{3+} for different integrated doses D and grain size. Dose rate was 0.23 Mrad/hour*

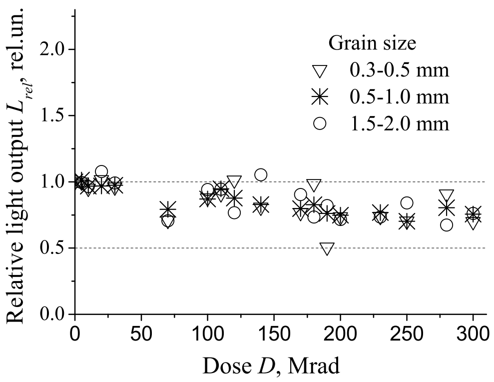

Fig.14. *Relative light output L_{rel} of the single-layer scintillators based on Al_2O_3:Ti^{3+} for different integrated doses D and grain size. Dose rate was 1500 Mrad/hour*

The data presented in Fig. 14 also confirm the high radiation resistance of composite scintillators based on grains of Al_2O_3 single crystals, which have been described above for rate irradiation 0.23 Mrad/hour.

Comparison of the Figs. 13 and 14 show that the irradiation with high dose rate 1500 Mrad/hour results in fluctuations of light output L_{rel} with lower swing then for the case of irradiation with low dose rate 0.23 Mrad/hour.

So, the composite scintillators based on grains of Al_2O_3:Ti^{3+} are radiation-resistant at least up to doses of $D = 100$ Mrad and 300 Mrad when the dose rate was 0.23 Mrad/hour and 1500 Mrad/hour respectively.

For doses $D > 200$ Mrad some of the samples demonstrate a weak tendency for embrittlement of the gel composition.

GSO and GPS scintillators

Fig. 15 demonstrates the relative light output L_{rel} versus dose D for composite scintillators based on GSO:Ce and GPS:Ce single crystal grains. The results were obtained for single-layer scintillators as well as for scintillators a 4 mm thick.

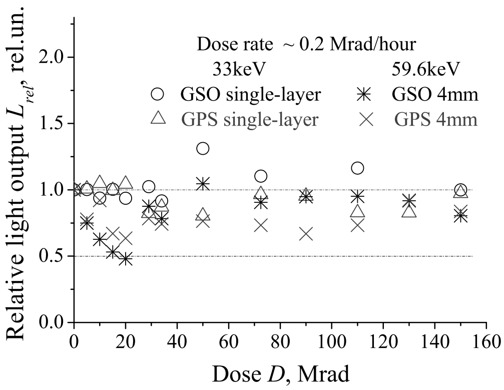

Fig.15. *Relative light output L_{rel} of the scintillators based on GSO:Ce or GPS:Ce single crystal grains for different integrated doses D. Dose rate was 0.23 Mrad/hour*

Fig. 15 shows that L_{rel}-value does not change significantly with D. It indicates the high radiation resistance of composite scintillators based on the grains of GSO and GPS single crystals.

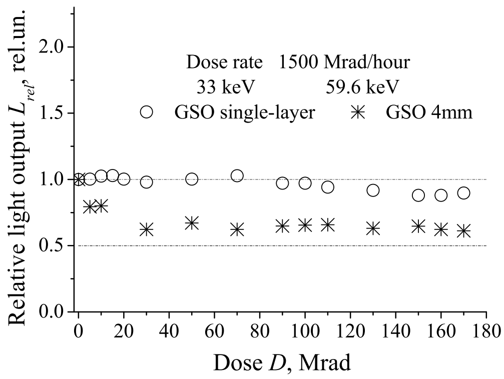

Fig.16. *Relative light output L_{rel} of the scintillators based on GSO:Ce single crystal grains for different integrated doses D. Dose rate is 1500 Mrad/hour*

Fig. 16 shows the results of measurements of the relative light output L_{rel} for composite scintillators based on GSO:Ce single-crystal grains before and after irradiation. Dose rate was 1500 Mrad/hour.

The data presented in Fig. 16 also confirm the effect, which has been described above for composite scintillators based on GSO:Ce single-crystal grains were irradiated with dose rate 0.23 Mrad/hour.

Fig. 15 shows that the light output can fluctuate with the dose D growth. This effect for GSO:Ce single crystals has been described in [7]. It explained as follows. Luminescence centers of GSO:Ce crystal is Ce^{+3}. They are excited as a result of energy transfer from Gd^{+3}. There are a lot of impurities in the GSO crystal, which are the quenchers of the excitation of Gd^{+3}. Due to the influence of radiation such an impurity center can capture an electron. It increases its energy level in energy diagram of the crystal. As the result such a center could not quench the luminescence of the crystal. The lifetime of the intermediate level can have a large spread. According to [7] the time of the existence of the most stable of them could reach a few months.

The results obtained for GPS:Ce have the same character as for GSO:Ce. Therefore the above-described mechanism can also be considered to explain the similar results observed for GPS:Ce.

Fig. 16 shows that for dose rate 1500 Mrad/hour light output is not only $L_{rel} > 0.5$, but also significantly reduces it variations.

Thus, the composite scintillators based on single crystals GSO:Ce and GPS:Ce are radiation-resistant to doses $D = 150$ Mrad for dose rate about 0.23 Mrad/hour and $D = 170$ Mrad for dose rate 1500 Mrad/hour. The scintillation light output and the luminous transmittance T of composite scintillators based on grains of GSO:Ce and GPS:Ce single crystals does not change significantly with dose rate increase.

The study of the scintillators for higher dose is planned.

4. CONCLUSIONS

1. Radiation-resistant composite scintillators based on the grains of inorganic single crystals (GSO:Ce, GPS:Ce and Al_2O_3:Ti^{3+}) were studied.

2. The technology of introduction of the single crystals grains in gel-composition Sylgard-184 was designed.

3. Composite scintillators based on grains of Al_2O_3:Ti^{3+} are radiation-resistant at least up to $D = 100$ Mrad and $D = 300$ Mrad when the dose rate is 0.20 Mrad/hour and 1500 Mrad/hour respectively.

4. Composite scintillators based on single crystal grains of GSO and GPS are radiation resistance at least for $D = 150$ Mrad and $D = 170$ Mrad when the dose rate is 0.2 and 1500 Mrad/hour respectively.

5. For $D > 200$ Mrad some of the samples demonstrate a weak tendency for embrittlement of the gel composition.

6. The variations in the relative light output L_{rel} is higher for lower dose rate.

7. Transmittance decreases less than 2 times over the range of the luminescence of the scintillator, which also indicates that these scintillators are radiation resistant

ACKNOWLEDGEMENTS

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РАДИАЦИОННО СТОЙКИЕ КОМПОЗИЦИОННЫЕ СЦИНТИЛЛЯТОРЫ НА ОСНОВЕ НЕОРГАНИЧЕСКИХ КРИСТАЛЛОВ GSO:Ce, GPS:Ce и Al_2O_3:Ti

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Были изучены радиационно стойкие композиционные сцинтилляторы на основе неорганических монокристаллов GSO:Ce, GPS:Ce и Al_2O_3:Ti. Для получения композиционного сцинтиллятора зёрна были введены в диэлектрическую гель-композицию Sylgard-184. Образцы облучались на электронном (10 МэВ) линейном ускорителе ХФТИ непосредственно электронами пучка с энергией 9,2 МэВ (при мощности дозы 1500 Мрад/час) и тормозными фотонами (при мощности дозы 230 крад/час). Измерение коэффициента пропускания и относительного светового выхода сцинтиллятора проводили до и после облучения. Представлены и проанализированы результаты, полученные для сцинтилляторов, облучённых до доз порядка 170 Мрад и 300 Мрад для GSO:Ce, GPS:Ce и Al_2O_3:Ti соответственно.

РАДІАЦІЙНО СТІЙКІ КОМПОЗИЦІЙНІ СЦИНТИЛЯТОРИ НА ОСНОВІ НЕОРГАНІЧНИХ КРИСТАЛІВ GSO:Ce, GPS:Ce та Al_2O_3:Ti

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Були вивчені радіаційно стійкі композиційні сцинтилятори на основі неорганічних монокристалів (GSO: Ce, GPS: Ce та Al_2O_3:Ti). Для отримання композиційного сцинтилятора зерна були введені в діелектричну гель-композицію Sylgard-184. Зразки опромінювалися на електронному (10 МеВ) лінійному прискорювачі ХФТІ безпосередньо електронами пучка з енергією 9,2 МеВ (при потужності дози 1500 Мрад/год.) і гальмівними фотонами (при потужності дози 230 крад/год.). Вимірювання коефіцієнта пропускання та відносного світлового виходу сцинтилятора проводили до і після опромінення. Представлені та проаналізовані результати, отримані для сцинтиляторів, опромінених до доз порядку 170 Мрад та 300 Мрад для GSO:Ce, GPS:Ce та Al_2O_3:Ti відповідно.