COMPOSITE SCINTILLATORS BASED ON SINGLE CRYSTAL GRAINS Y_2SiO_5:Ce (YSO) AND $Y_3Al_5O_{12}$:Ce (YAG)

N.Z. Galunov[1,2], Ia.V. Gerasymov[1], T.E. Gorbacheva[1], B.V. Grinyov[1], N.L. Karavaeva[1], S.U. Khabuseva[3], A.V.Krech[1] *, L.G. Levchuk[4], E.V. Martynenko[1], V.F. Popov[4], O.Ts. Sidletskiy[1], P.V. Sorokin[4], O. A. Tarasenko[1]

[1] *Institute for Scintillation Materials, National Academy of Sciences of Ukraine, 61001, Kharkiv, Ukraine;*

[2] *V.N. Karasin Kharkiv National University, 61022, Kharkiv, Ukraine;*

[3] *State Scientific Institution "Institute for Single Crystals", National Academy of Sciences of Ukraine, 61001, Kharkiv, Ukraine;*

[4] *National Science Center "Kharkiv Institute of Physics and Technology", 61108, Kharkiv, Ukraine*

(Received April 10, 2017)

Radiation-resistant composite scintillators based on grains of inorganic single crystals of the yttrium orthosilicate Y_2SiO_5:Ce and yttrium aluminium garnet $Y_3Al_5O_{12}$:Ce were studied. The irradiation of the samples by KIPT 10 MeV electron Linac causes the dose rate of 1,500 Mrad/h or 0.1 Mrad/h when it is 9.2 MeV beam electrons or bremsstrahlung photons, respectively. We run the measurements of light transmittance, luminescence spectra and scintillation light outputs of a scintillator before and after irradiation. The paper presents and analyzes the results obtained for scintillators exposed to doses about 25 Mrad (the dose rate is 0.1 Mrad/h) and 150 Mrad (the dose rate is 1,500 Mrad/h).

PACS: 42.88.+h, 81.05.Zx, 81.40.Wx

1. INTRODUCTION

Experiments that 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. The maximum dose accumulated in the CMS HE "critical" zones upon the LHC ultimate shutdown is estimated as 30 Mrad [1]. Furthermore, there are plans of gradual increase of the LHC luminosity by the order of the magnitude in the future. Therefore, a development of materials for radiation detectors with high radiation resistance becomes an important issue.

In the very begging of our study, we investigated the dielectric gel compositions for radiation resistance. The study showed that the light transmittance T-values, practically, did not change with increase in the radiation dose D about 90 Mrad. More details you can see in paper [2].

In previous papers [3],[4] we investigated the composite scintillators based on single crystal grains Gd_2SiO_5:Ce (GSO), $Gd_2Si_2O_7$:Ce (GPS) and Al_2O_3:Ti^{3+} as a radiation-resistance materials. The aim of this work is to study the other promising materials, namely the composite scintillators based on single crystal grains of the yttrium orthosilicate (Y_2SiO_5:Ce (YSO)) and yttrium aluminium garnet ($Y_3Al_5O_{12}$:Ce (YAG)).

As in the previous papers, we are following the classical definition by Birks. If the amplitude of the scintillation signal before irradiation $I(0)$ decreases to $I(D)$ after irradiation by dose D then according to this definition a scintillator is considered as radiation-resistant up to dose D, in the case when the relative amplitude of scintillation pulses $I(D)/I(0) \geq 1/2$, where [5]. Below the same approach we will use not only for scintillation output, but also for the optical transmittance of a scintillation material.

The light yield of YSO single crystal is about 9,200 ... 10,000 photons/MeV. The value of decay times for the single crystals is 42 ns [6]. The light yield of YAG single crystal accepted to estimate 9,000 ... 11,000 photons/MeV. The value of decay times for the single crystals is 120 ns [7].

*Corresponding author E-mail address: AntonKrech@gmail.com

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 [8].

To obtain a composite scintillator we mechanically grind up a single crystal boule. After that, the necessary fraction of the grain sizes we select using a set of calibrated sieves. The grains we introduced in dielectric gel according to the following technique. Firstly, we introduced the grains 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 the composite scintillators with thickness 4 mm (the size of the grains was 0.5...2 mm).

2.2. Irradiation of the samples

As in our earlier studies [2]-[4], we irradiate the samples at the KIPT 10 MeV electron Linac. During the irradiation at the room temperature, the dose rate was practically uniform over the sample surfaces. Inhomogeneity of irradiation of the samples did not exceed 5%. Direct irradiation of a sample by 9.2 MeV beam electrons provided the highest dose rate, namely, 1,500±5 Mrad/h. The samples irradiation by bremsstrahlung photons causes the considerably lower rate. It was equal to 0.10±0.01 Mrad/h. 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 we outlined in [4]. One of the composite scintillators in each series we used as a check sample and it was not exposed to radiation.

2.3. Measurements of scintillation light output and decay time

The set of gamma sources allowed us to calibrate the energy scale of the measuring setup. We used not irradiated composite scintillators of Y_2SiO_5:Ce and $Y_3Al_5O_{12}$:Ce as the reference for corresponding composite scintillators. For all the scintillators the relative light output we obtained as the result of measurements of scintillation amplitude spectra. For YSO:Ce and YAG:Ce the scintillators were excited by alpha particles from radionuclide sources ^{239}Pu (5.15 MeV). Measurements we run before and after irradiation.

The measurements of scintillation pulse shape we run using a delay coincidence technique (see [9]). Gamma radiation from a ^{152}Eu radionuclide source we used to measure a decay time τ of YSO and YAG scintillators.

2.4. Measurements of transmittance

To measure the light transmittance T in the range from 300 to 700 nm we used Shimadzu-2450 spectrophotometer with the integrating sphere. The comparison channel remained blank and the light flux inside it is 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 we 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.

2.5. Measurements of luminescence and excitation spectra

To obtain luminescence spectra and absorption spectra we used spectrofluorimeter Varian Cary Eclipse. In our experiments, the range of wavelengths is 300 - 700 nm.

3. RESULTS AND DISCUSSION

3.1. Measurements of luminescence

Figs. 1 and 2 show photoluminescence spectra for the composite scintillators containing YSO and YAG grains before ($D = 0$) and after irradiation ($D = 150$ Mrad) with dose rate 1,500 Mrad/h.

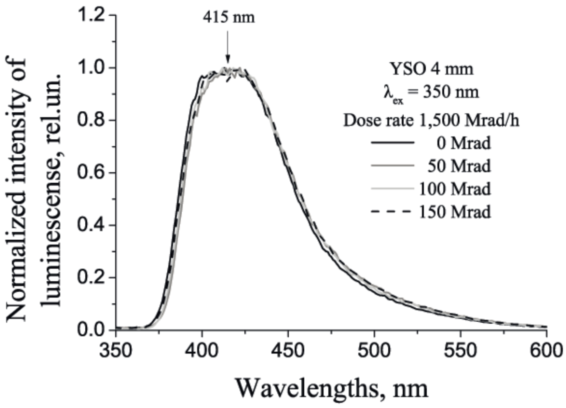

Fig.1. *Normalized luminescence spectra of 4 mm composite scintillator containing grains YSO*

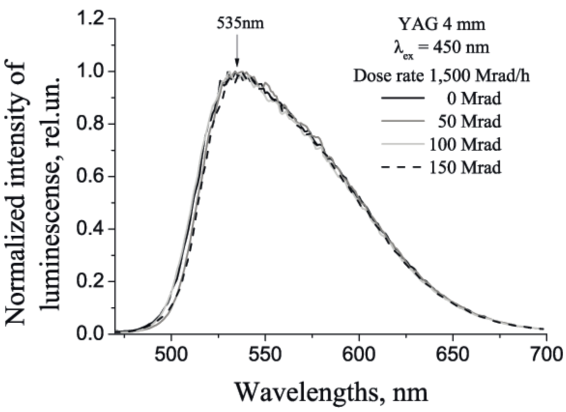

Fig.2. *Normalized luminescence spectra of 4 mm composite scintillator containing grains YAG*

Figs. 1 and 2 represents the normalized spectra and shows the typical example of the luminescence spectra obtained for the composite scintillators based on YSO and YAG grains after irradiation with different D. They demonstrate that the luminescence spectra of composite scintillators do not change their shape with irradiation. This means that new peaks do not appear in this spectrum, and therefore new luminescent centers do not appear too.

3.2. Relative light transmittance

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

Fig.3 and Fig.4 demonstrate the luminous transmittance T of the composite scintillators based on grains of single crystals YSO and YAG, respectively, for different doses of irradiation and the dose rate equal to 1,500 Mrad/h.

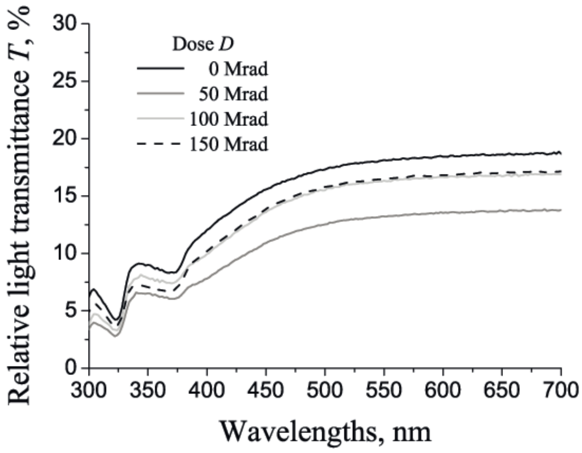

Fig.3. *T versus λ for 4 mm thick composite scintillator based on the grains of a YSO single crystal. The dose rate was 1,500 Mrad/h*

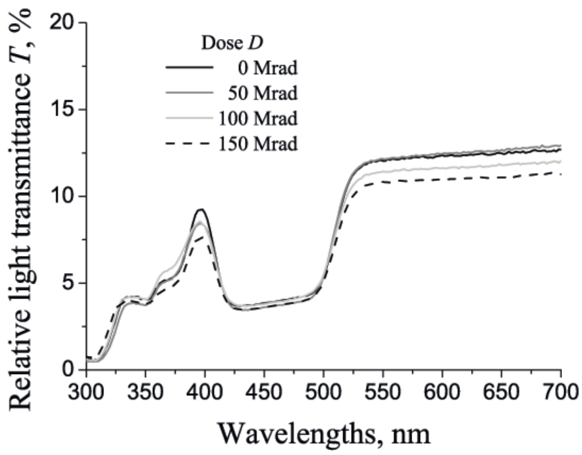

Fig.4. *T versus λ for single-layer composite scintillator based on the grains of a YAG single crystal. The dose rate was 1,500 Mrad/h*

Fig.3 and Fig.4 show that the composite scintillators based on YSO and YAG at least for $D = 150$ Mrad is radiation resistant, because the T-value for the band of luminescence ($\lambda > 390$ nm for YSO and $\lambda > 520$ nm for YAG) reduce less than to half.

3.3. Relative light output and decay time

Fig.5 shows the results of measuring the relative light output L_{rel} for composite scintillators based on the grains of an YSO single crystals.

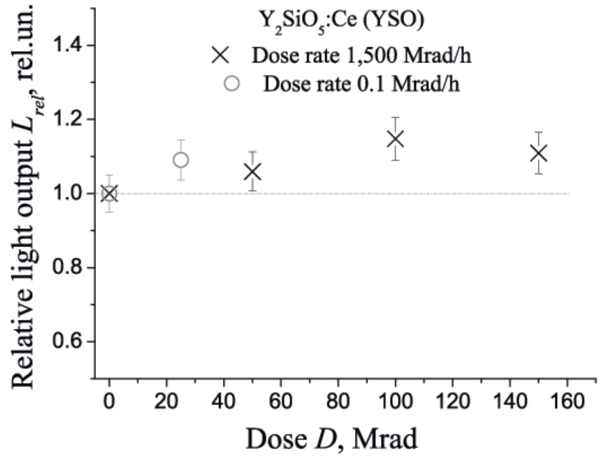

Fig.5. *Relative light output L_{rel} of the scintillators based on Y_2SiO_5:Ce (YSO) for different integrated doses D and dose rate*

Fig.5 demonstrates not only that L_{rel}-value does not change more than 2 times with the doses increase, but also increases with increasing dose. It gives the evidence of the high radiation resistance of composite scintillators based on the grains of an Y_2SiO_5:Ce single crystal.

Fig.6 shows the results of measuring the relative light output L_{rel} for composite scintillators based on the grains of a YAG single crystals.

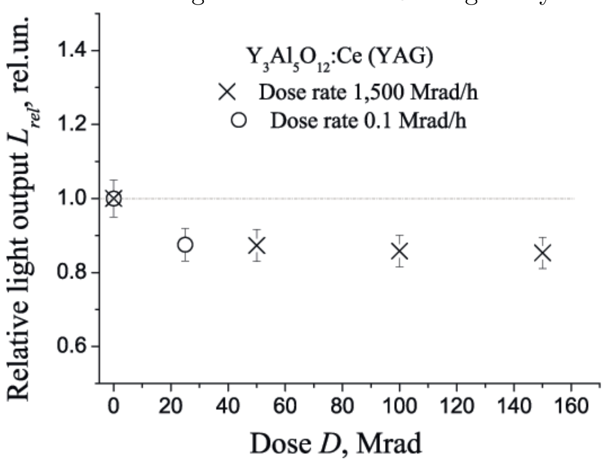

Fig.6. *Relative light output L_{rel} of the scintillators based on $Y_3Al_5O_{12}$:Ce (YAG) for different integrated doses D and dose rate*

Fig.6 demonstrates that L_{rel}-value does not change more than 2 times with the doses increase. E.g., the L_{rel}-value of composite scintillators for dose rate 1,500 Mrad/h and 0.1 Mrad/h are within 0.85 and 0.88 relative units, respectively.

Fig.5 and Fig.6 demonstrates that composite scintillators based on the grains of YSO and YAG are radiation resistance to dose above 25 Mrad with dose rate 0.1 Mrad/h and 150 Mrad with dose rate 1,500 Mrad/h.

To understand the possible influence of dose D on the decay time of composite scintillators based on YSO and YAG, the measurement of the decay time was carried out before and after irradiation. Fig.7 and Fig.8 present these results.

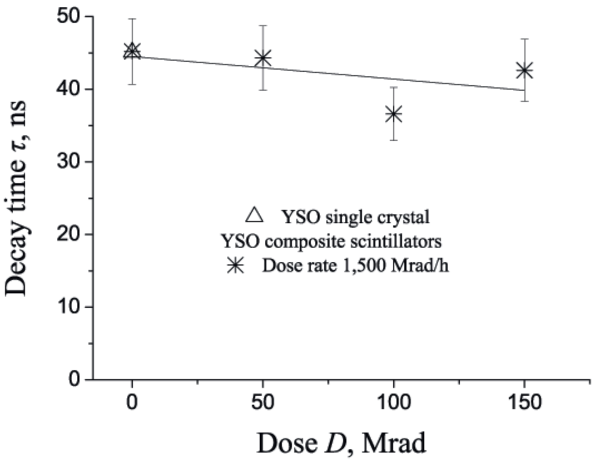

Fig. 7. *The value of decay time for scintillators based on YSO for different integrated doses D*

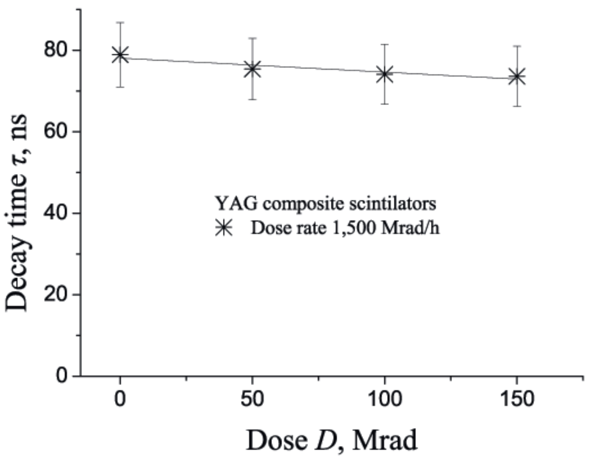

Fig. 8. *The value of decay time for scintillators based on YAG for different integrated doses D*

Comparison of the Figs. 7 and 8 shows that the irradiation of the scintillator leads to a slight decrease in the decay time.

Thus, the composite scintillators based on single crystals YSO:Ce and YAG:Ce are radiation-resistant to doses not less than $D = 150$ Mrad for dose rate about 1,500 Mrad/h and $D = 25$ Mrad for dose rate 0.1 Mrad/h. The scintillation light output and the luminous transmittance T of composite scintillators based on grains of YSO:Ce and YAG:Ce single crystals does not change significantly with dose increase. The normalized luminescence spectra show that new luminescent centers do not appear. The study of the scintillators for higher dose is planned.

4. CONCLUSIONS

1. Composite scintillators based on grains of Y_2SiO_5:Ce (YSO) and $Y_3Al_5O_{12}$:Ce (YAG) are radiation-resistant at least up to $D = 150$ Mrad and $D = 25$ Mrad when the dose rate is 1,500 Mrad/h and 0.1 Mrad/h respectively.

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

3. Luminescence spectra does not change, it means that the new luminescent centers do not appear.

4. Decay time constants of composite scintillators based on grains of the yttrium orthosilicate (YSO) and yttrium aluminium garnet (YAG) decrease with D-value increase from 45 to 40 ns and from 79 to 73 ns, respectively.

References

1. "CMS Collaboration. Technical proposal for the upgrade of the CMS detector through 2020, CERN-LHCC-2011-006, CMS-UG-TP-1, LHCC-P-004, CERN, Geneva, 2011.

2. A.Yu. Boyarintsev, N.Z. Galunov, N.L. Karavaeva, et al. Study of radiation-resistant gel bases for composite detectors // *Functional Materials.* - 2013, v.20, N6, p.471-476.

3. A.Yu. Boyarintsev, N.Z. Galunov, Ia.V. Gerasymov, T.E. Gorbacheva, N.L. Karavaeva, A.V. Krech, L.G. Levchuk, L.A. Litvinov, V.F. Popov, O.Ts. Sidletskiy, P.V. Sorokin, O.A. Tarasenko. Radiation-resistant composite scintillators based on inorganic crystals (GSO:Ce, GPS:Ce and Al_2O_3:Ti) // *PAST, Ser.: Nuclear Physics Investigations.* 2016, N5(105), p.59-65.

4. A.Yu. Boyarintsev, N.Z. Galunov, Ia.V. Gerasymov, N.L. Karavaeva, A.V. Krech, L.G. Levchuk, V.F. Popov, O.Ts. Sidletskiy, P.V. Sorokin, O.A. Tarasenko. Radiation-resistant composite scintillators based on GSO and GPS grains // *Nuclear Inst. and Methods in Physics Research*, A. 2017. v.841, p.124-129.

5. J.B. Birks. *The theory and practice of scintillation counting.* Oxford, London: "Pergamon Press Ltd". 1967, 662 p, (p.55-205).

6. Proceedings of the international conference on inorganic scintillators and their applications, Delft, The Netherlands, 28 August – September 1, 1995: SCINT 95

7. E. Mihokova, M. Nikl, J.A. Mares, A. Beitlerova, A. Vedda, K. Nejezchleb, K. Blazek, and C. D'Ambrosio. Luminescence and scintillation properties of YAG : Ce single crystal and optical ce-ramics // *Journal of Lumines-cence*, 126:77-80, 2007.

8. Sigma-Aldrich. 761036 Aldrich Sylgard-184. Available: http://www.sigmaaldrich.com/... .../catalog/product/aldrich/761036.

9. N.Z. Galunov, and V.P. Seminozhenko. *The Theory and Application of the Radioluminescence of Organic Condensed Media* Kiev: "Naukova Dumka". 1997, 280 p. (in Russian).

КОМПОЗИЦИОННЫЕ СЦИНТИЛЛЯТОРЫ НА ОСНОВЕ МОНОКРИСТАЛЛИЧЕСКИХ ГРАНУЛ Y_2SiO_5:Ce (YSO) И $Y_3Al_5O_{12}$:Ce (YAG)

Н.З. Галунов, Я.В. Герасимов, Т.Е. Горбачева, Б.В. Гринёв, Н.Л. Караваева, С.У. Хабусева, А.В. Креч, Л.Г. Левчук, Е.В. Мартыненко, В.Ф. Попов, О.Ц. Сидлецкий, П.В. Сорокин, О.А. Тарасенко

Исследованы на радиационную стойкость композиционные сцинтилляторы на основе гранул неорганических монокристаллов Y_2SiO_5:Ce и $Y_3Al_5O_{12}$:Ce. 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/ч) и тормозными фотонами (при мощности дозы 0,1 Мрад/ч). Значения пропускания света, люминесценции и светового выхода измерялись до и после облучения. Представлены и проанализированы результаты, полученные для сцинтилляторов, облученных до доз 25 Мрад при темпе 0,1 Мрад/ч и 150 Мрад при темпе 1500 Мрад/ч.

КОМПОЗИЦІЙНІ СЦИНТИЛЯТОРИ НА ОСНОВІ МОНОКРИСТАЛІЧНИХ ГРАНУЛ Y_2SiO_5:Ce (YSO) ТА $Y_3Al_5O_{12}$:Ce (YAG)

М.З. Галунов, Я.В. Герасимов, Т.Є. Горбачова, Б.В. Гриньов, Н.Л. Караваєва, С.У. Хабусєва, А.В. Креч, Л.Г. Левчук, Є.В. Мартиненко, В.П. Попов, О.Ц. Сідлецький, П.В. Сорокін, О.А. Тарасенко

Досліджені на радіаційну стійкість композиційні сцинтилятори на основі гранул неорганічних монокристалів Y_2SiO_5:Ce та $Y_3Al_5O_{12}$:Ce. Для отримання композиційних сцинтиляторів гранули були введено 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 0,1 Мрад/год.). Значення пропускання світла, люмінесценції та світлового виходу вимірювалися до і після опромінення. Представлені та проаналізовані результати, отримані для сцинтиляторів, опромінених до доз 25 Мрад при темпі 0,1 Мрад/год. та 150 Мрад при темпі 1500 Мрад/год.