ISSN 1027-5495. Functional Materials, 27, No.1 (2020), p. 18-23.
doi:https://doi.org/10.15407/fm27.01.18

Radioactivity induced in radiation-resistant composite scintillators by irradiation with bremsstrahlung photons

***V.F.Popov*[1], *A.Yu.Boyarentsev*[2], *N.Z.Galunov*[2,3], *B.V.Grinyov*[2], *N.L.Karavaeva*[2], *A.V.Krech*[2], *L.G.Levchuk*[1]**

[1]National Science Center "Kharkiv Institute of Physics and Technology", 1 Akademichna Str., 61108 Kharkiv, Ukraine
[2]Institute for Scintillation Materials, STC "Institute for Single Crystals", National Academy of Science of Ukraine, 60 Nauky Ave., 61072 Kharkiv, Ukraine
[3]V.Karazin Kharkiv National University, 4 Svobody Sq., 61022 Kharkiv, Ukraine

Received August 6, 2019

Radioactivity induced by bremsstrahlung photons in samples of composite scintillators containing grains of inorganic GSO:Ce, GPS:Ce, YSO:Ce and YAG:Ce crystals is studied. Measurements of the amplitude spectra of composite scintillators indicate the presence of intrinsic gamma activity resulting from the irradiation. The results obtained agree with the assumption of long-lived gamma-emitting isotope production. This should be taken into account, when composite scintillators are used for particle registration with detectors subjected to intensive irradiation.
Keywords: composite scintillators, radiation resistance, high energy physics.

Исследуется радиоактивность, наведенная при облучении тормозными фотонами в образцах композиционных сцинтилляторов, содержащих гранулы неорганических кристаллов GSO:Ce, GPS:Ce, YSO:Ce и YAG:Ce. Измерения амплитудных спектров композиционных сцинтилляторов указывают на наличие собственной гамма-активности, обусловленной облучением. Полученные результаты согласуются с предположением об образовании долгоживущих гамма-активных изотопов. Это явление должно учитываться при использовании композиционных сцинтилляторов для регистрации частиц детекторами, подверженными интенсивному облучению.

Дослідження наведеної радіоактивності у радіаційно-стійких композиційних сцинтиляторах при їх опроміненні гальмівними фотонами. *В.П.Попов, А.Ю.Бояринцев, М.З.Галунов, Б.В.Гриньов, Н.Л.Караваєва, А.В.Креч, Л.Г.Левчук.*

Досліджується радіоактивність, наведена при опроміненні гальмівними фотонами в зразках композиційних сцинтиляторів, що містять гранули неорганічних кристалів GSO:Ce, GPS:Ce, YSO:Ce і YAG:Ce. Вимірювання амплітудних спектрів композиційних сцинтиляторів вказують на наявність власної гамма-активності, обумовленої опроміненням. Отримані результати узгоджуються з припущенням про утворення довгоживучих гамма-активних ізотопів. Це явище має враховуватися при використанні композиційних сцинтиляторів для реєстрації частинок детекторами, які підлягають інтенсивному опроміненню.

1. Introduction

Particle detectors (including their scintillator-based subsystems such as calorimeters) used in experimental high energy physics are often exposed to very high (up to several tens of megarads) irradiation doses. This is the case in particular for the main experiments at the Large Hadron Collider (LHC) at CERN. Furthermore, the LHC upgrade to the high-luminosity (HL) collider parameters (see e.g., Ref. [1]) will make the dose pressure upon the detector subsystems even considerably more severe. Therefore, search of new radiation-tolerant and relatively cheap scintillation materials is now of great relevance and importance. We have already studied earlier (see Refs. [2–5]) the radiation-resistance of composite scintillators based on single crystal grains Gd_2SiO_5:Ce (GSO:Ce), $Gd_2Si_2O_7$:Ce (GPS:Ce), Al_2O_3:Ti, Y_2SiO_5:Ce (YSO:Ce) and $Y_3Al_5O_{12}$:Ce (YAG:Ce).

A composite scintillator it is a transparent non-scintillating gel composition, which contains single-crystal scintillation grains. Composite scintillators have a number of advantages in comparison with other scintillation materials those are continuous mediums [2, 6, 7]:

– In some cases, it is possible to exclude the expensive stage of growing a single crystal from the process of producing grains using a polycrystalline ingot obtained during the zone melting process, or use the waste arising from the treatment of single crystals.

– If it necessary, a specially prepared gel composition can unite the separate parts of composition scintillator in one uniform sample. Therefore, it is possible to create, practically, a detector with a very large input window.

– It is possible to vary both the sample size and grains size independently.

– Unlike classical scintillators, the basis of composite scintillators is not the luminescent material and the luminescence occurs in grains. This means that if we choose a non-luminescent base material, then a change in its luminescence will not affect the properties of the scintillators, since such a luminescence is absent.

We use the polydimethylsiloxane gel-composition that does not contain benzene rings. It is a non-luminescent material, and thereby only a change in the transparency of gel-composition occurring in the luminescence band of the grains can influence on the scintillation amplitude of the composite scintillator [2, 8, 9].

In our studies, irradiation is carried out at an experimental facility the NSC "Kharkiv Institute of Physics and Technology" (KIPT) based on an electron linear accelerator (Linac). The electron beam is incident onto a converter to produce an intensive flux of bremsstrahlung photons, which is used to irradiate the scintillator samples. There is also some admixture to this flux of neutrons (from photo-nuclear reactions) as well as of low-energy photons and electrons/positrons coming from "secondary" interactions with target atoms. In this work, we present a study of activation of composite scintillators containing grains of inorganic crystals GSO:Ce, GPS:Ce, YSO:Ce and YAG:Ce under such irradiation conditions and estimate its possible effect on the distortion of the scintillation signal.

2. Experimental

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

The following approach was applied to prepare the composite scintillator samples investigated in this work. Initially, we grind up a single crystal boule mechanically to obtain scintillation grains. After that, we use a set of calibrated sieves to select the necessary fraction of their sizes. The grains were introduced in dielectric gel according to the following technique. In the begging, we entered the grains into the first component of the gel. After adding the second component, we thoroughly mix the gel composition. Finally, we introduce this gel composition into the molding tank, in which we leave it until full polymerization. After that, we removed the scintillator from the forming container. We investigated the composite scintillators with thickness 4mm (the size of the grains was 0.5–2 mm) [2–4].

As in our earlier studies [2–4, 10–12], we irradiated the samples at the KIPT 10 MeV Linac at the room temperature. The dose rate was practically uniform over the sample surfaces, with deviations within 5 %. The highest dose rate, 1500±5 Mrad/h, was provided, when the samples were irradiated directly by 9.6 MeV beam electrons. Other samples were subjected to irradiation by bremsstrahlung photons at the considerably lower rate of 0.10±0.01 Mrad/h (mainly photons of bremsstrahlung radiation). The samples (by one item of each type) were successively exposed to radiation until they accumulated the necessary integrated radiation dose. The dose was measured by the Harwell Red 4034

Table 1. Results of the measurements of the amplitude γ-scintillation spectra for the samples of composite scintillators containing single-crystal grains of GSO:Ce

No.	E_γ, keV	N_{sum} after 17 h	Isotope	Reference values		Reaction	Half life
				E_γ, keV	I_γ, %		
1	58	158					
2	79.6	8					
3	97.4	1865	^{153}Gd	97.431	29	γ, *n*	240.4 d
4	103	1786	^{153}Gd	103.18	21.11	γ, *n*	240.4 d
5	145.4	406	^{141}Ce	145.44	48.2	*n*, γ	32.5 d
6	165.7	1890	^{139}Ce	165.9		γ, *n*	137.6 d
7	226.1	1673					
8	290.5	183					
9	305.6	350					
10	348.1	1185					
11	363.5	60121	^{159}Gd	363.55		γ, *n*	18.48 h
12	560	94					
13	581	226					
14	617–619	54					
15	1460.8	5	^{40}K	1461	10.7		4.7^{11} d

plastic dosimeters to an accuracy of ±10 %. The details of the measurements are outlined in [2, 3, 10–12]. All the absorption dose (and dose-rate) values presented below are given for a water equivalent. One of the samples in each series was taken as the reference scintillator and was not exposed to radiation.

To study the induced gamma-radioactivity of the composite scintillator samples, we measured the amplitude gamma-scintillation spectra. For these measurements, we used the BVEG2-22 detection unit containing the ∅40×40 mm^2 cylindrical NaI(Tl) scintillation single crystal. To eliminate the gamma background, the detector and the studied sample were placed into a shielding lead box with the wall thickness of 80 mm.

The irradiation of the samples of composite scintillators by bremsstrahlung photons consisted from several successive runs, for which the samples got the absorption dose rate of 0.113 Mrad/h (for a water equivalent). The total integral dose was 190 Mrad.

3. *Results and discussion*

To measure the amplitude gamma-scintillation spectra, we used the Canberra semiconductor detector. We measured the spectra 17 and 17.5 hours after irradiation for composite scintillators containing grains of GSO:Ce and GPS:Ce, respectively. Table 1 and Table 2 show the results of measurements for the GSO:Ce (exposed during 900 sec.) and GPS:Ce (exposed during 1200 sec.) samples, respectively. In 21 days, the measurements (with the same exposition times) were repeated, and practically the same results were obtained.

Samples of composite scintillators based on single crystal grains of YSO:Ce and YAG:Ce were irradiated by bremsstrahlung photons to the integral dose of 26.2 Mrad at the dose rate of 0.11 Mrad/h. The technical light output of the YSO:Ce and YAG:Ce samples was measured before and after their irradiation. The value of the light output of the scintillator under the ^{239}Pu alpha source is represented by the channel number (Mean$_i$) of the charge-digital converter (QDC). The scintillator samples were placed on the photocathode of a photomultiplier without optical contact.

Fig. 1 shows the results. According to these results the light output after the sample irradiation remained almost unchanged. For, e.g., the YSO:Ce sample, the alpha-peak centroid position corresponds to channel 506±15 and 528±16 before and after irradiation, respectively. However, a huge number of small- and large-amplitude signals emerge after the irradiation that may indicate the sample "activation". (The YAG:Ce sample demonstrates the similar behavior). To test this assumption, we measured the sample gamma-scintillation amplitude spectra exploiting the same experimental technique as the one used for the GSO samples. The measurements were carried out in 15 days after the irradiation.

Table 2. Results of the measurements of the amplitude γ-scintillation spectra for the samples of composite scintillators containing single-crystal grains of GPS:Ce

No.	E_γ, keV	N_{sum} after 17.5 h	Isotope	Reference values		Reaction	Half life
				E_γ, keV	I_γ, %		
1	57.9						
2	79.6						
3	97.4	2549	^{153}Gd	97.431	29	γ, *n*	240.4 d
4	103	2519	^{153}Gd	103.18	21.11	γ, *n*	240.4 d
5	145.4	15377	^{141}Ce	145.44	48.2	*n*, γ	32.5 d
6	165.7	80063	^{139}Ce	165.9		γ, *n*	137.6 d
7	225.9	1355					
8	290.5						
9	305.6	246					
10	348.4	1132					
11	363.4	53994	^{159}Gd	363.55		γ, *n*	18.48 h
12	560	55					
13	581	201					
14	617–619						
15	1460.8	6	^{40}K	1461	10.7		4.7^{11} d

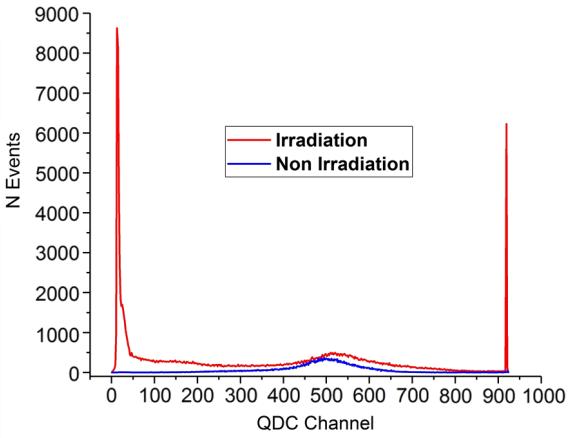

Fig. 1. Spectra of scintillation under α source (^{239}Pu) for composite scintillator based on YSO:Ce grains measured before and after irradiation with bremsstrahlung photons.

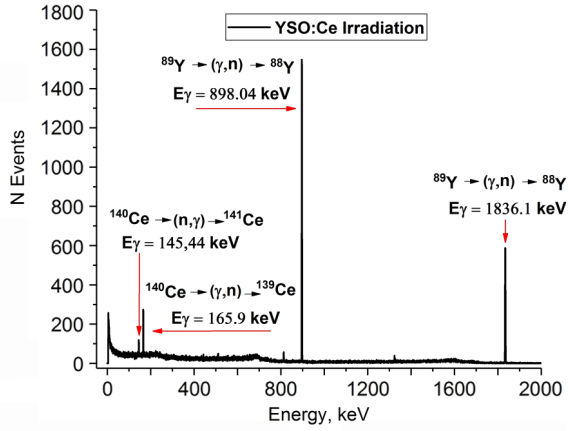

Fig. 2. Amplitude scintillation γ-spectrum for composite scintillator based on YSO:Ce grains; measurements were carried out 15 days after completion of the irradiation to dose of 26.2 Mrad.

The results for the YSO:Ce (YAG:Ce) samples are presented in Table 3 and Fig. 2 (Table 4 and Fig. 3). The measurement time (T_{exp}) was 2000 sec. and 1000 sec. for the YSO:Ce and YAG:Ce samples, respectively. The measurements indicate that composite scintillators based on YSO:Ce and YAG:Ce grains were "activated" by the bremsstrahlung-photon irradiation with production of the long-lived ^{141}Ce, ^{139}Ce and ^{88}Y gamma-active isotopes.

The same measurements were carried out also for the YSO:Ce- and YAG:Ce-based composite scintillators irradiated to dose of 51.8 Mrad (see Fig. 4) and 100 Mrad (see Fig. 5). The results were obtained in 10 days after completion of the irradiation. The NaI(Tl) detector was calibrated by the ^{137}Cs and ^{60}Co radionuclide sources, and the corresponding amplitude spectra are also shown on Figs. 4 and 5 as a reference plot.

Fig. 2–5 clearly demonstrate the presence of the ^{88}Y isotope gamma peaks with energies of 898 and 1836 keV, as well as the overlap of two peaks with energies of 145 and 166 keV from the ^{141}Ce and ^{139}Ce isotopes, respectively. The presence of the

Table 3. Results of the measurements of the amplitude γ-scintillation spectra for the samples of composite scintillators containing single-crystal grains of YSO:Ce

No.	E_γ, keV	N_{sum} (in peak)	N_{sum}/T_{exp}, 1/sec	Isotope	Reference values		Reaction	Half life
					E_γ, keV	I_γ, %		
1	145.1	434	0.217	^{141}Ce	145.44	18.2	n, γ	32.5 d
2	166	1011	0.5055	^{139}Ce	165.9	80	γ, n	137.6 d
3	812.8	304	0.152	?				
4	897.5	8664	4.332	^{88}Y	898.04	93.7	γ, n	107 d
5	1323.4	315	0.1575	?				
6	1460.8	52	0.026	^{40}K	1461	10.7		4.7^{11} d
7	1834.5	4655	2.3275	^{88}Y	1836.1	99.2	γ, n	107 d

Table 4. Results of the measurements of the amplitude γ-scintillation spectra for the samples of composite scintillators containing single-crystal grains of YAG:Ce

No.	E_γ, keV	N_{sum} (in peak)	N_{sum}/T_{exp}, 1/sec	Isotope	Reference values		Reaction	Half life
					E_γ, keV	I_γ, %		
1	145.1	403	0.403	^{141}Ce	145.44	18.2	n, γ	32.5 d
2	166	710	0.71	^{139}Ce	165.9	80	γ, n	137.6 d
3	812.8	131	0.131	?				
4	897.5	4239	4.239	^{88}Y	898.04	93.7	γ, n	107 d
5	1460.8	14	0.014	^{40}K	1461	10.7		4.7^{11} d
6	1834.5	2171	2.171	^{88}Y	1836.1	99.2	γ, n	107 d

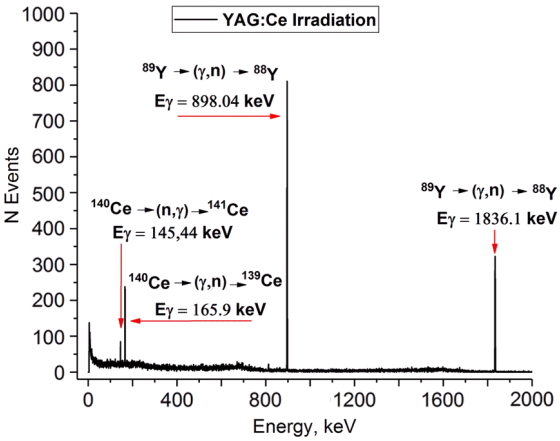

Fig. 3. Amplitude scintillation γ-spectrum for composite scintillator based on YAG:Ce grains; measurements were carried out 15 days after completion of the irradiation to dose of 26.2 Mrad.

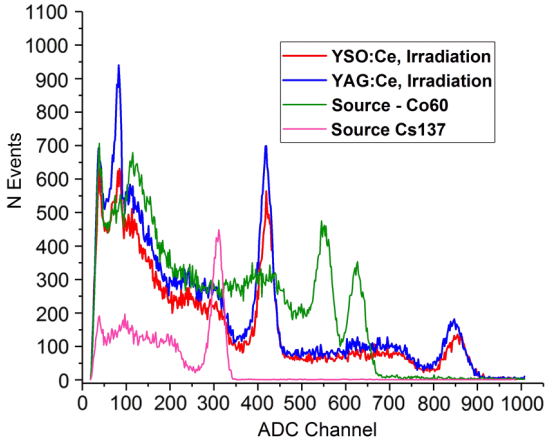

Fig. 4. Amplitude scintillation γ-spectra for composite scintillators based on YSO:Ce and YAG:Ce grains; measurements were carried out 10 days after completion of the irradiation to dose of 51.8 Mrad. For comparison, the scintillation γ-spectra for ^{137}Cs and ^{60}Co sources are also shown.

^{141}Ce isotope is due to the admixture of neutrons in the irradiation area of the KIPT Linac. It should be stressed that the Linac electron beam energy is not restricted by 9.6 MeV. Actually, the beam energy distribution has a long tail up to 17 MeV, which is responsible for extension of the bremsstrahlung photon spectrum to higher energies. The neutrons are mainly produced in photo-nuclear reactions with high-energy photons.

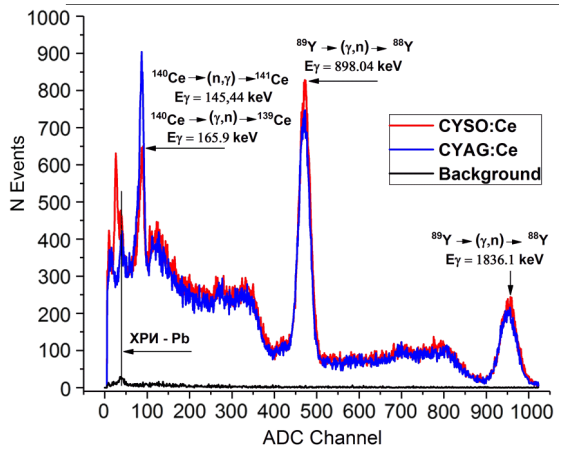

Fig. 5. Amplitude scintillation γ-spectra for composite scintillators based on YSO:Ce and YAG:Ce grains; measurements were carried out one day after completion of the irradiation to dose of 100 Mrad.

In Fig. 5, the background spectrum due to gamma-radiation of the detector environment is also shown. This, in particular, includes the amplitude spectrum of the characteristic X-ray radiation from lead detector shielding.

4. Conclusions

The measured amplitude scintillation spectra of the composite scintillators manifest the presence of intrinsic gamma activity resulting from irradiation with bremsstrahlung photons. The results obtained are consistent with the assumption of the long-lived gamma-active isotope production. Upon irradiation with bremsstrahlung photons, the composite-scintillator samples containing grains of GSO:Ce and GPS:Ce are activated with production of the ^{153}Gd, ^{159}Gd, ^{141}Ce and ^{139}Ce isotopes, which have relatively long life time. The ^{141}Ce, ^{139}Ce, ^{159}Gd and ^{153}Gd half-life is 32.5 days, 137.6 days, 18.48 hours and 240.4 days, respectively. Similarly, the YSO:Ce- and YAG:Ce-based composite scintillators are activated with the production of the long-lived ^{141}Ce, ^{139}Ce and ^{88}Y isotopes with half-lives of 32.5, 1 37.6 and 107 days, 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, Ia.V.Gerasymov et al., *Nucl.Inst. Meth. Phys. Res., A*, **841**, 124 (2017).
3. N.Z.Galunov, T.E.Gorbacheva, B.V.Grinyov et al., *Nucl. Inst. Meth. Phys. Res, A*, **866**, 104 (2017).
4. N.Z.Galunov, Ia.V.Gerasymov, T.E.Gorbacheva et al., *Problem Atom. Scien. Techn*, **3**, 35 (2017).
5. A.V.Krech, N.Z.Galunov, *Ukr. J. Phys.*, **62**, 569 (2017).
6. N.Z.Galunov, B.V.Grinyov, N.L.Karavaeva et al., *IEEE Trans. Nucl.Scien.*, **56**, 904 (2009).
7. N.Z.Galunov, B.V.Grinyov, N.L.Karavaeva et al., *IEEE Trans. Nucl. Scien.*, **58**, 339 (2011).
8. E.Biagtan, E.Goldberg, J.Harmon et al., *Nucl. Instr. Meth. Phys. Res., B*, **93**, 296 (1994).
9. A.Quaranta, S.Carturan, T.Marchi et al., *Nucl. Instr. Meth. Phys. Res., B*, **268**, 3155 (2010).
10. A.Yu.Boyarintsev, N.Z.Galunov, N.L.Karavaeva et al., *Functional Materials*, **20**, 471 (2013).
11. A.Yu.Boyarintsev, N.Z.Galunov, Ia.V.Gerasymov et al., *Probl. Atom. Scien. Techn.* , **5**, 59 (2016).
12. A.Yu.Boyarintsev, N.Z.Galunov, Ia.V.Gerasymov et al., *Probl.. Atom. Scien. Techn.* , **3**, 60 (2019).