RADIATION RESISTANCE OF COMPOSITE SCINTILLATORS

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Radiation resistance of composite scintillators containing grains of organic or inorganic single crystals has been analysed. The analysis is based on the study of composite scintillators containing grains Gd_2SiO_5:Ce (GSO:Ce), $Gd_2Si_2O_7$:Ce (GPS:Ce), Al_2O_3:Ti, Y_2SiO_5:Ce (YSO:Ce) or $Y_3Al_5O_{12}$:Ce (YAG:Ce). This paper presents the results of research and suggests possible mechanisms as well as processes of radiation changes that occur in scintillators due to irradiation. It also discusses how these effects can affect the scintillation characteristics of composite scintillators exposed to radiation.

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1. INTRODUCTION

A large number of experiments conducted on charged-particle accelerators indicate that radiation dose accumulated by the detectors and, in particular, by the scintillation materials contained in them a significant. For example, in experiments at the Large Hadron Collider (LHC), the dose of radiation for the scintillator detectors can reach 10...100 Mrad, and in the future (according to LHC modernization plans, see, e.g., [1]) will be even more. In this regard, it is especially important to search for new radiation-resistant scintillation materials.

As a definition of "radiation-resistant", we will use the classical one formulated by the Birks [2]. According to this definition, a scintillator is considered to be radiation resistant up to a dose D, if after irradiation the amplitude of the scintillation signal obtained before irradiation $I(0)$, reduces to $I(D)$ after irradiation so that the relative amplitude of the scintillation pulses $I(D)/I(0) \geq 1/2$.

Previously [3, 4, 5, 6] we investigated the 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) as a radiation-resistance materials.

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 [3, 7, 8]:

1) A composite scintillation material is cheaper and easier in production than a single crystal. In a number of cases, it is possible to pass a costly stage of growth of a single crystal, or to use the waste arising from processing of single crystals. A main loss of the scintillation material that appears during machining is absent, because machining is absent.

2) It is possible to create an almost infinite area. A specially prepared gel composition can unite the separate parts of composition scintillator in one uniform sample.

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

4) Unlike classical scintillators, the basis of composite scintillators should not be luminescent because the luminescence occurs completely 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

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that does not contain benzene rings. It is the non-luminescent material, and thereby a change in the transparency of gel-composition occurring in the luminescence band of the grains can only influence on the scintillation amplitude of the composite scintillator [3, 9, 10].

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 [11].

To make a composite scintillator we use the following approach. 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. Firstly, we introduced them in the first component of the gel. After adjunction of the second component, we carefully mix the gel composition. Finally we introduce this gel composition 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). [3, 4, 5].

2.2. Irradiation of the samples

As in our earlier studies [3, 4, 5, 12, 13], we irradiated the samples at the KIPT 10 MeV electron Linac 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/h, 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 0.20±0.01 Mrad/h (mainly photons of braking radiation). 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, 4, 12, 13]. One of the samples in each series was taken as reference scintillator and was not exposed to radiation.

2.3. Measurements of scintillation light output

The set of gamma sources allowed us to calibrate the energy scale of the measuring setup. We used not irradiated composite scintillators 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. The values of the relative light output were obtained by irradiation with the radionuclide sources of (i) photons of gamma radiation emitted by ^{137}Cs, (ii) alpha particles emitted by ^{239}Pu, and (iii) photons emitted by ^{241}Am. 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:

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

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

As was shown in previous works [3, 4, 5, 6, 13], the value of the relative light output L_{rel} of composite scintillators did not decrease more than two times after irradiation up to limiting dose when scintillators could start to crack. The limiting doses that were obtained for the developed composite scintillators are presented in Table.

Comparison of radiation resistance

Scintillator	Dose rate, Mrad/h	Limiting dose, Mrad	Causes of loss of radiation resistance
Organic single crystal scintillators	0.2	< 1	$L_{rel} < 0.5$
SCSN-81 plastic scintillator	0.2	$\sim 10...15$	$L_{rel} < 0.5$
Composite scintillator based on Al_2O_3:Ti grains	0.2 1500	~ 125 ~ 550	Mechanical self-destruction
Composite scintillator based on GSO:Ce or GPS:Ce grains	0.2 1500	~ 200 ~ 250	Mechanical self-destruction
Composite scintillator based on YSO:Ce or YAG:Ce grains	1500	> 150	Steady work is observed

In the Table also shows the radiation resistance of known scintillators (Organic single crystal scintillators and SCSN-81 plastic scintillator). The difference in the radiation resistance of the developed composite scintillators with known scintillators can reach three orders of magnitude.

According to [3, 4, 5, 6], the value of the relative light output L_{rel} can fluctuate with increasing radiation dose D. These fluctuations are observed against the background of the main decrease in relative light output L_{rel} values. Such fluctuations may arise because the radioluminescence of scintillators whose luminescent centres are damaged by radiation cannot be described by a monotonically decreasing function. The irradiation of the scintillator material not only leads to irreversible destruction of its base material, but also gives rise to secondary processes.

Therefore, the dependencies of the relative light output L_{rel}, optical transmission T, and others on the irradiation dose include a stochastic component. Such stochastic components can arise as result of material activation, modification of luminescence centres, changes in the decay efficiency of impurity centres and ions, and short-term damage of luminescence centres in grains with their further rapid recovery. Of course, the ability of a material to restore its parameters after irradiation will constantly decrease as the dose D increases.

It is possible to consider two alternative versions of the scenario for the development of these stochastic secondary processes.

In the first case, we have to take into account that all the scintillation materials contain defects, impurity ions, molecules, and others, which can create levels in the energy diagram of the crystal and play the role of quenching centres [6]. Ionization creates free electrons. When an electron is captured by one of these quenching centre, such the modify centre ceases to be quenching centre during its lifetime. By reducing the quenching effect, the light output increases. For this process, a characteristic feature is the invariance of the luminescence spectrum, as long as the scintillator is capable of luminescence. This is because new luminescent centres are not formed.

In the second case, the process has a different nature. Let us assume that molecules, ions, etc., which is a primary luminescent center of type A are damaged during irradiation, as a result of which centres of type B appear. If centres of type B are new additional luminescent centers with a higher light output than the light output of centres of type A, then the intensity of the scintillator luminescence increases after irradiation during the lifetime of type B centres. After this, it should decrease [13]. This type of process includes those for which irradiation is possible both direct and reverse transitions between the main centres and those that are formed under the action of radiation.

Consequently, there are two possible options for the development of processes after irradiation, which may be responsible for the stochastic component of the fluctuations in the luminescence intensity. In the first case, the spectrum of luminescence of the scintillator does not change its shape for different doses D, since new luminescence centres are absent. An alternative process is determined by the change in the luminescence centres during irradiation and should lead to a change in the spectral characteristics of the luminescence.

In the composite scintillators presented in our previous studies [3, 4, 5, 6, 13, 14, 15] under the influence of ionizing radiation, only one of these processes is observed, and this process is characteristic of a particular material. For organic scintillators, changes in their luminescence spectra are observed, and these systems are relatively non-radiation-resistant (up to 0.5 Mrad for organic crystals [15]). At the same time, for the inorganic scintillators, the change in the luminescence spectra can be observed (Al_2O_3:Ti) or not be observed (GSO:Ce, GPS:Ce, YSO:Ce, YAG:Ce) [3, 4, 5, 6]. Let's consider each case separately.

In organic scintillators, the luminescence spectrum can be shifted, since these systems contain benzene rings (they are responsible for luminescence). Under ionizing radiation, these rings are destroyed. The probability of their recovery is low. This process degrades the optical properties of the matrix. At the same time, however, a new organic molecule can be formed, with luminescence in a different spectral range.

The spectra of luminescence of composite scintillators based on grains of Al_2O_3:Ti were presented in papers [4, 6, 13]. Figures in these papers show a change in the luminescence spectra. It is observed in [4] that with increasing accumulated dose D in the blue luminescence band, the luminescence intensity rises sharply with subsequent saturation or even a significant decrease. This luminescence is associated with centres containing Ti^{4+} ions. With increasing dose, the luminescence intensity in IR intervals (this luminescence is associated with centres containing Ti^{3+} ions) decreases at the beginning, and then either slightly increases or it reduction stops.

These facts allow us to conclude that, at low doses, the formation of centres containing Ti^{4+} ions that arise upon ionization of Ti^{3+} is the dominant process. Thus, the concentration of centres containing Ti^{4+} ions increases, and the probability of direct exposure to these centres grows. Therefore, with increasing dose D, the probability of destruction for centres containing Ti^{4+} ions increases. Upon irradiation, the latter process may lead to a reverse transition from Ti^{4+} to Ti^{3+}.

The luminescence spectra of the composite scintillators based on inorganic GSO:Ce, GPS:Ce, YSO:Ce or YAG:Ce grains not change with the dose D increase [3, 5, 6, 13, 14]. The intensity of luminescence increases, only for the time of regeneration of the quenching centres. As a result, after this time interval, the luminescence intensity should decrease due to the restoration of the quenching efficiency. The energy levels of these centres are located in the for-

bidden band of the energy spectrum of single-crystal grains. Consequently, these centres mainly affect on the light transmittance T. The modification of the luminescence parameters is due to the radiation damage of the grains of the scintillation material. However, self-restoration accompanies all the processes. As a result, we get a complex variation of the properties of the scintillator.

Consequently, as noted above, the change in the relative light output L_{rel} that occurs upon irradiation to different doses D should have a stochastic component. This component arises as a result of changes in the quenching efficiency of impurity centres and ions, as well as because of temporary damage to the luminescence centres in grains with their further rapid recovery. For higher doses D, the radiation resistance of the material will gradually decrease on the background of stochastic fluctuations associated with secondary processes, until the destruction effect for luminescence centres becomes a limiting factor.

In all composite scintillators based on inorganic grains (Al_2O_3:Ti, GSO:Ce, GPS:Ce, YSO:Ce or YAG:Ce) considered in the papers, the average L_{rel} has a weak tendency to decrease with increasing dose D. This fact indicates that when the dose of irradiation increases, the amount of destroyed luminescence centres increases, that is, it becomes more and more important. In the end, this will result in the L_{rel}-value being below 0.5.

Proceeding from the foregoing, we can conclude that it is impossible to determine from general considerations which of the mechanisms will be observed in a particular scintillator. This question needs to be analysed in each individual case. At the same time, the criterion associated with a change in the luminescence spectrum or its invariance with increasing dose can be considered proven. Also, these two processes can be observed simultaneously in one scintillation material, but one of them will prevail.

4. CONCLUSIONS

Possible processes and their mechanisms of radiative changes arising in scintillators during irradiation were proposed and studied. The influence of these processes on the radiation resistance was analysed. All crystalline scintillation materials contain defects that can play the role of luminescence or quenching centres. Irradiation destroys and changes not only the basis material, but also these centres. As a result, the luminescence intensity can grow for some time. In this case, the scintillator luminescence spectrum does not change its form at different doses D, since new luminescence centres are not generated. In this case, the scintillator luminescence spectrum does not change its shape at different doses D, since new luminescence centres are not generated. Another process involves the change of luminescence centres. This should lead to a modification of the spectral characteristics of the luminescence.

For organic scintillators observed change in the form of luminescence spectra under irradiation. The composite scintillators containing grains of inorganic crystals change form luminescence spectra can both an observer (Al_2O_3:Ti), or not (GSO:Ce, GPS:Ce, YSO:Ce and YAG: Ce). Therefore, in order to identify possible processes causing radiation changes in scintillators, it makes sense to conduct such spectral studies before and after irradiation.

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РАДИАЦИОННАЯ СТОЙКОСТЬ КОМПОЗИЦИОННЫХ СЦИНТИЛЛЯТОРОВ

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Проанализирована радиационная стойкость композиционных сцинтилляторов, содержащих гранулы органических и неорганических монокристаллов. Анализ основан на изучении композиционных сцинтилляторов, содержащих гранулы Gd_2SiO_5:Ce (GSO:Ce), $Gd_2Si_2O_7$:Ce (GPS:Ce), Al_2O_3:Ti, Y_2SiO_5:Ce (YSO:Ce) или $Y_3Al_5O_{12}$:Ce (YAG:Ce). Представлены результаты исследований и предлагаются возможные механизмы и процессы радиационных изменений, происходящих в сцинтилляторах под действием ионизирующего облучения. Было также представлено, как эти эффекты влияют на радиационную стойкость композиционных сцинтилляторов.

РАДІАЦІЙНА СТІЙКІСТЬ КОМПОЗИЦІЙНИХ СЦИНТИЛЯТОРІВ

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Проаналізовано радіаційну стійкість композиційних сцинтиляторів на основі гранул органічних та неорганічних монокристалів. Аналіз ґрунтується на вивченні композиційних сцинтиляторів, що містять гранули Gd_2SiO_5:Ce (GSO:Ce), $Gd_2Si_2O_7$:Ce (GPS:Ce), Al_2O_3:Ti, Y_2SiO_5:Ce (YSO:Ce) або $Y_3Al_5O_{12}$:Ce (YAG:Ce). Представлено результати досліджень та запропоновано можливі механізми та процеси радіаційних змін, що виникають у сцинтиляторах під дією іонізуючого опромінення. Було також представлено, як ці ефекти впливають на радіаційну стійкість композиційних сцинтиляторів.