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Radiation resistant composite scintillators based on Al_2O_3:Ti grains and their properties after irradiation

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ABSTRACT

The effect of irradiation on the scintillation light output, optical transmittance, and luminescent spectra of composite scintillators based on single crystal grains of Al_2O_3:Ti, is studied. The dielectric gel Sylgard-184 is the base and the binder for the grains inside the composite scintillator. The paper presents and analyses the results obtained for the scintillators exposed by 10 MeV electrons from the linear electron accelerator at room temperature. For exposure doses at least up to $D \sim 550$ Mrad when dose rate is 1500 Mrad/h and $D \sim 125$ Mrad when dose rate is 0.2 Mrad/h the composite scintillators are radiation-resistant.

1. Introduction

A large number of experiments performed at accelerators of high-energy particles 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.

In previous paper [2] we investigated the composite scintillators based on single crystal grains of cerium-doped gadolinium silicate Gd_2SiO_5:Ce (GSO) and gadolinium pyrosilicate $Gd_2Si_2O_7$:Ce (GPS) as the radiation-resistance materials. This paper is devoted to the study of the other promising material, namely, composite scintillators containing the single crystal grains of Al_2O_3:Ti. As in the previous paper, we will use the classical definition formulated by the Birks [3]. 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 $I(D)/I(0) \geq 1/2$. Such the criterion, we will use below both for the relative scintillation output, and for the optical transmittance of scintillation material.

As in previous work [2], we use the polydimethylsiloxane gel-composition that does not contain benzene rings. Therefore, 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 [2,4,5].

Composite scintillators have a number of advantages in comparison with other scintillation materials [2,6,7]:

(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 the 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 agglutinate the separate parts of composition scintillator in one uniform sample.
(3) It is possible to vary both the sample size and grain size independently.

The crystals based on Al_2O_3 and doped by Ce, Pb, Ca, Tl, Ti etc. are widely used. They are used as the modern laser materials, in the thermoluminescence investigations and in radiation protection

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dosimetry, they were proposed as candidate for Dark Matter detection techniques, etc. [8–18]. The light yields (relatively to CsI:Tl) of the crystals activated by Ce^{3+}, Pb^{+}, and Ti^{3+} are about 0.27–0.35, 0.12–0.17 and 0.3, respectively. The luminescence decay time of the crystals activated by Ce^{3+}, Pb^{+}, and Ti^{3+} are about 0.05, 1.4–1.5, and 3.4 μs, respectively [15].

The chemical composition of the crystal lattice (Al_2O_3 in our case) will determine the radiation resistance of the material, because the concentration of addition agents (Ce, Pb, Ca, Tl, etc.) is too small. The information about Al_2O_3:Ti crystals is the most extended and consistent one in comparison with other crystal of such the type. Therefore, this paper studies the properties of the composite scintillators containing the crystalline grains of Al_2O_3:Ti.

According to [18], Al_2O_3:Ti crystals have a broad visible absorption band at 490 nm, and a strong absorption UV edge begging near 300 nm with a shoulder at 240 nm, which is related to Ti^{3+}–Ti^{4+} pairs (see below). The authors of the work [18] reported that the excitation by the light with the wavelength λ_{ex} = 232 nm (as well as 266 nm) results in photoluminescence in the spectral band of λ = 400–410 nm. According to [8–14,16] it accompany by photoluminescence with λ = 420 nm, 425 nm and 430 nm, respectively. These bands of luminescence are often combined under one name "blue band". This luminescence appears under irradiation due to the formation centres containing Ti^{4+} ions [14,16]. The Ti^{3+} ions give the near infrared (IR) luminescence in the spectral region 650–800 nm with a maximum of around 750 nm [8–10,15–18]. It usually associated with radiative transition $^2E_2 \rightarrow {}^2T_2$ [16,18].

Assuming that the Ti^{3+} centres are effective electron donors, one can expect that the outer electron of Ti^{3+} will move to the oxygen vacancy and the spatially correlated complexes (Ti^{4+}–F^{+}) or (Ti^{4+}–F) will appear. Really, it becomes very likely that F^{+} and/or F-defect centres (an oxygen vacancy occupied by one or two electrons, respectively) take part in the excitation of the luminescence if we take into account the spatial proximity between the dopant and such the defect. In this case, the energy transfer from defect centres Ti^{4+} has to result in the blue luminescence, and the excitation spectrum of Ti^{4+} represents the superposition of the bands of charge transfer and F and/or F^{+}-centres excitation. In [16,18], the other possible scenario was proposed when the F^{+}-centres may cause the band with maximum λ = 420 nm [16], λ = 410 nm [18]. A physical model of such a process should include the ionization of Ti^{3+} ions and leads to the generation of the conduction electrons. The migration of the electrons in the end should lead to their capture by the F^{+}-centres. It can initiate the following recombination process [18]:

$$F^{+} + e^{-} \rightarrow F^{*} \rightarrow F + h\nu \text{ (420 nm [16], 410 nm [18])}, \quad (1)$$

where F^{*} is excited F-centre.

In [16] was shown that due to the existence of blue and IR luminescence the decay time of Al_2O_3:Ti crystals is described by several components and does not exceed one microsecond. Under photo-excitation ($\lambda_{ex} \sim$ 200 nm) the decay time for 290 nm luminescence at 300 K was equal to 138 ± 2 ns. The measurements of light output shown that it value rises from 1800 to 2550 photons/MeV with energy of X-ray photons from 16.9 to 122.1 keV [16].

The cited above papers studied the crystals containing up to 20 wt% of Ti. According to all the works the optimal concentration of Ti in a single crystal was about 0.08–0.12 wt%. The technology features of crystal growth and the technique of the measurements determine a significant spread of the luminescent characteristics of the crystals. However, all the papers argue that the common feature of all the crystals is the following. If the luminescence spectrum demonstrates the IR luminescence in the spectral region 650–800 nm with a maximum of around 750 nm, or blue luminescence then it means that such the luminescence is mainly causes by Ti^{3+} ions, or by the centres containing Ti^{4+} ions, respectively.

2. Experimental

2.1. Preparation of composite scintillators

In this work, we choose the polydimethylsiloxane dielectric gel Sylgard-184 [2,19] as the base composition and the binder for the grains of the composite scintillator. The properties of this gel composition we described in details in our previous work [20]. Original crystal Al_2O_3:Ti contain 0.12 ± 0.02 wt% Ti. Single crystals of Al_2O_3:Ti during the growth and their processing can crack forming the fragments those are useless for production the scintillation detector. To obtain the grains we grind these waste products as well. The necessary fraction of their sizes we select using a set of calibrated sieves in accordance with procedure that was described in [6,7].

To prepare the composite scintillator, firstly, we introduce grains in the main component of the gel composition. After that, we thoroughly mix it with the second component of the gel. To form the scintillator we put this mixture in a cylindrical glass container. After it fully solidification, we remove the composite scintillator from the container. We study the single-layer composite scintillators. Therefore, the grain size, which was about 0.3–0.5, 0.5–1 and 1.5–2.0 mm, determined a scintillator thickness. During the preparation of the gel composition, we placed a single-layer composite scintillator on a substrate from a radiation-resistant Mylar film [21].

2.2. Irradiation of the samples

As in the previous study [2], 10 MeV electrons from the linear electron accelerator of NSC "Kharkiv Institute of Physics and Technology" irradiate the scintillators at room temperature. During irradiation, the dose rate is almost uniform over the sample surface. The degree of heterogeneity is less than 5%. The plastic dosimeters Harwell Perpex 4034 and radiachromic film dosimeters FWT-60-00 determine the dose rate (see [22]). The measurement error was ±5%. These dosimeters have a limitation on the dose value. Therefore, each of the same dosimeters accumulates the dose for separate (relatively small) interval of the dose. The integral dose D, which a scintillator gains during an irradiation time, we determine by summation of the results of such the sequent measurements. We run the irradiation for two dose rates namely for 0.2 ± 0.01 Mrad/h (mainly photons of braking radiation) and 1500 ± 5 Mrad/h (an electron beam directly scans over the sample surface). It is important to note that here and below all the dose values D refer to the absorbed radiation dose in the water equivalent of the irradiated material.

2.3. Measurements of the relative light output

To calibrate the energy scale we use a set of gamma lines with the following energies 17 keV (^{241}Am), 32.7 keV (^{137}Cs), 41 keV (^{152}Eu), 59.6 keV (^{241}Am), 77.9 keV (^{152}Eu), 122.0 keV (^{152}Eu). The light output of the scintillators relative to gamma energy is linear within ±5%. To obtain the relative output of a composite scintillator we compare it scintillation signal with a signal from a reference single crystal. As the reference sample we used a single crystal of Al_2O_3:Ti with dimensions 30 × 25 × 1.5 mm. In such the measurements, the alpha particles from radionuclide ^{239}Pu source excite the scintillators.

2.4. Measurements of the optical transmittance

To perform the measurements of light transmittance T in the range from 300 to 700 nm we use Shimadzu-2450 spectrophotometer with the integrating sphere. The comparison channel of the spectrophotometer remained blank and the light flux in it is the same as the light flux falling on a sample in measuring channel. The inaccuracy of the calibration