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CREATION OF COMPOSITE SCINTILLATORS WITH A SHORT DECAY TIME

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To date, LuAG:Ce crystals are one of the most common scintillators, since they have been known for a long time and there are technologies for mass production of large-volume crystals. However, the scintillation properties of such crystals can be improved by creating mixed crystals with the replacement of some ions by others. In this work, composite scintillators based on the grown LuYAG:Ce inorganic crystals were produced. For the obtained samples, studies of optical transmission, luminescence, light output, and decay time were carried out. The optimal conditions and sizes of crystalline grains for the creation of composite scintillators have been determined.

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INTRODUCTION

The work is devoted to the search for new alternative scintillation materials for solving modern problems of radiation materials science and instrumentation. This is, first of all, obtaining scintillation materials for creating ionizing radiation detectors for conducting the latest experiments in high-energy physics, which requires registration of super-large fluxes of ionizing radiation with a short decay time.

A characteristic feature of experiments planned and carried out at new generation charged particle and ion accelerators (such as, for example, the Large Hadron Collider (LHC) at CERN) is the exposure of detectors to high doses of radiation (up to several tens and even hundreds of Mrad). The dose load on the subsystems of the detectors of experiments at the LHC is proportional to the peak luminosity of the collider, which by the end of the 2nd session of its operation (2018) had already reached 20 $fb^{-1}s^{-1}$, which was 2 times higher than the corresponding design value. Over the next few years, it is planned to increase the luminosity of the LHC several times more. By the end of the work of the accelerator complex (in ~20 years), it is planned to obtain a data array that will correspond to an integral luminosity of 3,000 fb^{-1}. At present, none of the detectors of the main experiments at the LHC meets such conditions, and work on their modernization is becoming extremely relevant. In this case, the search for and development of new scintillation materials characterized by high radiation resistance and fast response are of particular importance [1].

Given the above, interest in fast radiation-resistant scintillators is growing. YAG-based garnet crystals are considered to be an efficient material for laser, phosphor, and scintillation materials. It was also shown that YAG-based garnet crystals are radiation-resistant to doses of more than 100 Mrad [2, 3]. The Ce-activated garnet did not attract much attention. Everything changed after the discovery of effective solid multicomponent solutions of garnets in which the substitution of Al/Ga cations with a light output of up to 58,000 photons/MeV is used. This value approaches the theoretical limit given the band gap of these materials. The improvement in the light output was due to the disappearance of the electronic levels of the charge carrier traps. This occurred due to the modification of the electronic structure by cationic substitution.

Therefore, it should be investigated whether the capture of charge carriers can be reduced and the efficiency of complex crystals based on available YAG:Ce can be improved.

One possible solid solution with a modified band structure is LuYAG. Thus, LuYAG:Pr crystals have an improved light output, reaching 33,000 photons/MeV compared to 19,000 photons/MeV in LuAG:Pr, and a shorter decay time. Thus, LuYAG:Pr crystals have an improved light output, reaching 33,000 photons/MeV compared to 19,000 photons/MeV in LuAG:Pr, and a shorter decay time.

1. EXPERIMENTAL DETAILS

1.1. CRYSTAL GROWTH

LuYAG:Ce crystals were obtained by the Czochralski method using tungsten crucibles in a reducing surrounding. Until recently, the main method for obtaining rare-earth garnet crystals was their growth by the Czochralski method from iridium crucibles. The only alternatives were crucible-free methods or growth in molybdenum by the Stockbarger method [4].

Recently, the possibility of growing YAG:Ce and YAG crystals by the Czochralski method using tungsten crucibles was shown in [5, 6]. Thus, evaporating oxygen from the surface of the melt interacts with carbon contained in the atmosphere according to the following formulas:

$$O_2 + C = CO_2,$$
$$CO_2 + C = 2CO \text{ (at > 950 °C)}.$$

This prevents oxidation of the wolfram tooling.

Thus, in this work, LuYAG:Ce crystals were obtained by the Czochralski method using wolfram crucibles in a CO-containing atmosphere.

Crystals $(Lu_xY_{1-x})_3Al_5O_{12}$:1%Ce were grown. After that, the samples 5x5x2 mm were cut and polished from the obtained crystals. The samples were cut at the same height in order to cancel the influence of the uneven distribution of Ce.

Also, crystals of YAG:Ce,Ca with different concentrations of Ce and Ca were grown.

1.2. PREPARATION OF COMPOSITE SCINTILLATORS

In this work, we used the dielectric polydimethylsiloxane gel Sylgard-184 as the base material for composite scintillators [7]. Its optical properties were shown in the work [8].

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 the adjunction of the second component, we carefully mix the gel composition. Finally, we introduce this gel composition into a forming container, in which it is left up to its complete polymerization. As the result, the scintillator is obtained and can be taken from the forming container. To be able to compare the results of research on composite scintillators with the results obtained for the relevant single crystals, it was decided to choose the same linear size, namely 5x5x2 mm [9]. In this way, composite scintillators based on LuYAG:Ce and YAG:Ce,Ca crystals with a grain size of 0.3-0.5, 0.5-1.0, 1.0-1.5, 1.5…2.0 mm were created.

1.3. MEASUREMENTS OF SCINTILLATION LIGHT YIELD AND DECAY TIME

Light yield and energy resolution were measured under irradiation with a ^{137}Cs γ-source at the 662 keV energy. As a photodetector R1307 Hamamatsu PMT ran at 800…960 V with a linear dynode voltage divider was used. The photomultiplier tube (PMT) output was connected to a charge-sensitive preamplifier BUS 2−94 and then to a BUI-3K shaping amplifier. The signal from the preamplifier was formed by a shaping amplifier with a 2 μs shaping time. To collect the whole scintillation light, the samples and the open part of PMT photocathode were covered with a Teflon reflector. No optical contact between the samples and the PMT window was provided. For absolute light yield determination, the light yields were compared to $Bi_4Ge_3O_{12}$ (BGO) etalon with a light yield of 8,600 phot/MeV produced at ISMA. The energy resolution of ^{137}Cs 662 keV peak was determined by approximation of the obtained pulse height spectra with the Gaussian function.

Scintillation decay curves were measured with Hamamatsu R6231 PMT and excitation with 662 keV gamma ray from ^{137}Cs source. A signal from the PMT anode was fed to the 50 Ω terminated input of the Rigol DS6064 oscilloscope.

1.4. MEASUREMENTS OF LIGHT 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 the measuring channel. The inaccuracy of the calibration was limited by 0.5%. The value of *T* was calculated as follows:

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

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

1.5. MEASUREMENTS OF LUMINESCENCE AND EXCITATION SPECTRA

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

2. RESULTS AND DISCUSSION

First of all, measurements of the scintillation yield of samples of different chemical compositions were carried out, but they were taken from the same part of the crystal. The results of measuring the light output (*L*) and resolution (*R*) for studied samples of single crystals are shown in Table 1. At a value of 100% light output, the light output of the BGO crystal was chosen.

Table 1

Relative light output L_{rel} and resolution *R* of samples based on $(Lu_xY_{1-x})_3Al_5O_{12}$:Ce

Sample	Without optical contact		With optical contact	
	L_{rel}, %	*R*, %	L_{rel},, %	*R*, %
x = 0.25	233	14.7	217.9	12
x = 0.5	228.4	14.2	226.1	13.1
x = 0.75	194.2	13.7	192.7	12.5
x = 1	252.3	13.3	220.1	12.2

Table 1 shows that the light output decreases with increasing lutetium concentration in the crystal. In this case, high light output is observed for samples that do not contain yttrium in their composition. Also, samples of this composition show the high efficiency of registration of ionizing radiation.

The results of measurements of optical transmittance, luminescence, light output, and decay time for samples of single crystals LuYAG:Ce and YAG:Ce,Ca was shown in our previous work [9]. For example, the scintillation decay time of single crystals LuYAG:Ce with different ratios of Y/Lu was 190, 181, 206, and 70 ns with increasing lutetium concentration in the crystal from 25 to 100%, respectively [9].

The results of measuring the light output (*L*) and resolution (*R*) for studied samples of composite scintillators are shown in Table 2.

Table 2

Relative light output L_{rel} and resolution R of samples composite scintillators based on $(Lu_xY_{1-x})_3Al_5O_{12}$:Ce

Sample	Without optical contact		With optical contact	
	L_{rel}, %	*R, %*	L_{rel}, %	*R, %*
x = 0.25	196.5	19.4	193.8	13.4
x = 0.5	202.9	15.6	196.0	16.0
x = 0.75	169.5	16	171.5	14.3
x = 1	221.2	16.5	199.7	12.5

As can be seen from Table 2, the results of measuring the light output of composite scintillators based on $(Lu_xY_{1-x})_3Al_5O_{12}$:Ce grains showed that the light output of these samples is very high. The results of the measurements with optical contact show slightly better resolution than without. Compared to single crystal samples of the same chemical composition, composite scintillators have a lower light output value. It is about 90% of the L_{rel}-value of single crystals of the same size. It is a good result and composite scintillators can be used as an alternative to single crystals.

At the same time, the decay time of composite scintillators almost did not change compared to single crystals, and the difference in values is within the measurement error (Table 3).

Table 3

The decay time of composite scintillators based on $(Lu_xY_{1-x})_3Al_5O_{12}$:Ce

Sample	Decay time, ns
$(Lu_{0.25}Y_{0.75})_3Al_5O_{12}$:Ce	192
$(Lu_{0.5}Y_{0.5})_3Al_5O_{12}$:Ce	182
$(Lu_{0.75}Y_{0.25})_3Al_5O_{12}$:Ce	208
$Lu_3Al_5O_{12}$:Ce	72

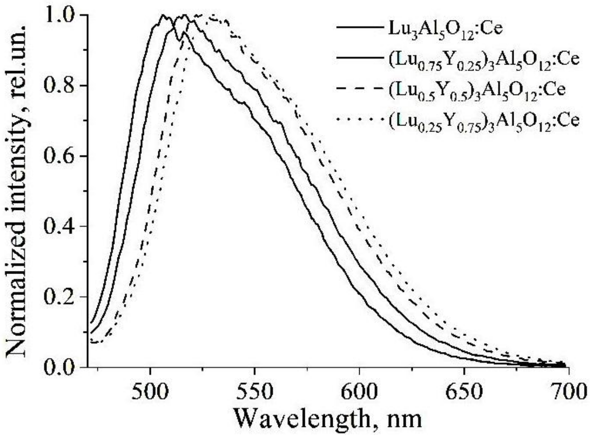
The luminescence and absorption spectra of these samples of composite scintillators have the same shape as single crystals, no additional lines or shifts in the spectra are observed. The value of the optical transmission of light in the region of luminescence of composite scintillators coincides with the transmission of similar samples of single crystals. These spectra (luminescence, light absorption, and transmittance), as examples, for single crystals can see in our previous work [9].

Figure shows normalized photoluminescence spectra of composite scintillators based on grains (size 0.3…0.5 mm) of single crystals $(Lu_xY_{1-x})_3Al_5O_{12}$:Ce with an excitation wavelength of 460 nm.

Composite scintillators from grains of different sizes have almost identical spectra and dependencies.

In both, single crystals and composite scintillators, with increasing Y content, a shift of the spectrum to a longer wavelength region, i.e., to the red region, is observed. The created composite scintillators have luminescence in the same wavelengths as single crystals.

Similar dependences were also observed for samples of composite scintillators based on grains YAG:Ce,Ca.



Comparison of the normalized luminescence spectra of composite scintillators based on grains $(Lu_xY_{1-x})_3Al_5O_{12}$:Ce (grains size 0.3…0.5 nm) with excitation of the light wavelength of 460 nm

CONCLUSIONS

1. LuYAG:Ce mixed crystals with different Y/Lu ratios and YAG:Ce,Ca crystals with different Ce and Ca concentrations were grown.
2. Composite scintillators based on LuYAG:Ce and YAG:Ce,Ca with a linear size of 5x5x2 mm were created.
3. Optical transmittance, luminescence, light output, and decay time studies for the obtained samples have been carried out.
4. The light output of composite scintillators (5x5x2 mm) based on LuYAG:Ce and YAG:Ce,Ca crystals with a grain size of 0.3…0.5 mm is within 85…95% of single crystals of the same chemical composition and linear size. The decay time is almost unchanged, within the measurement error. The luminescence and absorption spectra of these samples of composite scintillators have the same shape as single crystals, no additional lines or shifts in the spectra are observed. The value of the optical transmission of light in the region of luminescence of composite scintillators coincides with the transmission of similar samples of single crystals.
5. The optimum sizes of grains for creating composite scintillators based on LuYAG:Ce and YAG:Ce,Ca crystals are 0.3…2.0 mm.

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СТВОРЕННЯ КОМПОЗИЦІЙНИХ СЦИНТИЛЯТОРІВ ІЗ МАЛИМ ЧАСОМ ВИСВІТЛЮВАННЯ

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На сьогодні кристали LuAG:Ce є одними з найпоширеніших сцинтиляторів, оскільки вони відомі вже багато часу та існують технології масового виробництва великих об'ємних кристалів. Однак сцинтиляційні властивості таких кристалів можна покращити шляхом створення змішаних кристалів із заміщенням одних іонів іншими. У роботі були створені композиційні сцинтилятори на основі вирощених неорганічних кристалів LuYAG:Ce. Для отриманих зразків були проведені дослідження оптичного пропускання, люмінесценції, світлового виходу та часу згасання. Встановлено оптимальні умови та розміри кристалічних гранул для створення композиційних сцинтиляторів.