DETECTORS AND NUCLEAR RADIATION DETECTION

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DEVELOPMENT OF FAST SCINTILLATORS FOR HIGH-ENERGY PHYSICS PROBLEMS

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The scintillation materials are currently relevant, and developing the technology to create them is a priority. It is related to the search for new alternative scintillation materials and the development of detectors based on them in solving a wide range of modern problems of radiation materials science and instrument engineering, such as conducting the latest experiments in high-energy physics, which requires the registration of high doses of ionizing radiation with a short decay time. This work presents some features of the development of fast perovskite scintillators for high-energy physics applications.

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INTRODUCTION

Nowadays, the development of new scintillator materials with short decay times is more urgent than ever. This is due to the increased demand for such materials in high-energy physics. The increasing power of accelerators such as the LHC-HL (CERN, Sweden), where the planned particle collision energy will reach 7 TeV, requires the use of faster scintillator materials in new types of detectors. Cost is also a key parameter for scintillators in such applications, as large-scale production of detectors based on them is planned.

In recent years, the technology for growing YAG:Ce scintillator crystals has improved, and their characteristics are approaching the required level. The main advantage of these garnets is that they can be grown in inexpensive tungsten crucibles, significantly reducing their cost. For example, the cost of a tungsten crucible is 50–100 times lower than that of an iridium one. The lower cost of tungsten ones allows for the production of crucibles with thicker walls, which also increases the number of growth processes in these crucibles and, consequently, their service life.

One option for scintillator materials that can be produced using a similar, lower cost technology using tungsten/molybdenum crucibles is also YAP:Ce. The decay time of undoped YAP:Ce is 25 ns, and its density is 5.4 g/cm³. Its structure can also be modified by adding appropriate rare-earth oxides, which can form the $REAlO_3$ structure and form a solid solution (mixed crystal). This can increase the crystal's density and potentially improve its scintillation properties, similar to GAGG.

However, growing bulk perovskite crystals is somewhat more challenging than growing garnets, due to their tendency to crack upon cooling after growth. This is due to their orthorhombic lattice, which causes anisotropy in properties along different crystallographic directions. Heterostructured materials, particularly composites based on perovskite single crystal grains, may offer a solution to this problem. The creation of a composite material will enable the production of large-scale scintillators from perovskite crystal fragments, which may possess properties similar to those of single crystals.

Recently, the practical feasibility of growing YAG crystals using the Czochralski method using tungsten crucibles was demonstrated in [1], meaning that YAP crystals can now be grown using a similar method. Thus, evaporating oxygen from the melt surface reacts with carbon present in the atmosphere according to the following formulas:

$$O_2+C=CO_2,$$

$$CO_2+C=2CO \text{ (at } >950\ ^\circ C).$$

This prevents oxidation of the tungsten crucible.

1. EXPERIMENTAL

1.1. OBTAINING OF CRYSTALLINE INGOTS

Crystals of perovskites were grown in an "Oxide" growth chamber. The crystal extraction rate was 1.8…3.0 mm/h at 5…10 rpm. Three metal rods with a capillary inside were used as seeds, as this increases the likelihood of forming seeds with optimal growth orientation. After separation from the melt, rotation was stopped, and annealing began. The temperature was decreased linearly from the melting point to room temperature. The grown crystal was then removed from the growth chamber.

The obtained crystal ingots are shown in Fig. 1.

1.2. EXPERIMENTAL METHODS

Light output measurements were performed using the standard method [2], analyzing scintillation amplitude spectra. The 9208A photomultiplier tube was used as a photodetector [3]. We used the following types of ionizing radiations: alpha particles from ^{239}Pu with energy E_α = 5.15 MeV, conversion electrons from ^{137}Cs with energy E_β = 0.622 MeV.

The optical transmittance of samples of scintillators was measured on a Shimadzu UV-2450 spectrophotometer. The absolute error in determining the wavelength was 0.4 nm, and the measurement of spectral transmittances was 0.6% [4].



Fig. 1. Crystals of $YAlO_3$:Ce (top), $Y_{0.5}Gd_{0.5}AlO_3$:Ce (center), and $GdAlO_3$:Ce (bottom) after annealing

The luminescence characteristics of the studied samples were measured using a Lumina spectrofluorimeter (Thermo Scientific, USA), equipped with a continuous-wave xenon lamp as the excitation source and a photomultiplier tube (PMT) as the detector. The device allows recording photoluminescence and excitation spectra over a wide spectral range in the ultraviolet and visible regions.

The surface morphology of perovskite single crystals was studied using a Micromed POLAR 3 microscope in reflected light mode, without a polarizer or analyzer, with conventional bright-field illumination. Image recording was performed with a digital camera mounted on the microscope (UCMOS05100KPA-U-NA-N-C-SQ-NA).

To study the X-ray luminescence of scintillation materials, the following were used: Isovolt Titan 160 X-ray source (XRS) from a tri-energy scanner, a Hamamatsu spectrometer, and a personal computer running the Hamamatsu spectrometer software.

2. RESULTS OF STUDIES AND THEIR ANALYSIS

Figs. 2.1-2.3, for example, show tenfold magnification images of the surfaces of single crystals of the rare-earth perovskites YAP, YGAP, and GAP, respectively. The area on the surface of the single-crystal ingot was chosen so that the zone on which the lenses focus did not have visual cracks.

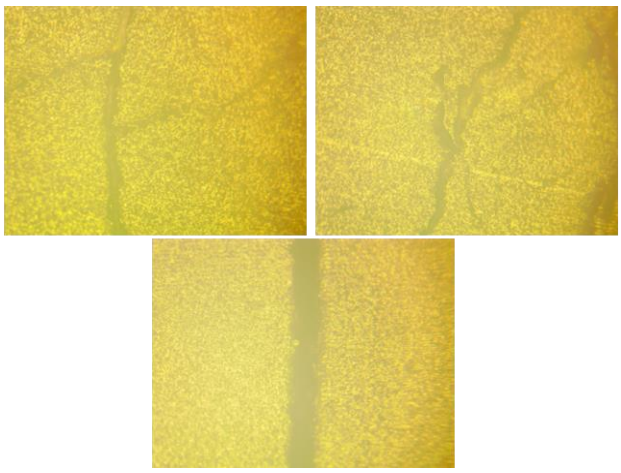


Fig. 2.1. Photographs of the surface of a single crystal of $YAlO_3$:1%Ce (YAP:1%Ce) under a microscope before annealing (magnification x10)

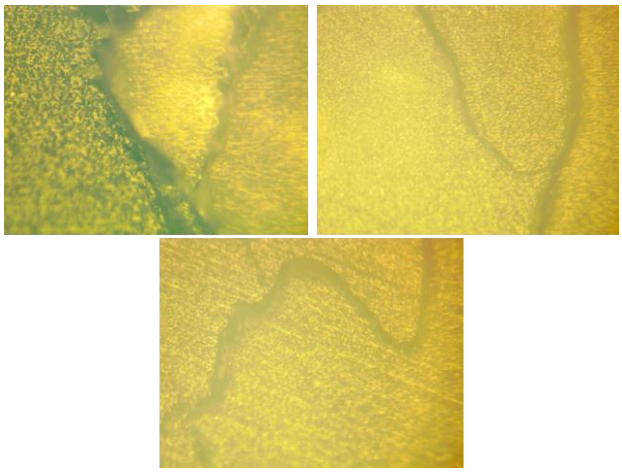


Fig. 2.2. Photographs of the surface of a single crystal of $Y_{0.5}Gd_{0.5}AlO_3$:1%Ce (YGAP:1%Ce) under a microscope before annealing (magnification x10)

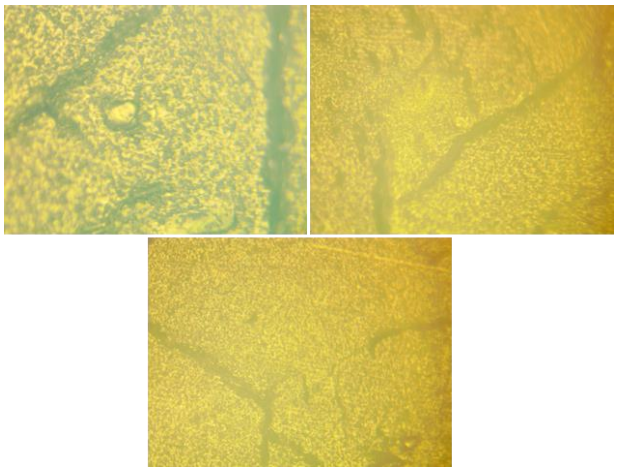


Fig. 2.3. Photographs of the surface of a single crystal of $GdAlO_3$:1%Ce (GAP:1%Ce) under a microscope before annealing (magnification x10)

Analysis of Figs. 2.1-2.3, which show images of crystal samples under a microscope, indicates the presence of microcracks in the crystal, which, during mechanical processing, can lead to further cracking of the single crystal into smaller fragments. At the same time, scattering and refraction of light can occur at the

boundaries of such cracks, leading to a decrease in optical transmission and, with it, a decrease in the scintillator's light output. It should be noted that after annealing, the number of such cracks increases. This makes it advisable to use them to further create composite scintillators based on perovskite crystals.

Also, the excitation and luminescence spectra of single crystal samples were investigated in the work. Perovskite single crystals exhibit luminescence in the 330…400 nm range, which overlaps with the maximum sensitivity range of many commercially available PMT. The luminescence peak maxima are in the range of 355…360 nm for YAP and YGAP crystals and 335 and 355 nm for GAP. Perovskite single crystals after annealing also have luminescence in the region of 330…400 nm, however, the maxima of the luminescence peaks shift to a longer wavelength region. Thus, the maxima of the luminescence peaks for YAP, YGAP, and GAP crystals are at 370, 365, and 355 nm, respectively.

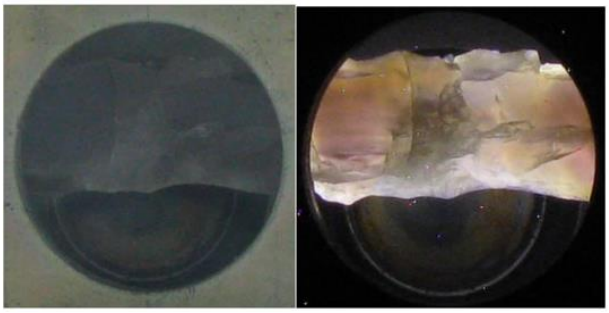


Fig. 3.1. Photographs of rare earth perovskite crystals $YAlO_3$ (YAP) activated with 1% Ce in a stand for X-ray luminescence research (left) and under the influence of X-ray radiation (right)



Fig. 3.2. Photographs of rare earth perovskite crystals $Y_{0.5}Gd_{0.5}AlO_3$ (YGAP) activated with 1% Ce in a stand for X-ray luminescence research (left) and under the influence of X-ray radiation (right)

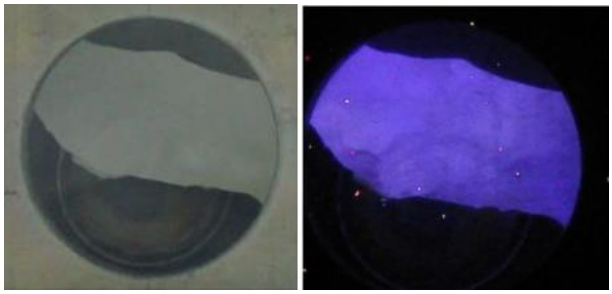


Fig. 3.3. Photographs of rare earth perovskite crystals $GdAlO_3$ (GAP) activated with 1% Ce in a stand for X-ray luminescence research (left) and under the influence of X-ray radiation (right)

Figs. 3.1-3.3 show photographs of perovskite single crystal samples placed in a stand for X-ray luminescence research and exposed to X-ray radiation.

Figs. 4.1-4.3 show the X-ray luminescence spectra of rare-earth element perovskite crystals.

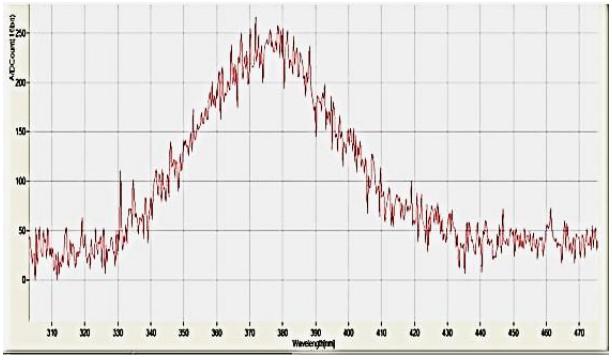


Fig. 4.1. X-ray luminescence spectrum of rare-earth perovskite crystals $YAlO_3$ (YAP) activated with 1% Ce

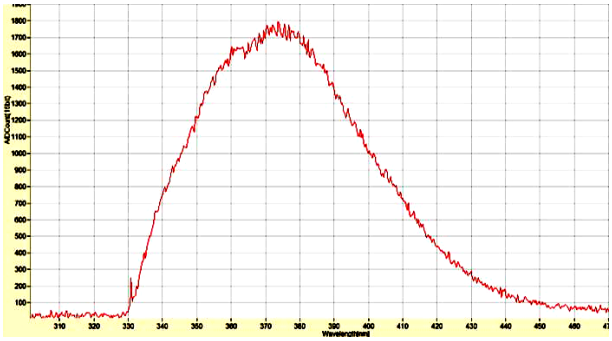


Fig. 4.2. X-ray luminescence spectrum of rare-earth perovskite crystals $Y_{0.5}Gd_{0.5}AlO_3$ (YGAP) activated with 1% Ce

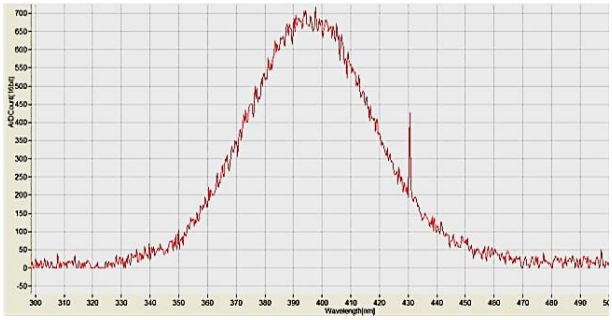


Fig. 4.3. X-ray luminescence spectrum of rare-earth perovskite crystals $GdAlO_3$ (GAP) activated with 1% Ce

As shown in Figs. 4.1-4.3, the X-ray luminescence of the obtained crystals, compared with that upon optical excitation, is in a longer wavelength region. However, these X-ray luminescence limits are still within the maximum sensitivity of most commercially available PMT. However, the maxima are located around 375 nm for YAP and YGAP, and for GAP – 395 nm.

After growth and annealing, the scintillation characteristics of the resulting crystals were studied. The light output was measured by irradiating samples of varying crystalline compositions with the sources and by the method specified in Section 1.2.

The most interesting results were obtained for YAP:Ce and YGAP:Ce samples. They are shown below.

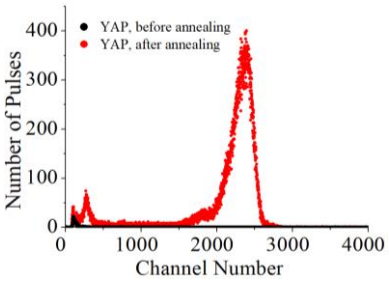


Fig. 5. Scintillation amplitude spectra for the unannealed (black points) and annealed (red points) YAP:Ce crystals. Irradiated with ^{239}Pu

The data presented in Fig. 5 clearly demonstrate that crystals measured immediately after growth and cooling exhibit close to zero light output. This confirms the necessity of annealing to obtain a scintillator crystal.

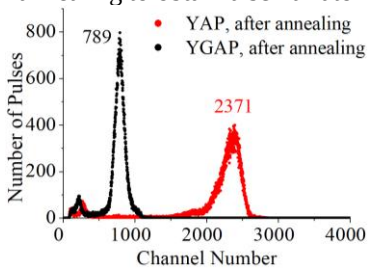


Fig. 6. Scintillation amplitude spectra for the annealed YGAP:Ce (black points) and annealed YAP:Ce (red points) crystals. Irradiated with ^{239}Pu

From Fig. 6, it can be seen that the YAP:Ce sample has a light output three times higher than that of the YGAP:Ce sample of the same geometry. This is because of the F-levels of Gd^{3+}, competing with the Ce^{3+} levels, are capable of creating traps that partially absorb the energy of ionizing radiation, reducing the number of photons produced.

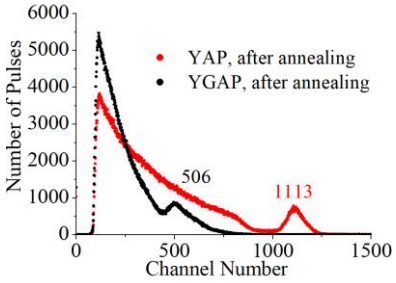


Fig. 7. Scintillation amplitude spectra for the annealed YGAP:Ce (black points) and annealed YAP:Ce (red points) crystals. Irradiated with ^{137}Cs

From Fig. 7 it can be seen that the YAP:Ce sample exhibits a superior light output compared to the YGAP:Ce sample of the same geometry. Furthermore, when processing the photopeak data, the position of which is indicated in the figure, the value for YAP:Ce is twice that of YGAP:Ce. This difference indicates that YAP:Ce is more sensitive to gamma radiation than YGAP:Ce, and may, to some extent, indicate that YAP:Ce is less sensitive to gamma radiation than to alpha particles.

CONCLUSIONS

1. Technological aspects for growing single crystals of rare-earth perovskites were worked out, and $YAlO_3$ (YAP), $Y_{0.5}Gd_{0.5}AlO_3$ (YGAP), and $GdAlO_3$ (GAP) crystals activated with 1% Ce were grown.
2. Preliminary studies of the optical transmittance, luminescence, light output, and decay time of the grown crystal samples were conducted.
3. The light output of unannealed crystals was below the detection limit of the measuring setup. Annealing the crystals for 48 h at 1400 °C in air resulted in an increase in the light output of the rare-earth perovskite crystals.
4. The light output of the annealed YAP:Ce crystals is 2–2.5 times higher than that of annealed YGAP:Ce crystals when irradiated with gamma radiation (^{137}Cs) and 3 times higher when irradiated with alpha particles (^{239}Pu). This difference indicates that YAP:Ce is more sensitive to gamma radiation than YGAP:Ce and may, to some extent, indicate that YAP:Ce is less sensitive to gamma radiation than to alpha particles.
5. It has been experimentally confirmed that rare-earth perovskite single-crystal ingots are prone to cracking upon cooling, both after completion of the growth stage and after annealing. This makes them suitable for creating and characterizing composite scintillators in the next stage of the work.

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РОЗРОБЛЕННЯ ШВИДКИХ КОМПОЗИЦІЙНИХ СЦИНТИЛЯТОРІВ НА ОСНОВІ ПЕРОВСКІТІВ ДЛЯ ЗАДАЧ ФІЗИКИ ВИСОКИХ ЕНЕРГІЙ

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Сцинтиляційні матеріали наразі є актуальними, а розробка технології їх створення є пріоритетним завданням. Це пов'язано з пошуком нових альтернативних сцинтиляційних матеріалів та розробкою детекторів на їх основі для вирішення широкого кола сучасних проблем радіаційного матеріалознавства та приладобудування, таких як проведення новітніх експериментів у фізиці високих енергій, що вимагає реєстрації високих доз іонізуючого випромінювання з коротким часом загасання. Наведено деякі особливості розробки швидких перовскітних сцинтиляторів для застосувань у фізиці високих енергій.