Sensors

# Investigation of gamma-ray sensitivity of YAG:Ce based scintillation structures

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**Abstract.** Ionizing radiation is widely used nowadays for diagnosing and probing a wide range of objects due to the high reliability and quality of the results obtained in such research. Use of highly sensitive ionizing radiation sensors enables the reduction of the radiation dose involved in the research. Moreover, sensitive systems for monitoring environmental parameters may be also created based on such sensors. In this work, the efficiency of a low density radiation detector with the composite scintillation structure based on powdery YAG:Ce phosphor as the converting coating of photosensitive detector was investigated. The possibility to detect gamma radiation from the <sup>241</sup>Am and <sup>137</sup>Cs based sources by the ionizing radiation detector comprising YAG:Ce<sup>3+</sup>composite converting scintillation structure and micropixel avalanche photodiode (MAPD) was found. The number of detected gamma rays emitted by the <sup>241</sup>Am source was shown to increase linearly with the thickness of the composite converting scintillation structure. The thickness of the composite converting scintillation structure of 495 μm was found to enable registration of gamma-rays with the energies in the range of 26 to 662 keV.

**Keywords:** ionizing radiation, scintillation structure, YAG:Ce phosphor, low density radiation detector.

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## 1. Introduction

Diagnostic equipment that uses ionizing radiation is increasingly exploited because of the high reliability and quality of the data provided [1]. Application of high sensitivity ionizing radiation sensors in such devices enables reduction of the radiation dose used in research, which is of great importance and highly demanded today [2]. Moreover, application of highly sensitive ionizing radiation sensors in environmental sensing systems allows detection of traces of hazardous radioactive substances, which is particularly important for control of their use, movement and compliance with the storage conditions [3].

One way to detect ionizing radiation is to convert the radiation energy into the radiation in the visible range, enabling its detection by a wider range of optoelectronic devices. Most radiation detectors consist of a scintillation structure coupled to an optical detector (photographic emulsion film, photocathode, photodiode, *etc.*). Scintillation structures [4, 5] or structures incorporating scintillation materials, in particular, based on phosphors [6], are used in radiation detectors as converters of ionizing radiation into visible light.

One type of highly sensitive detectors of optical radiation is micropixel avalanche photodiode (MAPD), which has maximum absorption in the green region [7–14]. Combined with a scintillation composite structure based on phosphors, it can be used as a highly sensitive detector of ionizing radiation.

The main criteria to be taken into account when evaluating the performance of scintillation or syndicating

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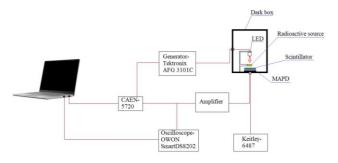
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structures for use in ionizing radiation detectors are as follows: quantum efficiency of detection and absorption, intrinsic efficiency of converting ionizing radiation into light, emission efficiency, emitted light spectrum and spectral compatibility with traditional optical detectors [15].

In recent years, cerium-doped yttrium aluminium (Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce or YAG:Ce) and gallium gadolinium (Gd<sub>3</sub>Al<sub>2</sub>Ga<sub>3</sub>O<sub>12</sub>:Ce or GAG:Ce) garnets have been widely used as the basic components of photophosphors [16]. Technological processes to produce such materials have been well established as they are actively used in light emitting diodes (LEDs) [17]. Quantum efficiency of light-converting structures based on the mentioned materials has almost reached the theoretical limit enabling creation of a new generation of high power LED lighting devices [18–21]. Use of such phosphors in white LEDs involves conversion of the narrowband short-wavelength radiation (360...460 nm) [22-24] into the broadband radiation (500...700 nm) to produce the resulting high-quality white light [26, 27]. In the ionizing radiation detectors, radiation is absorbed by the phosphor and then re-emitted having the wavelengths in the spectral range of 500...700 nm, which is typical for such phosphors [28].

Another attractivity of sensitive sensors based on garnet phosphors activated by  $Ce^{3+}$  ions is their fast response (~70 ns) [29]. Low response time is provided by the energy levels of  $Ce^{3+}$  ions including the configuration of the ground state  $4f^{1}$  containing one electron as well as the configuration of the energy level  $5f^{1}$  containing one electron and the configuration of the excited state  $5d^{1}$  split into five levels due to electric field effects in the crystal. The mechanism of the rapid scintillation decay consists in an allowed emissive transition taking place after excitation of the allowed state  $5d \rightarrow 4f$ .

YAG:Ce exhibits bright light emission with a broad peak in the green spectral region caused by strong crystal field effect. This property is of particular interest for application of this material with light emission detectors having similar spectral characteristics of maximum efficiency, such as MAPD considered earlier. Also important is the possibility of using this phosphor to manufacture the composite material with a transparent backing, which would allow the phosphor micro crystals in such composite to be suspended and evenly distributed over its volume thus providing maximum absorption and free over-emission.



**Fig. 1.** Schematic diagram of the experimental set-up used to measure the detector characteristics.

The purpose of this work is to investigate the efficiency of a low-density radiation detector when using a composite scintillation structure based on powdered YAG:Ce phosphor as the converting coating of a photosensitive receiver.

# 2. Experimental methodology and fabrication of scintillation structures

 $Y_3Al_5O_{12}$ :Ce phosphor obtained by the one of the simplest and the most technologically advanced methods based on solid-phase synthesis [30] was used as a scintillation material.  $Y_xCe_{1-x}Al_5O_{12}$  (x=0.03) was obtained from high purity  $Al_2O_3$  (99.99%),  $Y_2O_3$  (99.99%) and  $CeO_2$  (99.9%) precursors. The pre-calcined precursors were milled in a Retsch PM 100 planetary mill at different speeds to achieve the average particle size of 20 μm. Solid phase synthesis was carried out for 6 h at 1600 °C in a reducing medium inside the Nabertherm LHT 04/17 muffle furnace. After the synthesis, the phosphor was reground using a planetary mill and sieved through a 20 μm sieve.

The resulting powdered phosphor was placed in a binder to produce the scintillation structure in the form of films best suited for technological applications together with the selected photodiode type. To obtain a homogeneous suspension, to remove gases adsorbed on the phosphor microparticles and to ensure uniform distribution of the suspension components in the composite film, a degassing unit and a special technique for preparing the phosphor suspension were used. Twocomponent epoxy resin was used as the binder, which enabled precise control of the YAG:Ce composite film thickness. Compared to other binders (silicone, polymethylmethacrylate etc.), epoxy resin does not change the geometry of the sample during polymerisation. Moreover, various forms of the final scintillation structure are also possible.

Composite films with the thicknesses of 55, 150, 255, 340, 450, and 495  $\mu m$  and the weight concentration of the  $Y_3Al_5O_{12}$ :Ce phosphor of 20% were fabricated for use as the scintillation structures of ionizing radiation detectors.

Performance of the MAPD type Si photomultiplier (SiPM) photodiode combined with the YAG based scintillation structure was studied at the temperature of 23 °C. A schematic diagram of the experimental set-up is presented in Fig. 1. The Keithley 6487 picoammeter voltage source served to power the tested samples. The gain and the position of a single photon absorption event by the MAPD were recorded using a light emitting diode  $(\lambda = 650 \text{ nm})$  fixed at the distance of 50 mm from the photodiode [31, 32]. A Tektronix AFG 3101C generator was used to power the LED. The signals from the MAPD were fed into an amplifier. They were amplified with a gain of 55 and a bandwidth of 50 MHz. A CAEN DT5720 (charge of ADC cannel is 40 fC) digitizer integrated the output signal from the MAPD. <sup>241</sup>Am and <sup>137</sup>Cs were used as the radioactive sources.

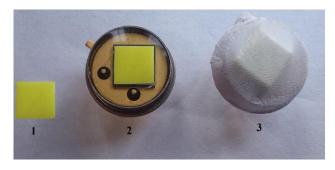
**Table.** Parameters of the MAPD photodiode [54, 55, 61].

Manufacturer	Zecotek Photonics
Туре	MAPD-3NM-II
Active area	3.7 × 3.7 mm
Total pixel number	6 1000 pixels
Photon detection efficency	>30 %
Gain	$\sim 2.5 \times 10^5$
Spectral response	300900 nm (max at 450 nm)
Operation voltage range	5456 V
Capacitance/channel	2 480/155 pF

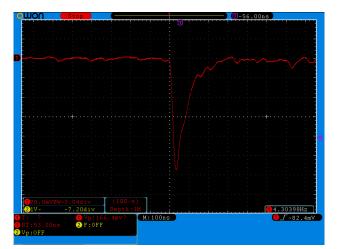
The MAPD type SiPM used here operates in the Geiger mode. The MAPD contains a Si substrate with n-type conductivity. Two p-type epitaxial Si layers are grown on this substrate [12, 33-36]. The device also contains a dense matrix of independent  $n^+$ -type pixels buried deep within the epitaxial layers. The pixels are connected in parallel and operate in the counting mode like a Geiger counter. The amplitude of the signal received from the MAPD corresponds to the number of triggered pixels. The number of fired pixels depends on the number of scintillation photons produced by the gamma rays. Therefore, increasing the pixel density is very important for the SiPM [35, 36]. The used design of the MAPD device ensures wide linearity range of photo response due to the high pixel density within the sensitive area [37-39]. A detailed description of the design and operation of this device can be found in [12, 33, 34, 40].

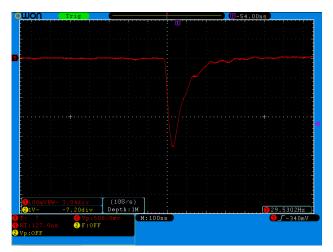
The parameters of the used MAPD photodiode are listed in Table.

The YAG:Ce based scintillation structure was used to convert gamma photons into visible light photons with the wavelength of 525 nm. YAG material provides a full light output of 18000 photons/MeV. The YAG:Ce based scintillation structure was covered by one layer of a reflective white Teflon tape (thickness of 200  $\mu m$ ) except for the bottom. This structure was coupled to the MAPD



**Fig. 2.** I – photo of the YAG:Ce based scintillation structure (3×3×0.255 mm), 2 – photo of the MAPD-3NM in combination with YAG:Ce, and 3 – photo of the MAPD-3NM + YAG:Ce covered by one layer of a reflective white Teflon tape.





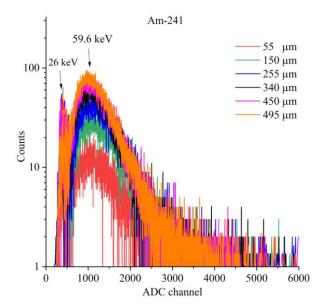
**Fig. 3.** The signals from the amplifier output for the cases of <sup>241</sup>Am (top) and <sup>137</sup>Cs (bottom) sources.

detector with silicone optical grease. The dimensions of the used YAG were 3 mm (length)  $\times$  3 mm (width). The thickness of the scintillation structure was 55, 150, 255, 395, and 495  $\mu$ m. The photo of the YAG:Ce based scintillation structure (0.255 mm) and its combination with the MAPD is shown in Fig. 2.

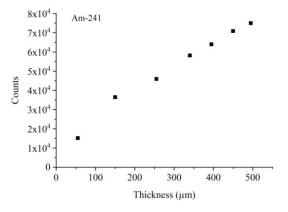
The MAPD+YAG:Ce based scintillation structure (495  $\mu$ m) amplified the output signals from the OWON Smart SD8202 Oscilloscope. Fig. 3 shows the signals from the amplifier output for the cases of  $^{241}Am$  (top) and  $^{137}Cs$  (bottom) sources. As expected, the length of each signal was 300 ns. The amplitude of the signal from  $^{241}Am$  was 116 mV, while the amplitude of the signal from  $^{137}Cs$  was 460 mV.

### 3. Experimental results and discussion

The numbers of the detected gamma rays by the MAPD with different thicknesses of the scintillation structure (55, 150, 255, 340, 395, 450, and 495  $\mu$ m) are presented in Fig. 4. As can be seen from this figure, the number of the detected gamma rays emitted by the <sup>241</sup>Am source increases with the thickness of the scintillation structure.



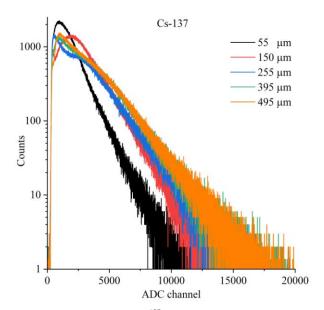
**Fig. 4.** Energy spectra of the  $^{241}$ Am source measured by the MAPD + YAG:Ce based scintillation structure at 22 °C. (Color online)



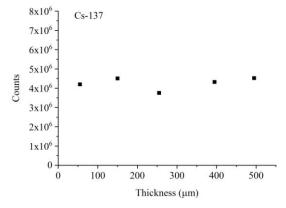
**Fig. 5.** Number of the detected gamma rays by the MAPD with different thicknesses of the YAG:Ce based scintillation structure (55, 150, 255, 340, 395, 450, and 495  $\mu$ m).

The number of detected MAPD gamma rays with different thicknesses of scintillation structure (55, 150, 255, 340, 395, 450, and 495  $\mu m)$  is presented in Fig. 5. The number of detected gamma rays emitted by the  $^{41}Am$  source increases with the thickness of the scintillation structure.

Increase of the number of detected gamma rays in the scintillation structure up to complete absorption of the quanta with the energies of 26 and 59.6 keV is caused by its increased thickness (up to 495  $\mu m$ ). The detection efficiency of the detector seems to increase with the thickness of the YAG:Ce based scintillation structure. As the thickness of the scintillation structure increases, some portion of scintillation photons can be absorbed in it (because YAG:Ce is not transparent) without being detected by the MAPD photodiode. In this case, some events in the low energy part of the spectrum (26 keV) may be lost.



**Fig. 6.** Energy spectra of the <sup>137</sup>Cs source measured by the MAPD + YAG:Ce based scintillation structure at 22 °C. (Color online)



**Fig. 7.** Number of the detected gamma rays by the MAPD with different thicknesses of the YAG:Ce based scintillation structure (55, 150, 255, 395, and 495  $\mu$ m).

The distribution of the amplitude of the gamma rays emitted by <sup>137</sup>Cs (662 keV) is shown in Fig. 6. The measurement time of 1000 s for each thickness was chosen. As the thickness of the YAG-Ce based scintillation structure increases, the high-energy part of the spectrum shifts up to the 20000ADC channel caused by the loss of ever more energy by the 662 keV gamma rays structure. This means that the 662 keV gamma rays produce more scintillation photons in the thick scintillation structures as compared to the thin ones.

The number of the detected gamma rays emitted from the  $^{137}Cs$  source does not significantly change with the thickness of the scintillation structure (up to 495  $\mu m$ ). The number of the detected gamma rays is in the range of 4258971 events (see Fig. 7). The mentioned thicknesses are not sufficient to absorb the 662 keV gamma rays, hence, we can see the shift of the high energy part of the spectrum. The obtained results demonstrate that the

thickness of the scintillation structure of 495  $\mu m$  ensures detection of gamma rays with the energies in the range of 26 to 662 keV. To increase the detection efficiency of the detector, it is suggested to increase the thickness of the scintillation structure up to several centimeters with the step of 1 mm.

Therefore, as can be seen from the presented results (see Fig. 7), the efficiency and sensitivity of the detector is possible to raise by increasing the thickness of the composite conversion scintillation structure. Furthermore, the efficiency of the detector based on such scintillation coatings can be increased if the detector has a hemispherical (convex, concave), pyramidal, conical form, the form of Fresnel lens *etc*. It may be assumed that the performance characteristics of such structure will also be affected by the heterogeneity of the distribution of phosphor particles in it and the presence of mineral scatterers, which requires further investigation.

### 4. Conclusions

Based on the research with <sup>241</sup>Am and <sup>137</sup>Cs sources, the possibility of registration of gamma rays by the ionizing radiation detector based on the YAG:Ce<sup>3+</sup> composite converting scintillation structure and MAPD is found.

The number of the detected gamma rays emitted by the <sup>241</sup>Am source is shown to increase linearly with the thickness of the composite converting scintillation structure. A certain geometric configuration of the structure is likely to increase the detector sensitivity, which requires further investigation.

The thickness of the composite converting scintillation structure of 495  $\mu m$  is found to enable registering gamma radiation quanta with the energies in the range of 26 to 662 keV.

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Дослідження чутливості до гамма-випромінювання сцинтиляторних структур на основі YAG:Се

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Анотація. Іонізуюче випромінювання сьогодні широко використовують для діагностики та дослідження широкого кола об'єктів завдяки високій достовірності та якості отриманих у результаті таких досліджень результатів. Використання високочутливих датчиків іонізуючого випромінювання дає змогу знизити дозу потрібного для досліджень іонізуючого випромінювання та створити чутливі системи для контролю параметрів середовища. У даній роботі досліджено ефективність датчика радіаційного випромінювання низької густини, в якому використано композитну сцинтиляційну структуру на основі порошкоподібного люмінофора YAG:Се в ролі конвертуючого покриття фоточутливого приймача. Для джерел на основі <sup>241</sup>Am і <sup>137</sup>Cs установлено можливість реєстрації гамма-випромінювання датчиком іонізуючого випромінювання на основі композитної конвертуючої сцинтиляційної структури YAG:Се<sup>3+</sup> і MAPD. Показано, що кількість виявлених гамма-променів, які випускаються джерелом <sup>241</sup>Am, лінійно збільшується зі збільшенням товщини композитної конвертуючої сцинтиляційної структури. Установлено, що вибрана товщина 495 мкм композитної конвертуючої сцинтиляційної структури дає змогу реєструвати кванти гамма-випромінювання, енергія яких перебуває в діапазоні 26 – 662 кеВ.

**Ключові слова:** іонізуюче випромінювання, сцинтиляційна структура, YAG:Се люмінофор, датчик радіаційного випромінювання низької густини.