

Summary of the Results of Research and Selection of Corundum Crystals for Development and Creation of UV and VUV Detectors

M.A. Hovhannisyan, J.G. Vardanyan*, A.R. Mnatsakanyan,
K.A. Mahtesyan, S.A. Mkhitarian

Institute of Applied Problems of Physics NAS RA, Armenia, Yerevan, 0014, St. Hr. Nersisyan 25

*E-mail: julia.vardanyan2011@yandex.ru

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Abstract: The paper summarizes the results of our research conducted on the study of corundum crystals with various activating agents for development and creation of UV and VUV detectors. Corundum crystals with certain parameters were chosen as the optimal converter of UV and VUV radiant energy into the visible region of radiation for creating UV and VUV detectors with high spatial and temporal characteristics, which also have high radiation and thermal resistance.

Keywords: UV and VUV detectors, convertor, wide-gap oxide, corundum crystals, thin sheets, corundum fibers

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1. Introduction

In recent decades, the characteristics of sources of synchrotron (electron, positron, etc.) radiation (power, continuous and wide spectrum, polarization, low divergence, etc.) have significantly improved, including the sources in the UV and VUV radiation regions. For example, in a relatively short period of time, the power of synchrotron radiation increased by 7–8 orders with high spatial and temporal stability, and the efficiency of using the radiation time reached to about ~ 95 % [1].

At the same time, progress in the development of UV and VUV detection systems, mainly associated with the use of SSDs, is not very effective, and the gap between achievements in these areas is still widening, although the characteristics of UV and VUV detectors are equally important [2].

In order to develop and create UV and VUV detectors with high spatial and temporal characteristics, which also have high radiation and thermal resistance, we carried out comparative studies of various corundum crystals with various activating agents using topographic (structural) and X-ray methods, to expand the frequency range and increase the spatial resolution of UV and VUV detectors on the basis of improving the structural and spectral characteristics of the converters created by our chosen corundum crystals.

After all these studies, as a converter of UV and VUV radiation into the visible region to create detectors with the above-described high technical characteristics, we selected and proposed a corundum crystal (aluminum oxide Al_2O_3) with certain activators (we used those chemical elements that provided good absorption in the UV and VUV spectrum).

Based on these considerations and taking into account the properties of corundum crystals (thermal and radiation resistance, necessary optical, spatial and temporal characteristics, etc.), we carried out experimental studies to find the optimal composition and concentration of mixtures at which the most complete conversion of UV and VUV radiation is achieved into the visible region [3].

Such crystals as corundum, being radiation-resistant materials, can be considered as a model object that has a set of technically important characteristics and is of particular interest both for

radiation physics of solids and applied purposes, and, for example, for the nuclear and electronic industries.

Our experiments showed that the use of corundum crystals in radiation dosimetry led to the increase in the accuracy of ionizing radiation monitoring and opened up new possibilities for using corundum crystals as effective dosimeters for synchrotron, X-ray, UV, and VUV radiation [4, 5].

The radiation resistance of these materials will make the conversion of electronic excitations into ionic ones unlikely, which leads to purely electronic relaxation processes. Our works also showed that this makes it possible to create radiation-resistant and radiation-sensitive sensors based on wide-gap oxides, which are corundum crystals [5]. The conducted experiments also showed that the spectral and luminescent characteristics of corundum crystals in the UV and VUV regions of the spectrum make it possible not only to record UV and VUV radiation, but also to obtain images of the emitted object [6, 8, 9].

Taking into the account the foregoing, on the basis of our conducted studies of corundum crystals with various impurities, we selected corundum crystals containing certain impurities with the required concentrations that are most suitable for use as converters of UV and VUV radiation into the visible region, for the development and manufacture of UV and VUV detectors. Despite the fact that, depending on the application, there are many criteria for assessing the optical qualities of crystals, in our spectroscopic studies to select the optimal corundum crystals for converters of UV and VUV detectors, we took into account the concentration of bubbles, as well as the concentration and composition of their impurities (the presence of impurities and their controlled introduction into the crystal, i.e., structure change), which are the determining parameters for the luminescent properties of crystals [6].

2. Experimental part

The implementation of the settled task included the sequence of the following works:

- Determination of the composition and concentration of impurities in the available and obtained by us crystals (measurements were carried out on an X-ray fluorescence spectrometer X-123SDD with a silicon drift SDD detector);
- Determination of the spectral properties of existing and obtained crystals (absorption and luminescence spectra) and their certification (measurements were carried out on NPF 448 and SP 8-1590UV/VIS spectrophotometers);
- Measurement of the number and size of bubbles in the samples (measurements were carried out with an optical eyepiece MIR 2);
- Thermal and radiation treatment of samples (thermal treatment was carried out on a vacuum furnace SMVE - 1.3.1/20, and radiation processing was carried out at the Research Center for Particle Physics DESY, Germany) [7].

On the basis of the selected crystals, two versions of UV and VUV radiation converters into the visible region of the spectrum were developed and created: a) an integrated converter - as a single crystal, and b) a converter with temporal and spatial resolution [5].

Based on the obtained results (relative characteristics of the spectral sensitivity, dependence of photoluminescence on the power and spectrum of the incident radiation, quantum yield, etc.), a corundum crystal with a chromium concentration of $Cr \sim 0.05\%$ (ruby) was chosen as the optimal crystal sample for use as a UV radiation converter for integrated detectors. Based on the selected ruby crystal as a UV radiation converter, an integrated UV detector with the $c \sim 0.9$ quantum yield was designed and created, in which the operating range was limited within 200–400 nm using appropriate filters.

One of the main factors in choosing the ruby crystal for use as a converter in UV integrated detectors was its absorption spectrum, which is characterized by the fact that two resonant absorption bands are observed in the region of 200–400 nm. At the same time, in the intermediate

300-350 nm range, the absorption of ruby is very small, while in the region of waves shorter than 250 nm, maximum absorption occurs, and the luminescence spectrum also has two resonance bands - in the range of 690-700 nm.

However, our studies have shown that the effective transformation of UV and VUV radiation into the visible region can also be carried out by corundum crystals, which include, in addition to chromium, other impurities, where chromium is not dominant [5]. In this case, transition metals - lanthanides, the atomic radius of which is larger than the atomic radius of aluminum, were used as additional impurities.

In the framework of these studies, their optical properties were studied and absorption and luminescence spectra were obtained, moreover, the study of the luminescence spectra was carried out at different frequencies of excitation irradiation of corundum crystals.

Theoretically, this is explained by the fact that, having large atomic radii, they can create new exciton-photon interactions, leading to new transitions and energy transformations. In particular, we have obtained and studied corundum crystal samples with impurities of chromium (*Cr*), manganese (*Mn*), iron (*Fe*), cobalt (*Co*), zinc (*Zn*), titanium (*Ti*), copper (*Cu*), and scandium (*Sc*). In the framework of these studies, their optical properties were studied, absorption, and luminescence spectra were obtained. Moreover, the study of the luminescence spectra was carried out at different frequencies of excitation irradiation of corundum crystals.

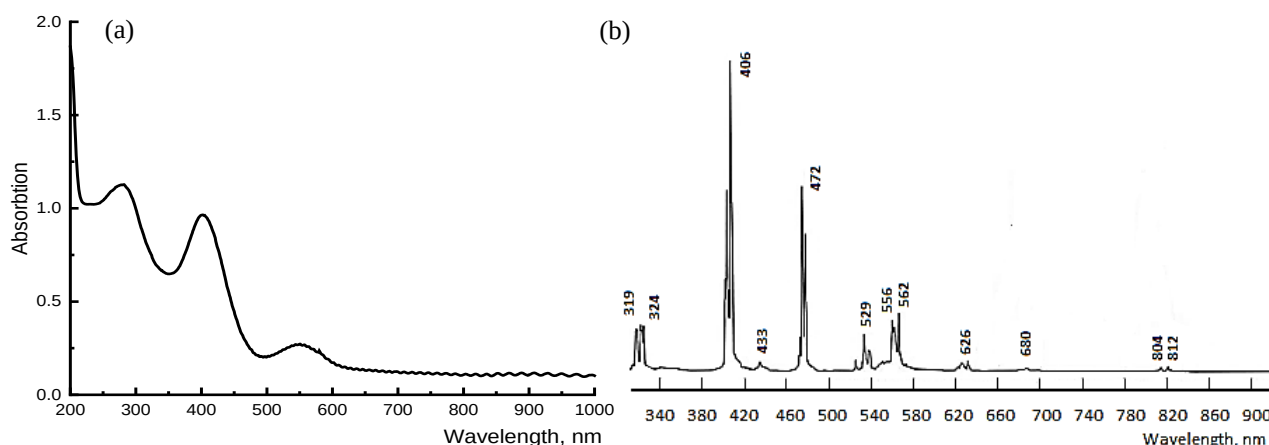


Fig. 1. (a) Absorption spectrum of a pink-yellow corundum crystal. (b) Luminescence spectrum of a pink-yellow corundum crystal exposed by the 257 nm radiation.

Fig. 1a, as an example, shows the obtained graph of the absorption spectrum of a yellow-pink corundum crystal. Table 1 shows the impurity concentrations of two corundum crystals (yellow-pink and dark-burgundy) measured on an X-ray fluorescence spectrometer.

Table 1. Composition and impurity concentrations of corundum crystals measured on an X-ray fluorescence spectrometer.

Samples of corundum crystals	Components							
	<i>Al</i>	<i>Cr</i>	<i>Mn</i>	<i>Zn</i>	<i>Ti</i>	<i>Fe</i>	<i>Cu</i>	<i>Ni</i>
	Relative weight concentrations, %							
Dark-burgundy	97.883	1.970	0.087	0.027	0.022	0.006	0.003	-
Yellow-pink	99.313	0.039	0.023	0.005	-	0.044	-	0.032

The absorption maxima of the yellow-pink crystal (Fig. 1a) are due to Ti^{4+} , Ti^{2+} и Ti^{3+} ions. These titanium impurities in various concentrations are always present in the samples of all crystals; however, the concentrations of impurity titanium ions cannot always be determined from absorption

curves. For this, other measurements were also needed, taking into account which we also carried out measurements of the luminescence of a yellow-pink crystal when the crystal was irradiated with different frequencies of monochromatic radiation. Fig. 1b, as an example, shows the luminescence spectrum of a yellow-pink corundum crystal upon irradiation with 257 nm radiation, which clearly expresses the nature of the color centers of the crystal.

In the framework of these studies, we also obtained the spectral characteristics of absorption and luminescence of a dark-burgundy crystal, where the study of the luminescence spectrum was carried out when it was irradiated with radiation with wavelengths of 468 nm, 370 nm, 357 nm, 300 nm and 250 nm with a band of 0.5 nm [5]. Fig. 2b, as an example, shows the luminescence spectrum of a dark-burgundy crystal when it was irradiated with the 250 nm radiation, from which it can be seen that the absorption maximum occurs in the wavelength range of 685–730 nm.

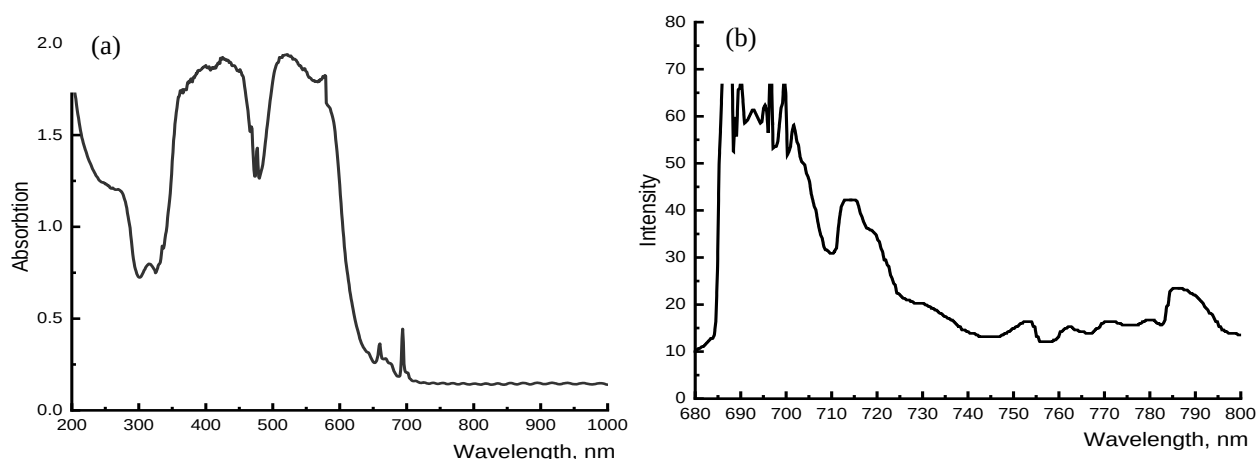


Fig. 2. (a) Absorption spectrum of a dark-burgundy corundum crystal. (b) Luminescence spectrum of a dark-burgundy corundum crystal exposed by the 250 nm radiation.

The conducted measurements also showed that the absorption of short-wavelength radiation by a dark-burgundy corundum crystal (Fig. 2a) with the indicated impurity composition increases in comparison with ruby, which makes a significant contribution to the total integrated luminescence flux.

For a more complete identification of the contributions of individual absorbed spectral components to the total integrated luminescence flux, we also recorded the luminescence spectra upon irradiation with continuous (white) light. Moreover, the luminescence spectra were obtained, first, upon irradiation of selected crystals with different frequencies of monochromatic radiation, and then, at irradiation with white light (Fig. 3).

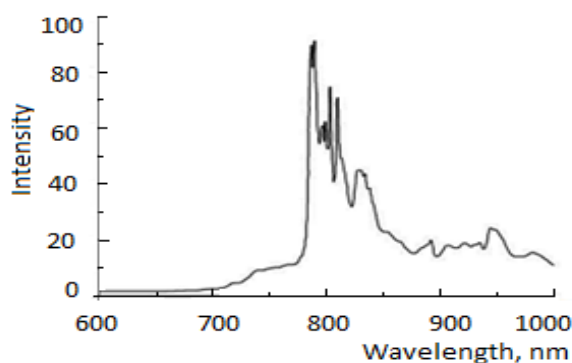


Fig. 3. Spectrum of luminescence of dark-burgundy corundum crystal when exposed by the "white" light.

Based on the analysis of the absorption and luminescence spectral characteristics of the above-mentioned studied crystals, we chose the dark-burgundy corundum crystal as the optimal one, the use of which as a converter in UV and VUV detectors led to an increase in the output current of the detector when exposed by white light and, consequently, to a 30 % increase of the luminescence power compared to ruby-based UV detector.

The correlation between the operating characteristics, which determine the spectral sensitivity of UV and VUV detectors, and the structural characteristics of corundum crystals, makes it possible to expand the research area of UV and VUV radiations, that is, to expand the research area of spectral and luminescent characteristics of corundum crystals, which will allow not only to register UV and VUV radiations, but also to receive the image of the emitted object. The study of the temporal characteristics of the luminescence of various samples of corundum crystals also allows us to understand the relaxation processes at the excited electronic levels of impurity atoms in order to receive pulse signals in the UV and VUV spectrum regions.

After systematization and certification of samples of corundum crystals with various impurities, we will create a "bank" of our studied samples with open access for researchers, where the results of our respective studies will also be available.

For the implementation of the second variant of a converter with temporal and spatial resolution, thin plates with a thickness of 0.15-0.2 mm were obtained from almost all the investigated samples of corundum crystals, and the spectral characteristics of these plates with different impurities were studied. Impurities were implanted in thin crystals from both sides by a flow of high-energy chemical elements, during which those elements were selected that were well absorbed in the UV and VUV spectrum ranges. Then they were polished (both chemically and mechanically), covered with a connecting material (nickel, copper, silver, platinum, etc.) and "connected" together in the form of matrices with dimensions of 6x7 (the dimensions of the matrix elements were - 0.2x0.2x2.5 mm) [5]. Further, a matrix converter was made from these fiber crystals connected together to be used for the creation of UV and UV detectors. At the same time, the conducted studies showed that the spatial distribution of the luminescence intensity is not only preserved, but also transferred into the visible part of the radiation, after which the visible image at the output of the matrix can be directly registered [5].

To further improvement of the spatial resolution characteristics of such detectors, it is necessary to obtain fiber-optic plates with a smaller thickness, as the spatial resolution of the detector is determined by the size of the elements assembled in the matrix.

3. Conclusions

Experimental studies of the obtained samples of corundum crystals with various impurities were carried out, in the framework of which, in particular, their optical properties were studied and absorption and luminescence spectra were obtained. Measurements of the luminescence spectra of the investigated crystals were carried out at several wavelengths within the capabilities of the available equipment, as well as when irradiated with white light.

Conducted experiments showed that the dark-burgundy corundum crystal studied by us has a sufficiently good absorption (more than 60 %). The inclusion of a dark-burgundy corundum crystal in the UV and VUV detector as a converter led to an increase in the output current of the detector and, therefore, the luminescence power by 30 %, compared to a ruby-based UV detector.

For the development and creation of UV and VUV image detectors with high spatial and temporal characteristics, thin plates of corundum crystals with a thickness of 0.15-0.2 mm were mechanically obtained and the spectral characteristics of the most of these plates with the addition of various impurities were studied. For use as a converter of a UV and VUV detector with spatial sensitivity, those plates were chosen that absorbed well in the UV and VUV spectrum. From these

plates, matrices with sections of matrix elements - 0.2x0.2x2.5 mm were collected and images were obtained with web cameras.

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References

- [1] H. Graafsma, J. Synchrotron Rad. **42** (2017) 418.
- [2] Reliability Workshop, ESRF, September (2022).
- [3] S.A. Mkhitarian, J.G. Vardanyan, H.S. Agajanyan, M.N. Nersisyan, A.R. Mnatsakanyan, H.R. Harutyunyan, M.A. Hovhannisyan, J. Contemp. Phys. **57** (2022) 289.
- [4] M.W. Allen, N. Swift, K.M. Nield, Atmosphere **11** (2020) 744.
- [5] M.W. Allen, N. Swift, K. M. Nield, B. Liley, R.L. McKenzie, IEEE Sens. J. **21** (2021) 18706.
- [6] J. Vardanyan, M. Hovhannisyan, S. Mkhitarian, M. Nersisyan, H. Agajanyan, Arm. J. Phys. **13** (2020) 382.
- [7] The Research Center for Particle Physics DES, <http://regae.desy.de/>.
- [8] Y. Zou, Y. Zhang, Y. Hu, H. Gu, Phys. Sens. **18** (2018) 2072.
- [9] S. Inamdar, V. Ganbavle, S. Shaikh, K. Rajpure, J. Phys. Stat. Solidi (a) **212** (2015) 1704.