ABSTRACT

dissertation work of Alpyssova Gulnur Kenzhebekovna on the thesis «Radiation synthesis conditions on luminescence YAG:Ce ceramics», submitted for the degree of Doctor of Philosophy (PhD) in the specialty «6D072300 - Technical Physics»

The aim of the dissertation research

The aim of this work is to identify the causes of the dependence of the reproducibility of the radiation synthesis of ceramics based on YAG:Ce depends on the conditions and modes of irradiation and preparation of the initial mixture.

Research objectives. In the course of the dissertation work, the following particular scientific tasks were set and solved:

1. Synthesis of ceramics of yttrium-aluminum garnet activated by cerium.
2. Investigate the morphology and structure of ceramics.
3. Measure the spectral and kinetic characteristics of samples of different series obtained by radiation synthesis, placement in the crucible, preliminary preparation of mixtures and their composition.
4. Conduct research on the dependence of the efficiency of radiation conversion by the phosphor on the conditions for the preliminary preparation of the charge.
5. Estimate the efficiency of luminescence conversion for different types of excitations.

Research methods

The spectral, kinetic and energy characteristics of the luminescence of YAG:Ce and YAGG:Ce-based ceramic samples synthesized in the field of high-power radiation fluxes were measured depending on the synthesis conditions. We studied ceramic samples obtained at different times, with different compositions, different prehistory of the initial oxide powders and charge bulk density, different degrees of compaction, and different arrangement of samples in the crucible. To obtain reliable information, all measurements were carried out for each case, usually 10 times, and the measurement error was determined. In total, more than 1500 measurements were carried out for each measured parameter. The obtained measurement results are systematized and analyzed.

The main provisions for defense:

1. The structure of ceramics produced by the YAG:Ce radiation method depends on the technological regimes of synthesis.
2. There is a spread in the values of the spectral characteristics of the luminescence of ceramics obtained in one crucible. The scatter of the values of the luminescence spectral characteristics weakly depends on the temperature difference of the crucible during synthesis, the position of the series samples in the crucible, the degree of compaction, and the beam scanning rate during synthesis.
3. Synthesized ceramic samples show 50-60% efficiency of conversion of excitation radiation into luminescence compared to industrial ones.
4. The relative luminous efficiency of the obtained YAG:Ce ceramic reaches 40% of the efficiency of the reference YAG:Ce scintillator. The photoluminescence quantum yield of synthesized ceramics and industrial (commercial) samples are comparable.

Description of the main results

Synthesized luminescent ceramics based on YAG:Ce by exposure to powerful flows of high-energy electrons directly on the mixture of metal oxides without their pre-

treatment and the use of additional and auxiliary materials. Synthesized YAG:Ce ceramics has characteristic properties for phosphors, ceramics based on YAG:Ce, YAGG:Ce obtained by other methods. Ceramics is a sticky particles with a size of 1 - 30 microns with pronounced planes, characteristic of crystals. This indicates the existence of a tendency for the formation of a crystal structure during synthesis. The diffraction patterns fully correspond to those known for YAG:Ce crystals in terms of the position and ratio of the peaks. The performed studies made it possible to detect differences in the spectral and kinetic characteristics of the luminescence of YAG:Ce ceramic samples obtained by radiation synthesis: the position and half-width of the luminescence bands, the luminescence decay kinetics, depending on the synthesis conditions. The scatter of characteristic values is observed both in samples of different series and in samples of the same series. These differences reach ±5 nm, 0.02 eV, and ±4 ns, respectively, and exceed the measurement confidence interval, which we determined as follows: ±1 nm, 0.01 eV, and ±2 ns. Thus, the scatter in the values of the luminescence spectral characteristics, i.e., the reproducibility of the synthesis results, does not depend on the temperature difference of the crucible during synthesis, the position of the series samples in the crucible, the degree of compaction, and the beam scanning rate during synthesis. The synthesized ceramic samples show 50-60% photoluminescence conversion efficiency compared to commercial YAG:Ce phosphors. In a series of samples modified with gadolinium, the conversion efficiency is higher than in those without gadolinium.

The kinetic characteristics of the luminescence of the synthesized ceramics are similar to materials based on YAG:Ce. Under photoexcitation, the kinetics has a characteristic decay time of ~60 ns, which is the dominant decay component. When cathodoluminescence is excited, the same trend is observed in terms of spectral and kinetic characteristics, but there are three damping components. The relative scintillation efficiency reaches 40% of the efficiency of the reference YAG:Ce scintillator. The quantum yield of photoluminescence of ceramics is comparable to industrial (commercial) samples.

Description of the novelty and importance of the results obtained. The scientific novelty and theoretical significance of the research results are as follows:

Comprehensive studies of the spectral-kinetic and quantitative characteristics of luminescence for the first time obtained by the method of radiation synthesis of YAG:Ce ceramic samples with statistical processing of their values were carried out. The ranges of deviations of values, luminescence characteristics from the statistical average, dependence of deviations on modes and conditions of synthesis are established. It is shown that the spread of deviations and luminescence characteristics of samples of different prehistory exceeds the confidence interval of measurements and is due to the difference in physicochemical processes during synthesis. It is shown that the luminescent properties of the outer and inner layers of the ceramics synthesized in the radiation field differ. It has been established that the main reason for the spread in the characteristics of the luminescent properties of YAG:Ce ceramics obtained by radiation synthesis is the high rate of synthesis and, especially, the high rate of cooling of the samples. It is concluded that, after radiation synthesis of ceramics based on YAG:Ce, thermal or radiation annealing is necessary to complete the formation of the ceramic YAG phase. A method has been developed for express evaluation of the efficiency of

converting the energy of exciting radiation into luminescence by comparing the brightness of the luminescence of the test sample with the reference one.

Compliance with the directions of scientific development or state programs

The dissertation work corresponds to the priority areas of science development that are being implemented in the Republic of Kazakhstan and contains new scientifically substantiated results, the totality of which is important for the development of the studied scientific areas. The work was carried out within the framework of grant funding for scientific projects of the Ministry of Education and Science of the Republic of Kazakhstan AP08052050 "Development and improvement of the synthesis of luminescent YAG:Ce nanoceramics in the field of powerful radiation fluxes" 2020-2022.

Description of the applicant's contribution to the preparation of each publication

The research results presented in the dissertation were obtained personally by the author, as well as jointly with the staff of the Department of Laser and Light Technology (Tomsk, Russia), which is reflected in the publications.

Discussion and analysis of the results were carried out jointly with the scientific consultant Dr. of Sciences, Professor V.M. Lisitsyn

The main results of the dissertation research were published in 15 publications that fully correspond to the topic of the dissertation. Of these, 7 articles were published in peer-reviewed scientific journals included in the Web of Science and Scopus database, and 5 abstracts were published in the proceedings of international scientific conferences.

The main provisions of the work and the results obtained were reported and discussed at international scientific conferences: 20th International Conference on "Radiation Effects in Insulators" (Nur-Sultan, Kazakhstan 2019); 7th International Congress on "Energy Fluxes and Radiation Effects" (Tomsk, Russia, 2020); II International Youth Congress "Modern materials and technologies of new generations" (Tomsk, 2019); Anniversary International Youth Conference on Luminescence and Laser Physics (Irkutsk, 2019); - 13th international conference "Functional Materials and Nanotechnologies" (Vilnius, 2020); XV International Scientific Conference of Students and Young Scientists "ǴYLYM JÁNE BILIM - 2020" (Nur-Sultan, Kazakhstan), 2021 Spring Meeting of the European Materials Research Society (E-MRS) to be held as a VIRTUAL Conference from May 31st to June 3rd , 2021, 11th International Scientific Conference “Chaos and Structures in Nonlinear Systems. Theory and experiment” (Karaganda, 2019).