YAG based phosphors, synthesized in a field of radiation

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Abstract. YAG:Ce, YAGG:Ce ceramics were obtained by sintering the oxide powders in the radiation field. The ceramics well luminesces when excited by radiation fluxes of chips on 460 nm, which are commonly used in white LEDs to excite luminescence. The results of investigations of the luminescence of powders of crushed ceramics are presented. The luminescence characteristics of powders exactly correspond to the phosphors luminescence used in practice obtained by solid-state synthesis methods.

1. Introduction

YAG:Ce based phosphors are the most promising for use in LEDs. The phosphors are crystalline multicomponent systems [1]. Their synthesis is usually carried out using solid-state reactions [2, 6, 7]. The melting points of the components are from 2455° C in Y_2O_3 to 2075° C in Al_2O_3 [3, 4]. Therefore, the reproducibility of the results of synthesis is low: the elemental composition of microcrystals differs significantly from that contained in the burden, the composition of different batches of phosphor varies, even when synthesized under identical conditions, and the same initial composition of the burden. Luminescent properties also change. This is explained by the strong influence on the result of synthesis of uncontrolled deviations of technological modes in the synthesis process [5, 7–9]. Consequently, work is constantly being done to improve the synthesis technologies.

In this paper, we present the results of investigations of phosphors synthesized in a radiation field. Synthesis in the radiation field should obviously promote the flow of solid-state reactions.

2. Experimental details

Phosphors of different compositions were synthesized with the content on burden Y_2O_3 from 22 to 36 weight %, Al_2O_3 from 56 to 62 weight %, Ce_2O_3 from 4.8 to 9.1 weight %, and Gd_2O_3 from 0 to 12 weight %. The synthesis was carried out by sintering samples from powders of oxides in the field of the flow of high-energy electrons. The synthesized samples were represented as ceramic balls with a diameter of 3-6 mm of yellow colour. The samples were crushed into powder, the photoluminescence

spectra were measured by excited chip emission on 365 and 460 nm. The main excitation bands of luminescence YAG:Ce based phosphors are in 340 and 460 nm. Narrow emission bands of the selected chips are in good matched with the position of wide excitation bands of YAG phosphors. The luminescence spectra were measured with an AvaSpec-2048 spectrophotometer. The characteristics of the obtained samples were also measured: elemental composition, X-ray analysis. Elemental analysis of the synthesized samples was determined using SEM, which includes the EDAX system (energy-dispersive analysis). To determine the phase composition, a Rigaku Miniflex 600 diffractometer was used.

3. Experimental results

Figure 1 shows photographs of the broken ceramic samples when illuminated by the radiation from the chips. The samples intensively luminesce when it illuminated. In the place of split, the luminescence is noticeably brighter than on the sample surface. This is explained by the fact that on the surface there are particles during the synthesis of dispersed substances of burden and melting pot are precipitated.

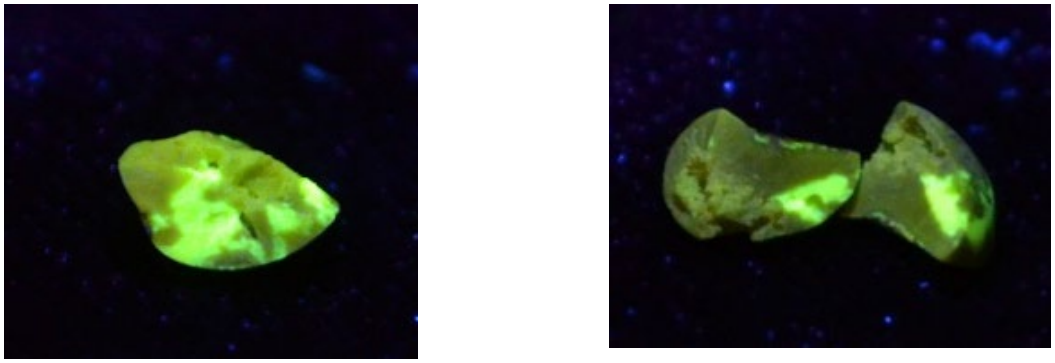

Figure 1. Broken ceramic samples.

Figure 2 shows photographs of powdered samples of the resulting ceramics. As can be seen from the results of the photographs, luminescence is observed when UV radiation is irradiated by chips. There is a difference in the colour of the luminescence upon excitation by 365 and 460 nm radiation. When the radiation is excited at 365 nm, the colour of the luminous powder is yellowish, whereas when excited at 460 nm, it is bright white. The difference is explained by the fact that when excited at 460 nm, the spectrum of the visible spectrum is complemented by the radiation of the chip, which is related within the visible range. In the powder of the phosphor, non-luminous particles are visible. Obviously, these particles are on the surface synthesized metal oxides samples found from which the melting pot is made.

Figure 2. Photographs of powdered ceramics.

The luminescent characteristics of synthesized phosphors are basically similar known as solid-state synthesis obtained by traditional methods. Figure 3 shows the results of an investigation of the luminescence spectra of samples of different compositions crushed into powder. In the phosphors of compositions Al_2O_3(56.8%)+Y_2O_3(34.1%)+Ce_2O_3(9.1%) (5.08.18) the band maximum is observed ~530 nm. The luminescence band has half the width 0.442 eV. In phosphors with gadolinium, the luminescence band position is shifted to the red region to 556 nm. The band has half the width 0.505 eV.

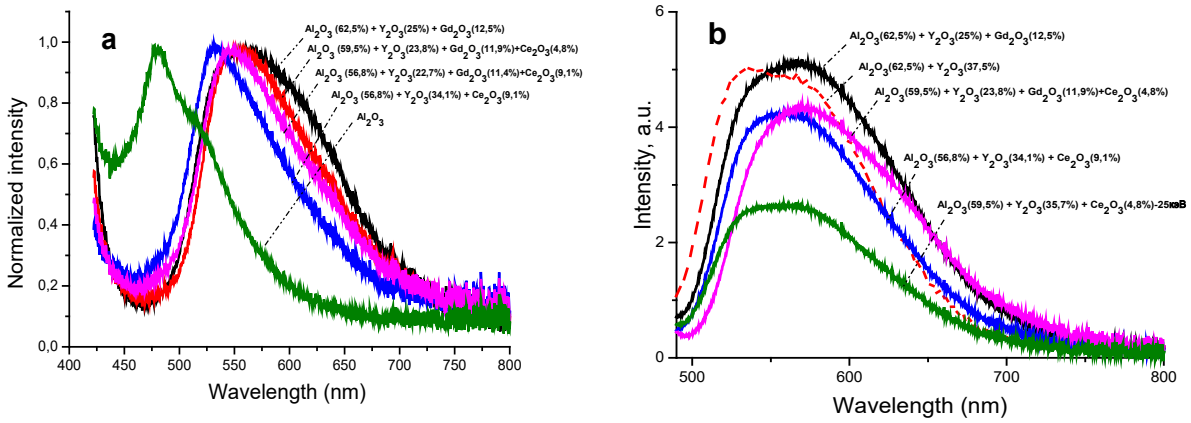

Figure 3. Luminescence spectra excited by radiation from the chip with λ_{ex}=365 nm (a) and λ_{ex}=460 nm (b).

The synthesized luminescence spectra of powders of phosphors are similar to industrial phosphors synthesized by solid-state methods [6, 7]. Figure 4a,b shows the luminescence spectra of the phosphors SDL 4000, 3500 and SDL 2700, differing in that part of the yttrium ions in SDL 2700 is replaced by gadolinium.

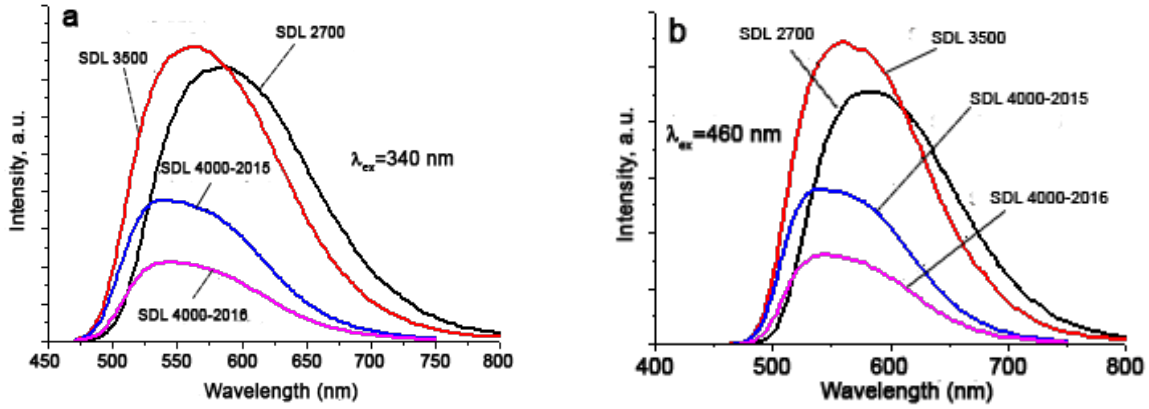

Figure 4. Luminescence spectra of industrial phosphors SDL 2700 ($Y_{1.24}Gd_{1.56}Al_5O_{12}$:$Ce_{0.2}$), SDL 3500 ($Y_{2.6}Gd_{0.25}Al_5O_{12}$:$Ce_{0.15}$), SDL 4000 ($Y_3Al_5O_{12}$) excited on λ_{ex}=340 nm (a) and λ_{ex}=460 nm (b).

The elemental composition of samples of ceramic phosphors was studied. Examples of the results of the study are shown in a figure 5. The figure shows the spectra of electron emission of elements of a composition typical for the investigated phosphors. The tables in figure 5 show the numerical values of the analysis results and the composition of the burden. As can be seen from the comparison of tabulated values of the ratio of elements in the initial composition for the synthesis and in the

synthesized samples are fairly close. For example, the ratio of the weight amounts of yttrium and aluminum in the initial sample is within from 34.1 to 37.5 wt % and from 56.8 to 62.5 wt %. The ratio of the relative amounts of cerium in the burden is 0.8: 1.9 for the initial proportions in the burden 4.8:9.1. The satisfactory correlation of the ratios in the initial and final products indicates that during synthesis there was a fairly good mixing of the elements.

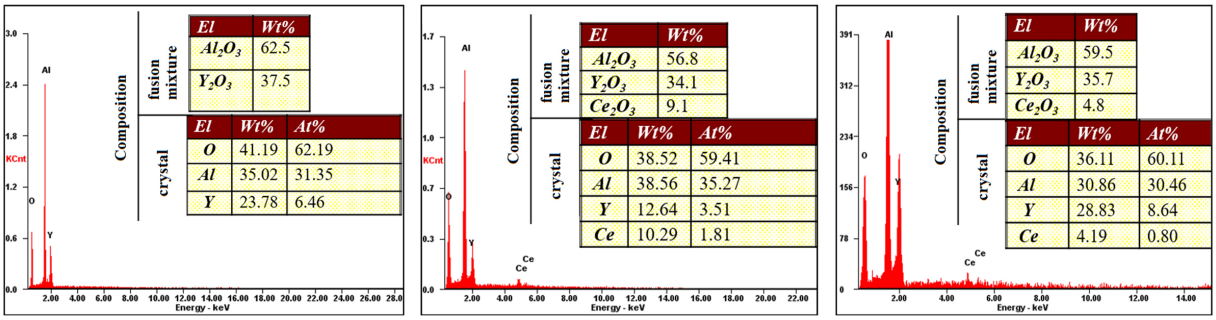

Figure 5. Element composition of synthesized samples of ceramics.

The examples of the results of X-ray diffraction analysis of the produced samples are given in a figure 6. From the presented results follows, the ceramic is a sintered crystallite with a phase characteristic of the phosphors. In phosphors Al2O3 (56.8%)+Y2O3 (34.1%)+Ce2O3 (9.1%) the dominant phase is yttrium aluminum garnet. In the phosphors Al2O3 (56.8%)+Y2O3 (22.7%)+Gd2O3 (11.4%)+Ce2O3 (9.1%) – yttrium-aluminum-gadolinium garnet phase.

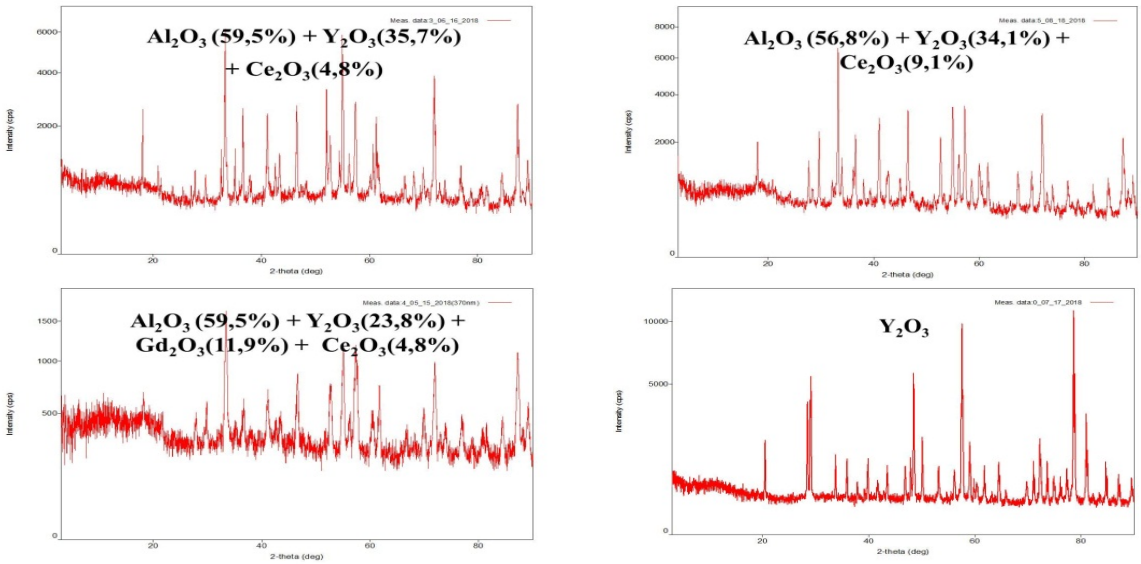

Figure 6. X-ray of synthesized ceramics powders.

Most of the spectrogram lines correspond to the standard for the YAG and YAGG phases. Some differences in the number of detected lines are due to the existence of a large background in the spectrum. This is the expected difference between the ceramic and the crystal.

As the results of the x-ray diffraction analysis, crystallites sizes in ceramics is within from 10 to 50 nm.

4. The discussion of the results

Yttrium-aluminum garnet (YAG: Ce) based ceramics is obtained by sintering in a field of high-energy electrons flow. The ceramics synthesis was carried out in a burden Y2O3, Al2O3, with Ce2O3 as an activator. Samples of ceramics with Gd2O3 as a modifier were also synthesized. The composition of the ceramics is close to that contained in the burden. The crystal structure corresponds to the standard for YAG and YAGG. Crystallites have sizes from 5 to 50 nm.

The obtained ceramic has good luminescent characteristics. The luminescence spectra of powders of ceramic samples after grinding are typical for phosphors with similar compositions. The luminescence band position shifts with the doping of gadolinium to 20 nm to the red region of spectrum. The brightness of the glow of powders when excited by the radiation of the chip is comparable with the brightness of industrial phosphors.

The ceramics synthesis in a field of radiation differs from synthesis in heat fields. The formation of a new phase from the others depends not only on the temperature, but also on the degree of ionization of the components. For example, the YAG ceramics synthesis is realized using a 15% greater flux power than the synthesis of MgF2-based ceramics. The melting points of the components for the YAG ceramics range from 2050 to 2450°C, whereas MgF2 has a melting point of 1260°C.

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