

# Luminescence decay kinetics of yttrium aluminium garnet phosphor at different temperature

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**Abstract.** The temperature dependence of the luminescence decay for a series of SDL YAG:Ce phosphors is investigated. Luminescence quenching excited by chip under heat-treatment is observed. The dependence of temperature on thermal luminescent intensity at elevated temperature (range from room temperature to 200°C) was tested. Noticeable luminescence quenching of SDL 2700 phosphor is observed at temperatures above 50°C, and by increasing temperature up to 200°C, the luminescence intensity decreases 5 times. Compare with sdl2700 phosphor, the temperature quenching of sdl3500 and sdl4000 phosphors is much lower. The investigated phosphors differ in the content of Gd<sup>3+</sup>. An explanation of the observed effects is proposed.

## 1. Introduction

For lighting, white light sources (white LEDs) are the most widely used [1]. In these LEDs, the emission of the chip in the blue region of the spectrum is partially converted into radiation in the range 500–800 nm. The combined radiation of the chip and phosphor overlap the entire range of visible light. Phosphors based on YAG: Ce are most often used to convert the radiation. YAG phosphors provide a high level of transformation of the radiation of the chip [2–3]. By changing the composition of activators, coactivators, it is possible to change the spectral luminescence characteristics of YAG: Ce phosphors and chromatic properties of LEDs [4-5].

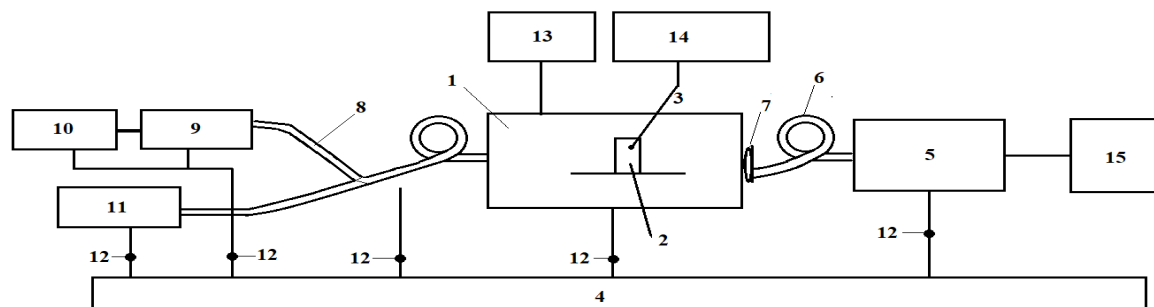
One of the main problems in the synthesis of phosphors is improving their thermal stability and the temperature quenching of luminescence. LEDs can work in conditions of high ambient temperatures. The phosphor is in close contact with the chip, which heats up to temperatures above 60°C, the phosphor itself is heated by the conversion of excitation energy [9-11]. In [6], a decrease in the luminescence efficiency with increasing temperature was observed. In [7] shows the influence of Ce<sup>3+</sup> on the thermal stability of YAG: Ce phosphor and the effect of temperature quenching of YAG: Ce phosphor. In [8], it shows that the temperature quenching is influenced by the presence of coactivators, for example Gd<sup>3+</sup>, which is introduced to improve the color rendition.

In this paper, it is devoted to study the temperature quenching for a series of SDL phosphors with different content of Gd<sup>3+</sup>.



## 2. Experimental

Industrial YAG phosphors of SDL4000, SDL2700 and SDL3500 with different ratio of elemental composition were used for the present research. Detailed information on the synthesis is described in [12]. The luminescence decay kinetics was measured at a constant present temperature with time. The diagram for measuring the kinetics of luminescence decay is shown in figure 1.



**Figure 1.** The stand circuit. 1 – Heating furnace, 2 – Cuvette for the phosphor, 3 – Thermal couple, 4 – Optical bench, 5 – Spectrophotometer, 6 – Optical waveguide, 7 – Lens, 8 – Optical waveguide with splitter, 9 – Telescopic system, 10 – Chip, 11 – Laser, 12 – Rack mount (riders), 13 – Power system, 14 – Multimeter, 15 – Laptop.

The relative temperature-dependent luminance of a certain phosphor at elevated temperature was tested by a controlled resistance-heating and a thermocouple, using an excitation source of blue LED with  $\lambda_{em}=455$  nm, which was directed through the optical system and a quartz fiber to a cuvette with phosphor. The luminescence was measured at regular intervals through a collecting lens, an optical fiber and an Avantes 3648 spectrophotometer. The information was processed by a computer. All the elements of the stand were rigidly fixed to the optical bench. The result of the measurement was the luminescence decay kinetics with time after heating at elevated temperature. The measurement time for the kinetics was usually 200 min. The measurements were carried out in the temperature range from 30 to 200°C. The relative intensity of the luminescence was measured. The accuracy of measuring the relative intensity in the experiment was achieved as follows. The relative placement of the stand elements remained unchanged, the power of the circuit elements was supplied from stabilized sources. To assure the accuracy of the tested temperature, the phosphors were embedded as close as possible to the thermocouple probe tip. The heating temperature is set to range from room temperature to 300°C.

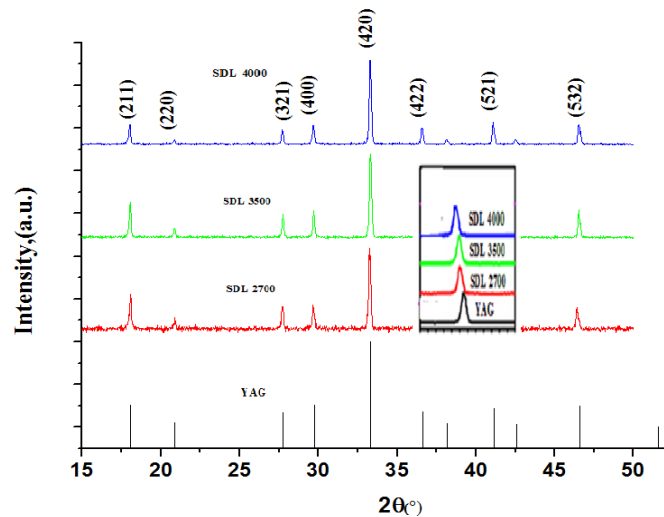
## 3. Results and discussion

### 3.1. Element composition and crystal structure of phosphors

The results of the elemental analysis of the investigated phosphors are shown in table 1.  $Gd^{3+}$  was not introduced into in SDL 4000 phosphor; the content of  $Gd^{3+}$  introduced into SDL 2700 was much larger than it in the SDL 3500. It is noted that the elemental composition of the investigated phosphors of one type with different batches and different years of synthesis differs for basic elements by almost 15%, for activator – by almost 35%. Such relationships are also characteristic of other industrial phosphors that we studied earlier. However, the number ratio between ions of  $Gd^{3+}$  and  $Y^{3+}$  in the investigated phosphors varies greatly.

The results of X-ray diffraction analysis of the investigated phosphors based on  $Y_3Al_5O_{12}:Ce^{3+}$ , doped with  $Gd^{3+}$  with different concentrations are shown in figure 2. All the investigated phosphors had a typical YAG structure, which is confirmed by the standard JCPDS09-1316  $Y_3Al_5O_{12}$  card. Some slight relative shift of the spectral lines is observed with a change in composition by introduction of  $Gd^{3+}$ . This

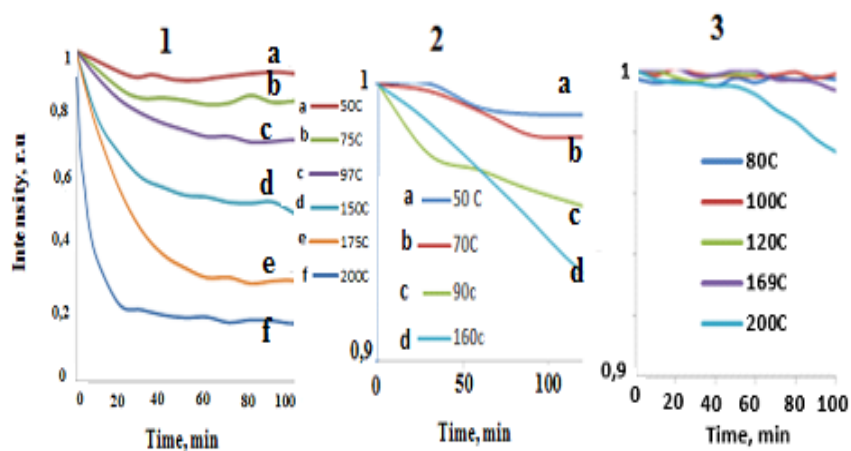
means that the lattice parameter increases with increasing gadolinium content in the crystal. The increase is due to the replacement of yttrium with gadolinium with large ion sizes.



**Figure 2.** XRD patterns of SDL4000, SDL3500, SDL2700 and standard  $Y_3Al_5O_{12}$ .

### 3.2. Luminescence decay kinetics

Figure 3 shows the kinetic curves for the luminescence decay of  $Y_3Al_5O_{12}:Ce^{3+}$  phosphors doped with  $Gd^{3+}$  ions with different concentrations at different temperatures. The change in the luminescence intensity kinetics was performed for more than 120 min in the temperature range from 50 to 200°C. The results of research of the luminescence decay kinetics (that are shown in figure 3-1) show that there is an intensity decrease with time after the start of heating of the SDL 2700 phosphor for 30–40 min. Then the luminescence intensity remains constant throughout the measurement time. The intensity decrease is insignificant at temperatures below 50°C. At temperatures above 175°C, the intensity decrease value reaches a maximum of 20% of the initial intensity, and remains almost unchanged. The luminescence decay in the SDL 3500 occurs over a longer period of time, the decrease value to 100 min at different heating temperatures is from 2% to 6% (figure 3-2). Even less is the intensity decrease value in the SDL 4000 phosphors of  $Gd^{3+}$  not activated (figure 3-3).

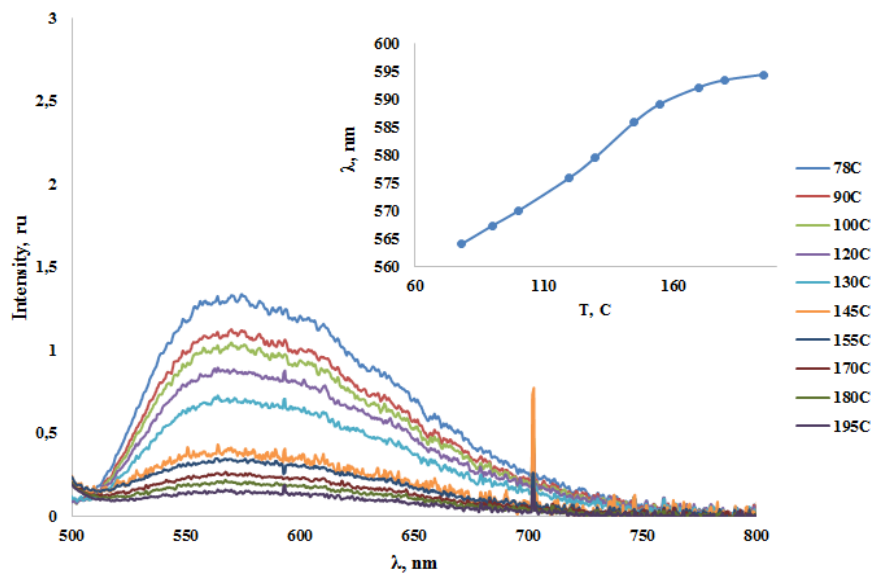


**Figure 3.** The luminescence decay kinetics of the phosphors:  
1 – SDL 2700; 2 – SDL 3500; 3 – SDL 4000.

Thus, the luminescence decay kinetics and the limiting value of the decay are quite clearly determined by the presence of  $Gd^{3+}$  in crystals.

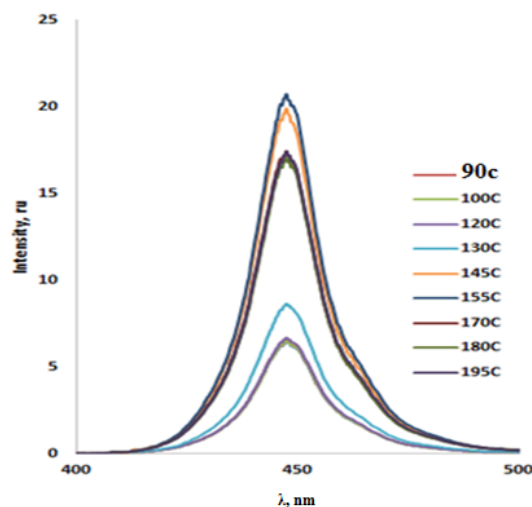
### 3.3. Dependence of the luminescence spectra of a phosphor and a chip on the temperature

Figure 4 shows the temperature dependence investigation results of the luminescence spectra of the YAG SDL 2700 phosphor. Luminescence spectra were measured at different temperatures after the end of the recession to a stable level. As follows from the presented results, there is a decrease in the luminescence intensity, broadening of the luminescence band. The inset to figure 4 shows the dependence of the maximum position of the luminescence band on temperature.



**Figure 4.** Temperature dependences of the luminescence intensities of the YAG SDL 2700 phosphor. The inset shows the temperature dependence of the positions of the maxima of the luminescence bands of the SDL 2700 YAG phosphor.

Figure 5 shows the results of the temperature dependence investigation of the electroluminescence spectra of the chip. The luminescence spectra were measured at different temperatures. As follows from the presented results, there is a decrease in the luminescence intensity. Band broadening is not observed.



**Figure 5.** Temperature dependence of the emission band of the chip.

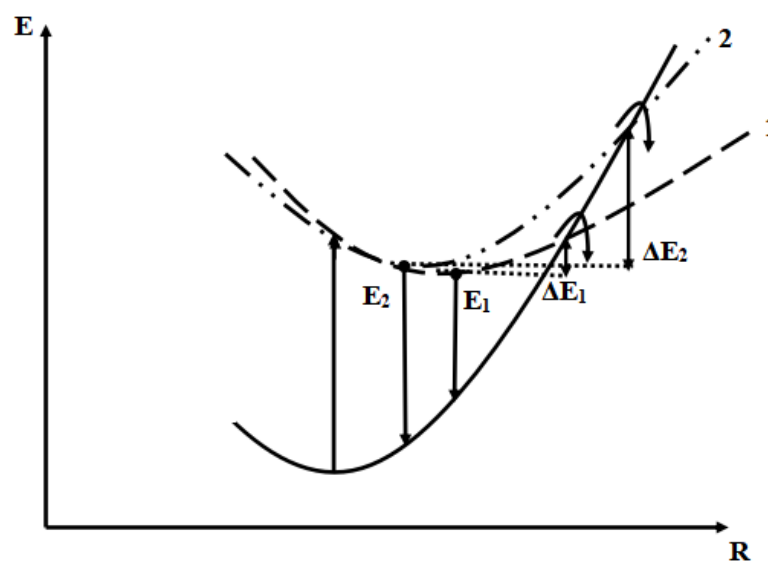
Table 1 shows the temperature dependence generalized characteristics of the emission band of a phosphor and a chip.

**Table 1.** The emission band characteristics of a phosphor and a chip.

Phosphor radiation										
$\lambda$ (nm)	563.95	567.27	569.92	575.89	579.53	585.83	589.14	592.11	593.44	594.43
T (C)	78	90	100	120	130	145	155	170	180	195
Half-width (eV)	0.432	0.433	0.438	0.442	0.448	0.466	0.489	0.524	0.566	0.575
Chip radiation										
$\lambda$ (nm)	447.42	447.42	447.42	447.42	447.424	447.42	447.42	447.42	447.42	447.42
T (C)	78	90	100	120	130	145	155	170	180	195
Half-width (eV)	0.093	0.0989	0.105	0.105	0.106	0.112	0.112	0.106	0.106	0.105

#### 4. Discussion

The conducted studies results have shown that there is a significant difference in the kinetic characteristics and the decay value in the SDL 2700 and SDL 3500, 4000 phosphors. The main difference between these phosphors is the difference in composition. In the SDL 2700 phosphor most of the  $Y^{3+}$  ions are replaced by  $Gd^{3+}$  ions. Hence, the replacement of  $Y^{3+}$  ions by  $Gd^{3+}$  ions leads to a decrease in the thermal stability of phosphors. It follows from structural investigations that the replacement of  $Y^{3+}$  ions by  $Gd^{3+}$  ions affects the crystal lattice parameter. Heating the SDL 2700 phosphor leads to a shift in the luminescence band from 564 to 594 nm, while the excitation band of its shape does not change. Perhaps the following explanation of the observed effects. The replacement of  $Y^{3+}$  ions by  $Gd^{3+}$  ions leads to a distortion of the configuration curves of the luminescent centers levels. The shape of the configuration curves changes more in the excited state. When  $Y^{3+}$  ions are replaced by  $Gd^{3+}$  ions, the curves and the curve minimum position are shifted because of the change in the mutual distance of the ions in the glow center region. An assumed image of the configuration curves is given in figure 6.



**Figure 6.** The configuration-coordinate diagram for the SDL 2700(1) and SDL 3500, 4000(2) phosphors.

Excitation by the radiation of the chip leads to the electron transition to the excited state, from which the electron passes into the radiative state in any of the considered phosphors, since they intersect. Radiation can occur in the main of the SDL 2700 (1) and SDL 3500, 4000 (2) different radiative states with the quantum energies  $E_1$  and  $E_2$ , since they have appreciably different interionic distances. Therefore, the luminescence bands of the 2700 (564 nm) and 3500 (555 nm) and 4000 (540 nm) phosphors are shifted. Since the configuration curves of the excited states of the SDL 2700 and SDL 3500, 4000 the luminescent centers intersect with the ground at different points, the activation energy of the SDL 2700 phosphor quenching is lower,  $\Delta E_1$  and  $\Delta E_2$ , respectively. Consequently, the temperature quenching of the luminescence in this phosphor will be more. The maximum position displacement of the luminescence band with increasing temperature is evidently due to the configuration curves asymmetry in the excited and ground states.

The strong dependence of the luminescence efficiency of the SDL 2700 phosphor on temperature should not be considered as a significant disadvantage of these phosphors. SDL 2700 phosphors provide the generation of light with a low color temperature and high intensity. It is necessary to use LEDs with these phosphors under conditions in which the heating of the LED is below 500 °C in light devices in which a good heat dissipation is provided. Such devices are, for example, household lamps.

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