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Temperature and Time Stability of Luminiferous Coating on Basis of Yttrium Aluminium Garnet alloyed By Cerium

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Abstract

The effect of temperature and operation time on the value of quantum efficiency of aluminiferous coating is determined. It is shown that the basic contribution to the decrease of its value is made by thermal quenching of luminescence and degradation processes in the aluminiferous coating, caused by the physicochemical interaction of luminophore with silicon-organic ligament.

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1. Introduction

A semiconductor light source represents an optical instrument, which transforms electrical energy in radiation with the spectral distribution close to that of white light. A crystal on the basis of a GaInN heterostructure generates monochromatic radiation with a wavelength range of 450–460 nm that is partially transformed to nonmonochromatic radiation in the red–yellow range of the spectrum by means of the luminophore. The intermixture of radiation flows of the crystal and luminophore permits to receive white light with the correlated color temperature from 3000 to 6000°Kelvin. The value of the color temperature is defined by the ratio of radiation flows in the dark blue and red–yellow ranges of the spectrum. To transform the radiation of the crystal to the red–yellow, firm crystalline luminophores are used, which are mostly based on the powder of the yttrium aluminium and yttrium gadolinium garnets alloyed by cerium [1–3]. The powder consists of spherical or irregularly shaped grains in the size range from 3 to 30 microns. To ensure the strength and manufacturability, the powder is mixed with an organic silicon compound that is applied on the crystal surface in the form of a layer with thickness from 200 to 500 microns. When transiting through a composite layer, stimulating radiation is reflected and absorbed in separate grains of luminophore. As a result, there is luminescence in the powder grains that is diffused and partially absorbed while being spread in the layer.

2. Materials and Methods

Let us assume that the luminophore is directed-scattering with an emphasis of passing by the 'murky medium'. Then, by way of finding and comparing the directional diagrams of crystal radiation with and without luminophore, it is possible to find the absorption and radiation dispersion constants of the coating. Thus, the radiation flow passing from the crystal is absorbed by the luminophore and undergoes absorption and dispersion. Its axial value is expressed as follows:

$$I(d) = I_{\alpha}(d) + I_{\rho}(d) = I_0 \exp[-(\alpha + \rho)d], \quad (1)$$

where *I (d)* is the radiation flow passing through the luminophore; I_0 is the radiation flow entering the luminophore; $I_{\alpha}(d)$ is the radiation flow absorbed by the luminophore; $I_{\rho}(d)$ is the radiation flow diffused by luminophore; *d* is the thickness of the luminophore layer; α is the luminophore absorption constant; ρ is the luminophore scattering factor. It was experimentally found that the change of the absorption constant is typically in the range from 30 cm^{-1} to 70 cm^{-1}, and the scattering factor is in the range from 20 cm^{-1} to 30 cm^{-1}.

The characteristics of a specific luminiferous layer depend on the quantity of luminophore, its structure, the density of packing, the thickness uniformity of the layer, grain-size composition and other factors. If the size of particles in the coating is high, the coating then becomes non-uniform and unstable. The size of particles influences the value of dispersion exponent ρ. It is found that ρ is directly proportional to the total area of a cross-section of particles per unit of volume [2, 5]. Experiments with various fraction sizes of luminophore have confirmed this dependence. Scattering factors and luminophore absorption influence its quantum efficiency, and size reduction of particles below a certain limit leads to efficiency decrease. The intensity of luminophore luminescence depends not only on the value of the absorbed radiation flow, but also on the temperature: the higher the operational temperature, the weaker the luminescence. The reduction of output optical power due to increased probability of nonradiative transitions is characteristic of the effect of thermal quenching of luminescence.

The temperature dependence of the radiation output from luminophore follows Mott scattering [1]:

$$\eta(T) = \frac{\eta_0}{1 + A \exp(-\frac{E_a}{kT})}, \quad (2)$$

where *A* is the quenching constant; E_{α} is the apparent activation energy of luminescence quenching; η_0 is luminescence quantum yield at room temperature.

The increased temperature influences not only the luminescent emission, but also the width of the line Δλ. The increase of the line Δλ can be derived using formula [4]:

$$\Delta\lambda(T) = \Delta\lambda(0)\left[th\frac{h\nu_i}{2kT}\right]^{-\frac{1}{2}}, \quad (3)$$

where ν_i is the oscillation frequency of Ce ion.

The thermal conditions of luminiferous coating depend not only on the ambient temperature, but also on the additional heat liberation caused by the processes of radiation absorption by the luminophore.

The temperature of the luminiferous coating can be estimated by use of a known solution of the thermal conduction equation for a flat plate with a uniformly distributed source of heat.

The quantity of heat produced in the volume of the luminiferous coating with thickness L and characteristic size *a* per unit of time is defined by the formula [6]:

$$q_2 = \alpha L\alpha^2 I_0 \exp(-\alpha L), \quad (4)$$

where α is the absorption constant; I_0 is a light stream.

At the same time, the temperature distribution along the coating is subject to parabolic law; moreover, the peak temperature is reached on the coating surface:

$$\frac{T_{\max}}{T_1} = 1 + \frac{q_2 L^2}{2\lambda T_1}, \quad (5)$$

where T_1 is ambient temperature.

3. Results

The observational test of the assumptions was carried out with AWB-3 luminophore (China). The luminophore powder was mixed with organic silicon compound with the mass ratio of 1:5. The received mixture was applied in a layer of 200 microns onto a glass substrate surface of the size 1 x 1 cm (see Figure 1).

The samples were placed in a heat chamber. The luminescence excitation in the samples was performed by means of the light-emitting diode module with radiation power of 3 W of the light guide over a wave length of 465 nm. The measurements of the correlated color temperature and the values of the light flux in the selected ranges of radiation with the light filters of the source were carried out by means of AvaSpec-2048 fiber spectrometer in the temperature range from ambient to +100C. The temperature was controlled both by means of a thermoelectric couple located on the surface of the sample and of the noncontact method (Figure 2).

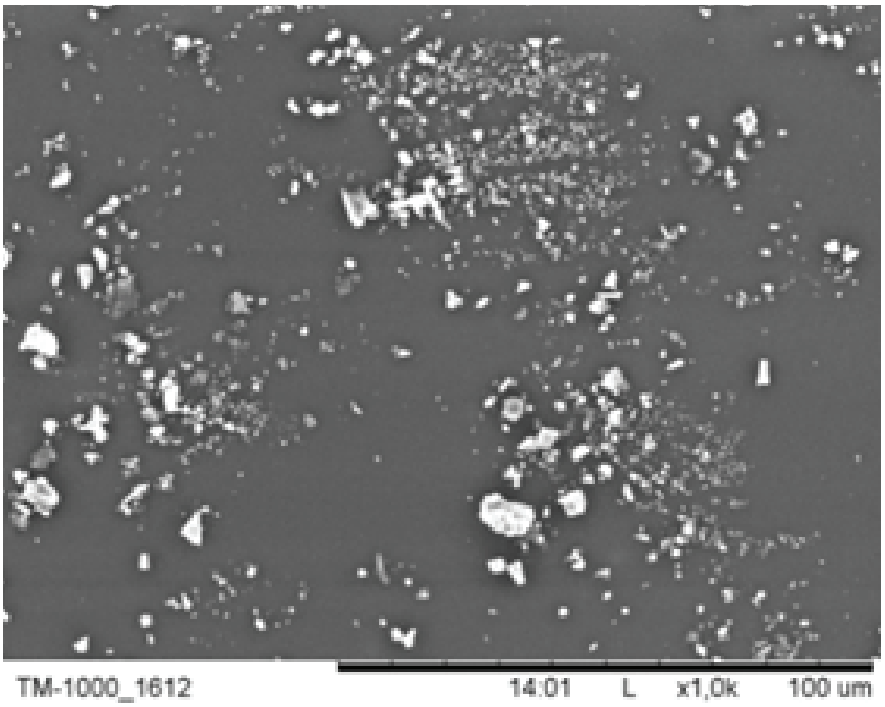

Figure 1: Luminophore distribution in organic silicon compound.

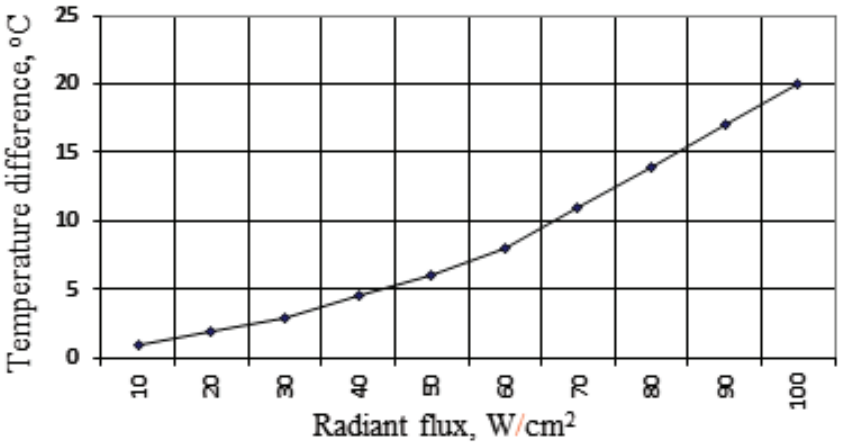

Figure 2: Dependence of increased temperature of luminophore from radiant flux.

Figure 2 shows that under the exciting radiation flux of 100 W/cm^2, the coating temperature is increased by 20°C.

Figure 3 shows the results of the measurement of luminous flux from the luminiferous coating. The results indicate that the luminous flux in the range of 420–480 nm practically does not change, whereas the luminous flux in the range from 500

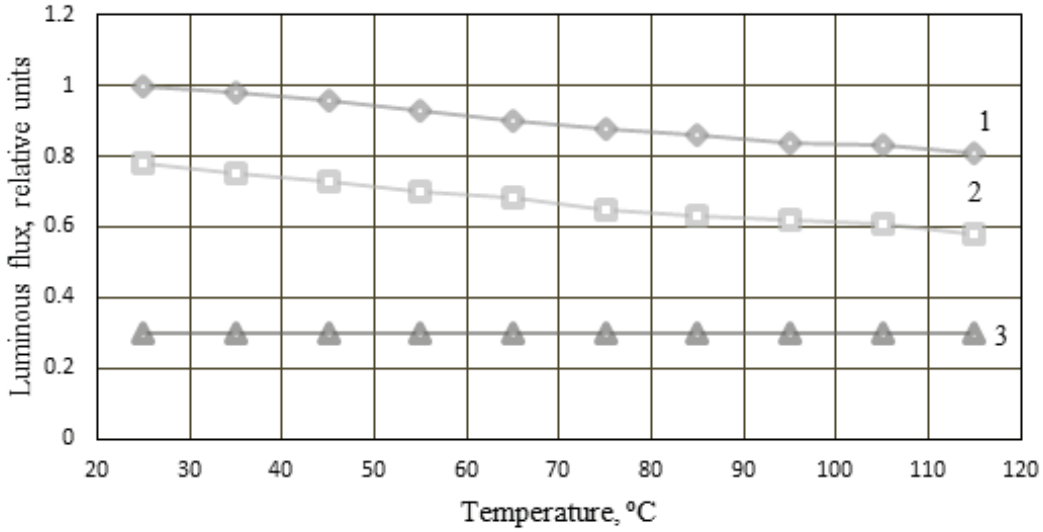

Figure 3: Dependence of the luminous flux of the luminophore coating from temperature. 1 – full luminous flux; 2 – luminous flux in the wavelength range from 500 to 800 nm; 3 – luminous flux in the wavelength range from 420 to 480 nm.

to 800 nm changes. This experiment confirms the determining influence of temperature dependence of luminophore quantum efficiency from the values of full luminous flux and the correlated color temperature. It also holds that under the range of temperatures used in the experiment, the dependence follows Mott law (3). The activation energy of the process of temperature luminescence quenching defined from the observational dependences lies within the limits from 0.1 to 0.12 eV, which is characteristic for inter-center radiation-less transition. The long-term operation of the coating at high temperature can lead to an irreversible process of decrease in the quantum efficiency of the luminophore. To determine the dependence of luminous flux and color temperature of luminiferous coating on time, the tests were conducted at the temperature of +85°C during 2000 hours, at the flux density of exciting radiation equal to 100 W/cm^2. Figure 4 shows the results of the tests.

4. Discussion

During the tests, the general decrease of light flux and increase of color temperature was observed. These changes are preferentially caused by physicochemical processes in luminophore and its interaction with organosilicon compounds. The apparent activation energy of the degradation process of luminous flux obtained from the experimental data was equal to 0.6–0.65 eV and the color temperature was 1.2–14 eV.

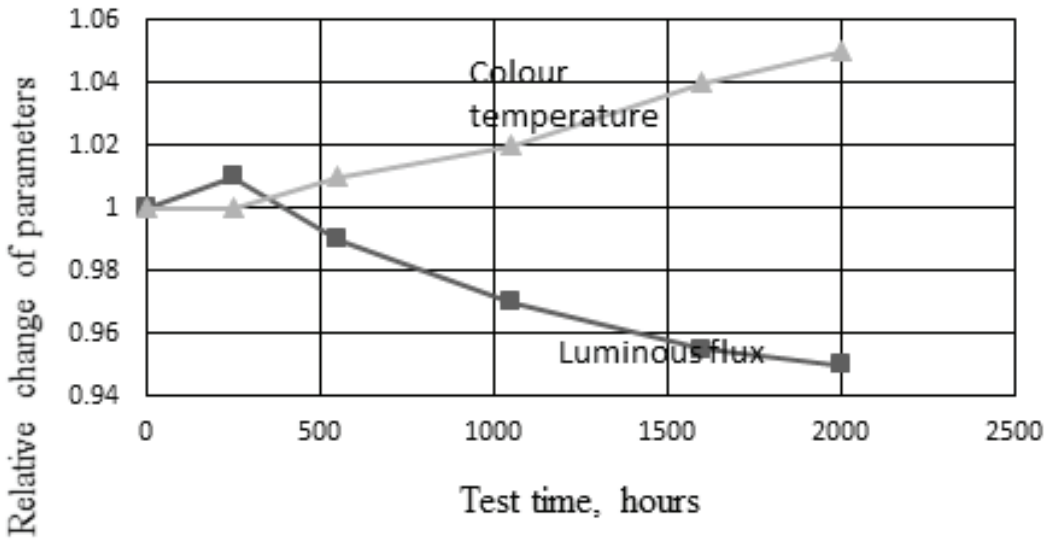

Figure 4: Change of the radiation flow value and color temperature during the test.

5. Conclusion

As a result of the undergone research, it was found that quantum efficiency of the luminiferous coating based on the powder of yttrium aluminium garnet alloyed by cerium has a strong dependence both from operation temperature and operation time. The decrease of the quantum efficiency of the luminiferous coating is caused by both a higher temperature and the physicochemical processes of degradation. The ratio change of light fluxes in the dark blue and red–yellow ranges also increase the color temperature of radiation.

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