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Optical model of light-converting film for white LED sources: Effect of distance between photoluminescent microparticles on specular reflectivity of exciting UV pump radiation

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Abstract. An optical model of a heterogeneous light-converting film based on a YAG:Ce^{3+} photoluminescent phosphor in epoxy resin is considered. The dependence of the distance between the phosphor microparticles in the suspension on the mass fraction of the powder and the densities of the components of the initial suspension is determined. Within the framework of the proposed model of the composite film, the coefficients of specular reflection and transmission within the region of isotropic scattering are simulated. It is demonstrated that optimizing the material composition by the amount of the photoluminescent component allows one to significantly reduce the coefficient of specular reflection of the UV pump radiation.

Keywords: turbid media, light-converting film, light absorption coefficient, yttrium-aluminum garnet (YAG), photoluminophore microparticle, white LED.

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1. Introduction

As a rule, white LED sources are built based on light-converting coatings with blue LEDs (460 nm) for pumping. Composite structures for fluorescent down-conversion of UV light are made of a suspension of photoluminescent powders with fractions of 1 µm to 15…20 µm in a binding agent, such as epoxy resin or silicone [1]. Phosphor powders with particle sizes of 5 to 10 µm are most commonly used, as white LEDs have maximum light output.

The efficiency of light conversion depends on a combination of propagation and absorption processes in such heterogeneous media. Passing through the suspension, the blue light of the LED pump can be absorbed, reflected from the surface of the dispersed phase particles, or scattered. The contribution of these processes depends on the particle size and their mutual arrangement. Ultimately, the efficiency of the light-converting layer is maximized at the optimum ratio between absorption and light scattering. The ratio between absorption (and, consequently, useful luminescence of the material) and light scattering (promoting uniform distribution of light in the material, including providing a short-wave contribution to the resulting white light emission) depends on both physical (density of components and mass fraction of powder in suspension) and geometric (powder particle size and distance between particles) parameters of the suspension. Recent studies have demonstrated the importance of optimizing both the spectral composition of lighting systems [2, 3] and the optical parameters of LEDs [4, 5] used in them to improve the overall efficiency and spectral controllability of such systems. It is obvious that to achieve a purposeful change of the ratio between absorption and light scattering, it is necessary to have an analytical dependence of the distance between particles and the mass fraction of powder particles in suspension.

2. A model of the light-converting film of a YAG:Ce^{3+} photoluminophore. Hypothetical cube

In [6, 7], a simple model of a light-converting film was considered, which allowed us to determine for the first time the value of the absorption coefficient of microparticles of aluminum oxide garnet powder doped with cerium. In this model of the composite film, it was assumed that the constituent components of the suspension – phosphor microcrystals and binder – have the same density. This naturally led to a decrease in the accuracy of the determination of the absorption coefficient of the phosphor powder microparticles and was acceptable only for their rough experimental evaluation. In this work, we used a more advanced approach to detail the analytical

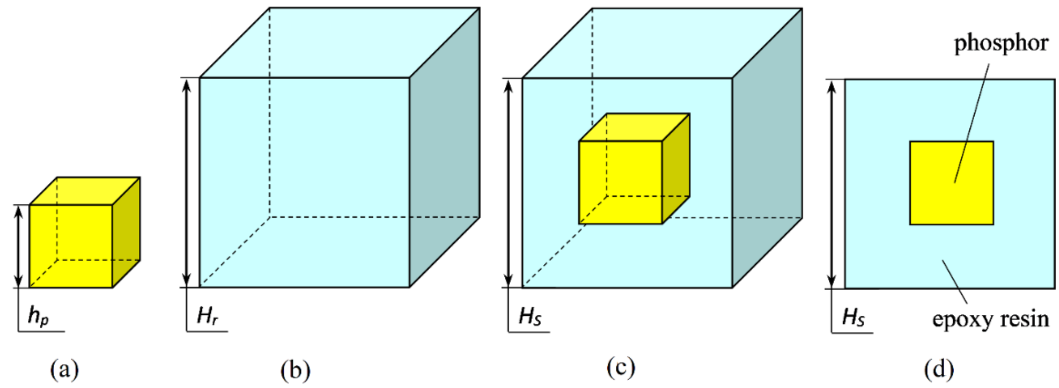


Fig. 1. Schematic representation of the main elements of the optical model: (a) phosphor cube, (b) homogeneous resin cube, (c) heterogeneous cube, (d) frontal projection of heterogeneous cube.

relationship between the physical (phosphor and epoxy resin densities, powder particle absorption coefficient) and geometric (size) parameters of the phosphor powder in a homogeneous and degassed suspension.

Let us consider an optical model of a light-converting film made from a photoluminescent suspension of cubic powder microcrystallites in an epoxy resin with a total mass of phosphor powder and epoxy resin *W*. The weight of one microparticle (cube) will be equal to $\rho_p \cdot h_p^3$, where ρ_p – density of phosphor material, h_p – length of cube edge. Let the mass fraction of phosphor particles in the suspension be equal to *C*. Then, the total weight of all *n* phosphor particles in suspension will be equal to

$$\rho_p \cdot h_p^3 \cdot n = C \cdot W . \quad (1)$$

In this case, the weight of epoxy resin binder in the suspension will be equal to $(1-C) \cdot W$, and, per phosphor microparticle, accordingly, – $(1-C) \cdot W/n$. The weight of one homogeneous resin cube will be equal to $\rho_r \cdot H_r^3$, and the weight of all the resin in the suspension will be equal to

$$\rho_r \cdot H_r^3 \cdot N_r = (1-C) \cdot W . \quad (2)$$

Here, N_r is the number of all homogeneous cubes of resin when making a suspension of weight *W*, and ρ_r is the density of resin. Obviously, the number of homogeneous resin cubes will be equal to the number of phosphor cubes, *i.e.* $n = N_r$.

Dividing equation (2) by equation (1), we obtain

$$\frac{\rho_r}{\rho_p} \cdot \frac{H_r^3}{h_p^3} = \frac{1-C}{C} . \quad (3)$$

The height of the homogeneous resin cube can be determined from Eq. (3) (Fig. 1b)

$$H_r = h_p \cdot \sqrt[3]{\frac{\rho_p}{\rho_r} \cdot \frac{(1-C)}{C}} . \quad (4)$$

Let's place a phosphor cube (Fig. 1a) inside a homogeneous resin cube (Fig. 1b) in the center of a homogeneous resin cube (Figs. 1c and 1d). In the following discussion, we will refer to such a heterogeneous cube of resin and phosphorus as a heterogeneous cube. The volume of the heterogeneous cube (V_s) is larger than the volume of the homogeneous cube made of binder material by the value of the volume of the cube (h_p^3) made of phosphor. We have

$$V_s = H_s^3 = H_r^3 + h_p^3 . \quad (5)$$

Here, H_s is the height of the heterogeneous cube (Figs. 1c and 1d), $H_s > H_r$. Combining equations (4) and (5), we obtain the height of the heterogeneous cube:

$$H_s = h_p \cdot \sqrt[3]{\frac{\rho_p \cdot (1-C)}{\rho_r \cdot C} + 1} . \quad (6)$$

Since each non-uniform cube contains only one cube of phosphorus, the number of non-uniform cubes N_s in a suspension of weight *W* will be equal to the number of cubes of phosphorus, *i.e.*, $N_s = n = N_r$. As follows directly from Eq. (6), the height of the heterogeneous cube (H_s) depends linearly on the height of the phosphor cube (h_p) and nonlinearly on the weight concentration of phosphor particles (*C*), and on the densities of phosphor and resin (ρ_p and ρ_r), respectively.

At the next stage, we construct a row from a certain number of heterogeneous cubes, as shown in Fig. 2a. Let the number of cubes in a row be equal to $\sqrt[3]{N_s}$ pieces). The distance between the centers of heterogeneous phosphor cubes is equal to H_s.

By arranging rows of cubes into an ordered three-dimensional structure, we obtain the final model of a heterogeneous film – hypothetical cube, where the lengths of edges are equal to $H_s \cdot \sqrt[3]{N_s}$, and the nearest phosphor cubes are at distances H_s along the axes *X*, *Y*, and *Z* (Fig. 3).

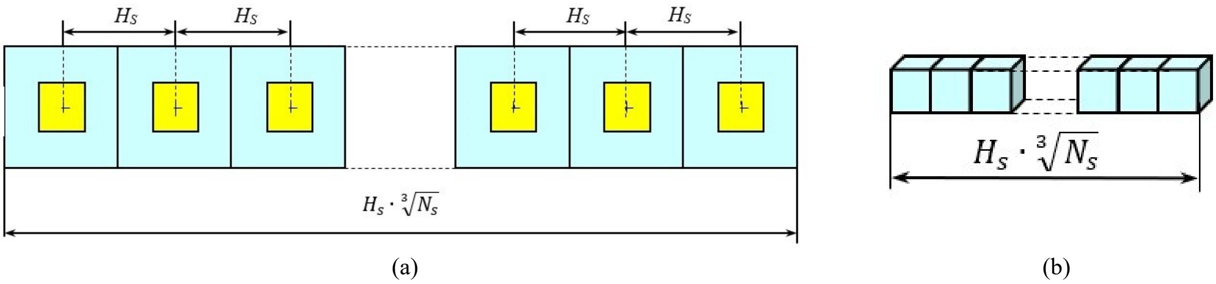


Fig. 2. Schematic representation of a row of cubes arranged in a row: (a) frontal projection of several heterogeneous cubes (crosses mark the positions of the centers of phosphor cubes), (b) reduced image of the frontal dimetric projection of several heterogeneous cubes shown in (a).

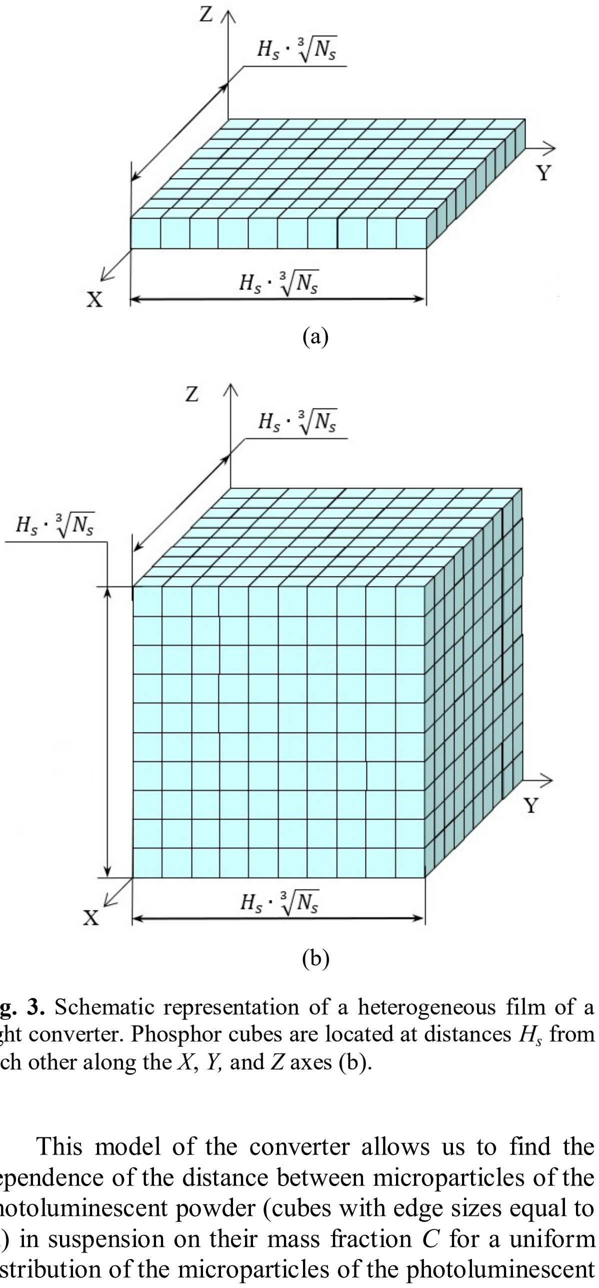


Fig. 3. Schematic representation of a heterogeneous film of a light converter. Phosphor cubes are located at distances H_s from each other along the *X*, *Y*, and *Z* axes (b).

This model of the converter allows us to find the dependence of the distance between microparticles of the photoluminescent powder (cubes with edge sizes equal to h_p) in suspension on their mass fraction C for a uniform distribution of the microparticles of the photoluminescent powder. It is obvious that this model best describes materials obtained from carefully homogenized suspensions, where the average distance between photoluminescent microparticles will be determined only by their mass fraction and the densities of the suspension components.

3. Dependences of the parameters of the solid-state converter model on the mass fraction of particles *C* of YAG:Ce powder

According to the considered model of a turbid medium, the distance between the centers of photoluminophore particles in homogeneous and isotropic suspension is equal to the height of the total suspension cube H_s (Figs. 2 and 3), the analytical expression of which is represented by Eq. (6). The distance between the centers of photoluminophore cubes in homogenized and degassed suspension depends on the size of phosphor cubes, the mass fraction of powder particles C (cubes) in the suspension, and the densities of phosphor and binder (epoxy resin).

The dependence of the height H_s on the height of the YAG:Ce phosphor model cube h_p is shown in Fig. 4. It is known that the distance between uniformly distributed YAG:Ce photoluminophore particles in the epoxy resin depends linearly on the height of the YAG:Ce [8] and phosphor model cube h_p [9]. That is, the larger the size h_p of the phosphor powder used, the larger the size H_s of the individual cells of the turbid medium model will be (at a given value of C). In this case, the size of such a cell H_s will depend on the size of the crystallite h_p, the weaker the more phosphor there is in the suspension (Fig. 4). It is worth noting that at the considered parameters of materials and powder mass fraction (0.1…0.4) the cell size of the turbid medium model (H_s) is approximately 2-3 times larger than the crystallite size (h_p) and grows with decreasing the powder fraction for a crystallite of some specific size.

The dependence of the distance H_s between the particles of YAG:Ce powder in material on the mass fraction of C is shown in Fig. 5. Fig. 5 shows that the distance between uniformly distributed particles of photoluminophore YAG:Ce [10] in epoxy resin nonlinearly depends on the mass fraction of powder particles C in suspension, as it directly follows from Eq. (6). The larger the phosphor crystallite size, the larger the distances between particles with decreasing fraction of powder in the suspension.

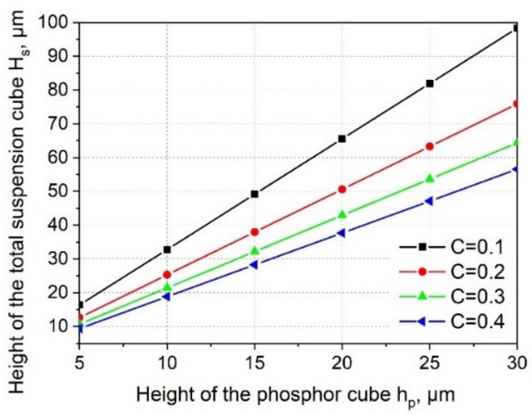


Fig. 4. Dependence of the height of the total suspension cube H_s on the height of the phosphor cube h_p for different values of the mass fraction of the powder C.

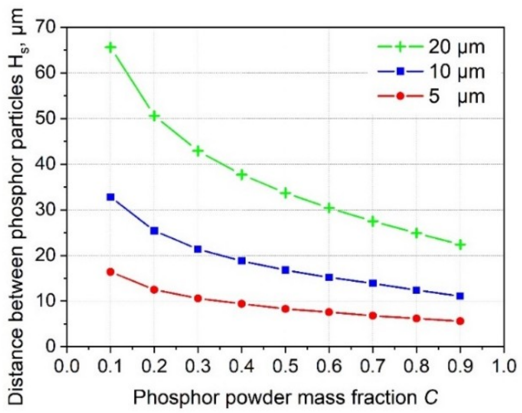


Fig. 5. Dependence of the distance between phosphor particles on their mass fraction.

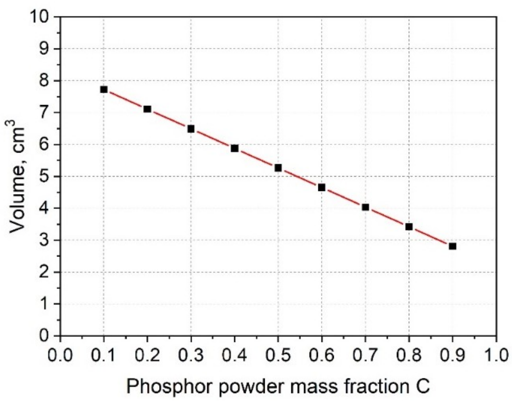


Fig. 6. Dependence of volume of hypothetical cube of suspension on mass fraction C of phosphor powder at W = 10 g.

Fig. 6 shows the dependence of the volume of a hypothetical cube (*i.e.*, the total volume of the suspension) on the mass fraction C of the phosphor at a constant mass (W = 10 g) of the suspension of the phosphor powder and binder.

From the data presented in Fig. 6, it is evident that the volume of the suspension depends linearly on the mass fraction of the photoluminescent powder. It follows that the projected area of the base of the hypothetical cube used to determine the light-shadow coefficient also depends on the mass fraction C of the photoluminescent powder.

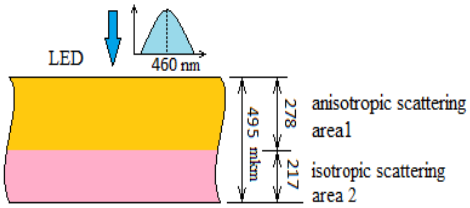
4. Modeling the coefficients of specular reflection and transmission of a layered structure of alternating layers of phosphor and binder of different thicknesses

As shown in Refs. [11, 12], when considering the propagation of light through a turbid medium obtained from a suspension of phosphor powder in epoxy resin, two qualitatively different regions can be distinguished. In the near-surface layer of the sample, on the side of the incident light, anisotropic light scattering occurs (Fig. 7a). Region 1 has a thickness of less than 278 µm for the case of a YAG:Ce powder of about 5 µm in size in epoxy resin [1, 11, 12]. At greater thickness, isotropic scattering is observed (Fig. 7a, region 2 with a thickness of 217 µm) with a spherical indicatrix.

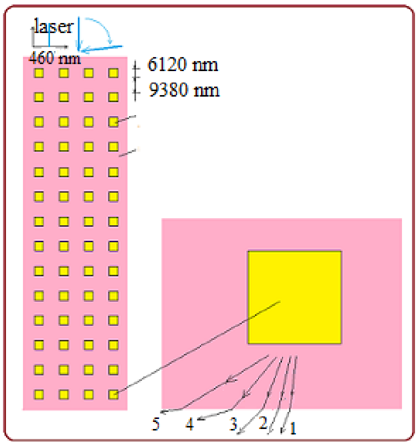
When light propagates in a turbid medium, a laser beam normally incident on the film surface, propagating in region 1, significantly expands (the so-called Tyndall cone) due to a significantly higher scattering coefficient than absorption [1, 16]. This means that laser radiation in the isotropic scattering region 2 will fall on the surface of phosphorus crystallites in a wide range of angles. This determines the appropriateness of analyzing the angular dependence when analyzing the processes of specular reflection and transmission in the isotropic scattering region 2.

To determine the effect of the film thickness on the optical properties of photoluminescent films, the reflection and transmission coefficients of films of various thicknesses were simulated. Model samples of films with heterogeneous cube dimensions H_s = 15500 nm were selected for analysis [13]. With a mass fraction of phosphor C = 0.2, this corresponds to a microcrystal with h_p = 6120 nm placed in the center of the cube, surrounded by a 4690 nm layer of resin binder. The simulation was performed using WinSpall 3.02 software [14] by numerically solving Fresnel equations for a layered structure when the angle θ of the incident radiation varied within (0…90)°, where "0" degrees corresponds to the normal incidence, and "90" – sliding along the surface of the film. Considering the selected crystallite sizes, the model is a set of plane-parallel layers: the phosphor layer has a thickness of 6120 nm, and the epoxy resin layer has a thickness of 9380 nm. In the case of 3 layers of epoxy resin and 3 layers of phosphorus, forming 3 H_s, we have a region of anisotropic scattering and 28 layers (14 layers of epoxy resin and 14 layers of phosphorus, forming 14 H_s), correspond to an isotropic scattering regime.

These calculations are presented as a dependence of the specular reflection coefficient (and transmission) on



(a)



(b)

Fig. 7. Schematic diagram of turbid media light-converting film pumping with a 460 nm LED (a) and diagram of pumping area 2 of the suspension film with a laser, λ = 460 nm (b).

the angle of incidence of light on a film, which is a stack of plane-parallel plates simulating layers of epoxy resin (n_r = 1.55 + i·0, thickness 9380 nm) and phosphorus (n_p = 1.8469 + i·0.000198, thickness 6120 nm). The film is inside the binder (n_r = 1.55 + i·0), the wavelength of light is 460 nm. The calculated optical model is placed in a binder layer to eliminate the influence of specular reflection at the boundary with air, a medium with a low refractive index. The presence of a binder layer necessitates the introduction of an additional semi-infinite final layer (seventh and twenty-ninth) of epoxy resin in calculations. The first layer is also a semi-infinite layer of epoxy resin in any case (which corresponds to the requirements for the thicknesses of the first and last layers of the WinSpall program). The calculation results allow us to estimate the amount of light that is reflected from this structure and, accordingly, cannot be used either to excite the phosphor or to add "blue" light to the phosphor radiation to obtain white light. Thus, the less specularly reflected light from such a model structure, the higher its efficiency as a converter of ultraviolet radiation. It should also be noted that the presented angular dependence is essentially similar to the configuration when the direction of propagation of the exciting light is constant, and the crystallite faces are rotated relative to it by a certain angle. Thus, it is possible to estimate which range of angles of incidence of light on equally located crystallites is more effective. Under normal manufacturing conditions, of course, a uniform distribution of crystallite orientation is most likely – in this case, the efficiency is estimated by the value of the integral scattering: the smaller it is, the higher the potential efficiency of the structure.

The results of modeling composite films of different thicknesses (consisting of different numbers of identical layers having an infinite length along the film surface) are presented in Fig. 8 and Fig. 9 for 28 layers (14 model cubes) and for 6 layers (3 model cubes), respectively. When the film thickness is reduced to three model cubes, the reflection coefficient corresponding to the first peak decreases from 0.78 to 0.18 in the area of incidence angle up to 60 degrees. The corresponding transmission coefficient increases from 0.63 to 0.9.

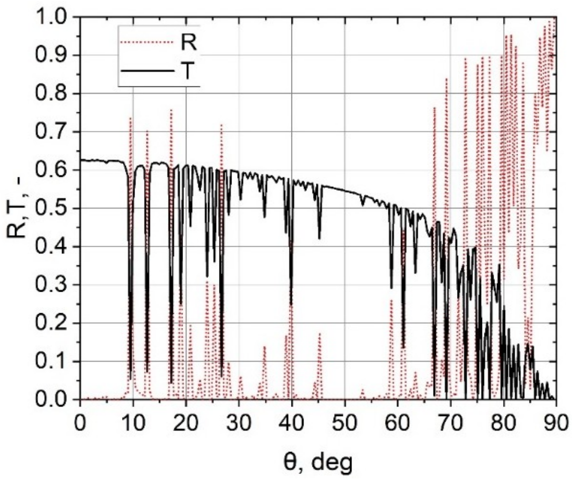


Fig. 8. Angular spectrum of specular reflection and transmission in area 2 (an isotropic scattering regime) of a suspension film with a thickness of 28 alternating layers of epoxy resin and phosphorus (14 model cubes H_s).

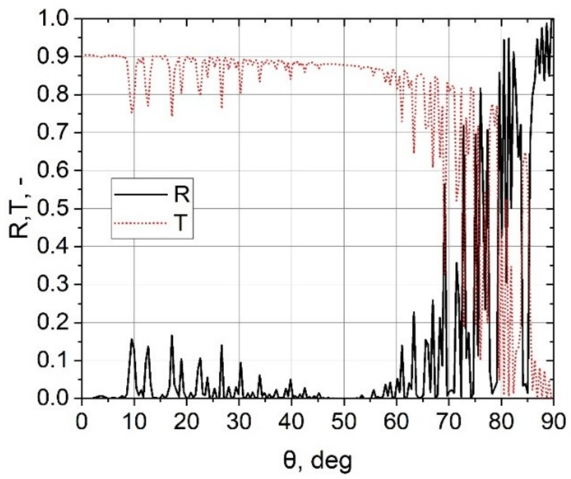


Fig. 9. Angular spectrum of reflection and transmission reflections in region 2 (isotropic scattering regime) of a suspension film with a thickness of 6 layers of epoxy resin and phosphorus (3 model cubes H_s).

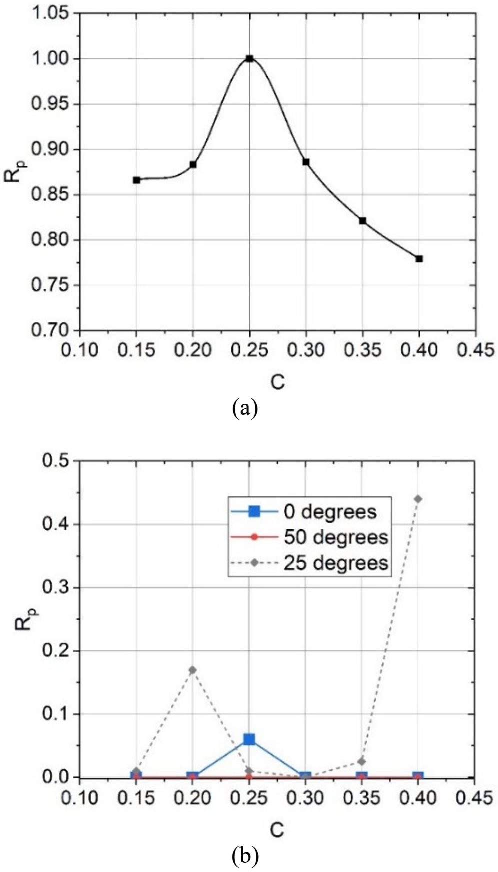


Fig. 10. Dependence of the magnitude of specular reflection on mass fraction of phosphor powder of a layered film structure with the thickness 217 μm and the model cube size H_s = 15500 nm (28 layers) for various angles of incidence (a) and integral dispersion within 0…90 degrees of incidence (b).

An analysis of angular dependences shows the presence of two selected areas where the intensity of specular reflection is minimal – approximately 0…9 and 45…55 degrees. At the same time, the greatest losses due to specular scattering are observed at incidence angles greater than 60 degrees. This allows us to recommend the development of methods for partial orientation of crystallites in the film, which will significantly increase its efficiency as a light-converting coating. If the "transparency region" and minimal reflection at angles of incidence close to the normal are not surprising, then the window in the 50-degree region raises some questions. Modeling has shown that this effect is associated with the implementation of the total internal reflection mode in the crystal phosphor layer. In this case, the light is localized in the layer, which, of course, not only reduces losses due to specular reflection but also promotes effective absorption in the material layer by increasing the trajectory of the beam in the phosphor region. This method of increasing the "absorption capacity" of the heterogeneous material due to the localization of light in the volume of individual crystallites seems very promising, but requires further detailed analysis of this potential mechanism for increasing the light conversion efficiency.

A comparison of the results of calculating the angular dependences of reflection and transmission, performed for films of various thicknesses, consisting of 14, 12, 10, 8, 5, and 3 identical model cubes with H_s = 15500 nm, showed that the positions of the peaks of specular reflection and transmission remain constant regardless of the thickness of the heterogeneous film. The shape of angular dependences is the result of interference in a layered system with uniformly distributed layers of the same composition. This is confirmed by the results of modeling with other characteristic dimensions of the layered structure – a change in the distance between the crystals, or their size, leads to changes in the position of the peaks of specular reflection and transmission on the angular dependences. This means that the angular position of the peaks of specular reflection and transmission can be used to estimate the distances between the phosphor particles, of course, in the case of their homogeneous spatial organization. The development of this approach requires additional research on real systems.

Fig. 10a shows the dependence of the magnitude of specular reflection on mass fraction of phosphor powder of a layered film structure with a thickness of 217 μm and a model cube size H_s = 15500 nm (28 layers) for various angles of incidence. As follows from the calculation results, the most effective angle for all values of C is 50°.

As it was noted above, this model can also be used to estimate an arbitrarily oriented set of crystallites in a film. In this case, the value of integral scattering in the entire range of angles will be informative. These data for a "uniform distribution of crystal orientation" (the same contribution for each angle) are shown in Fig. 10b. One can see that the optimization of the mass fraction of phosphor crystals from 0.25 to 0.4 allows one to reduce losses due to specular reflection by approximately 20%, which will significantly increase the efficiency of the structure. At the same time, an "unsuccessful" choice of the mass fraction of photoluminescent powder can significantly reduce the efficiency of the structure simply by increasing the contribution of the scattering compared to specular reflection.

5. Conclusions

An approach to the description of homogeneous light-converting films of photoluminophore is developed using the example of $YAG:Ce^{3+}$ powder in an epoxy resin binder. The dependence of the distance between luminophore microparticles in a suspension on the mass fraction of powder and the density of the suspension components is determined. The insights of a model cube for a suspension with a given radiating fraction of the luminophore, the size of which H_s corresponds to the distance between the luminophore crystals, introduced in the work, allows one to simulate the angular distribution of the specular reflection coefficients and evaluate the efficiency of the selected structure for generating white light.

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Оптична модель світлоперетворювальної плівки для білих світлодіодних джерел: вплив відстані між фотолюмінесцентними мікрочастинками на дзеркальну відбивну здатність збуджуючого УФ-випромінювання

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Анотація. Розглянуто оптичну модель гетерогенної світлоперетворювальної плівки на основі фотолюмінесцентного люмінофора YAG:Ce^{3+} в епоксидній смолі. Визначено залежність відстані між мікрочастинками люмінофора в суспензії від масової частки порошку та густини компонентів вихідної суспензії. В рамках запропонованої моделі композитної плівки змодельовано коефіцієнти дзеркального відбивання та пропускання в області ізотропного розсіювання. Показано, що оптимізація складу матеріалу за кількістю фотолюмінесцентного компонента дозволяє суттєво зменшити коефіцієнт дзеркального відбиття УФ-випромінювання накачки.

Ключові слова: мутне середовище, світлоперетворювальна плівка, показник поглинання світла, ітрій-алюмінієвий гранат, мікрочастинка фотолюмінофора, білий світлодіод.