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Published in:
Applied Physics Letters

Link to article, DOI:
10.1063/5.0073168

Publication date:
2021

Document Version
Peer reviewed version

Design of a β-SiAlON:Eu based phosphor-in-glass film with high saturation threshold for high-luminance laser-driven backlighting

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Abstract

β-SiAlON:Eu possesses an intense and narrow emission band peaked at ~530 nm, which makes it a highly suitable green-emitting phosphor for LCD backlight. However, it can hardly be used in high-power laser displays or projectors because of the laser-induced saturation effect. The aim of the present study is to develop a β-SiAlON:Eu based phosphor-in-glass (PiG) film that can withstand high-power density laser irradiation. A facile blade-coating method was employed to fabricate the PiG film. The morphology, quantum efficiency, luminescence spectra and laser-pumped colorimetric properties of the β-SiAlON:Eu PiG films were investigated. Under excitation of 450 nm, a typical PiG film showed internal quantum efficiency of 58%, which is an acceptable value. The laser-pumped photometric properties of the PiG films were measured in reflection mode. When irradiated with a blue laser, the PiG film attained a high luminous efficacy of 109 lm/W. Furthermore, its saturation threshold reached over

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26.4 W/mm^2, thus achieving a high lumen density of 1970 lm/mm^2 and a high luminous exitance of 1052 lm/mm^2. With the above excellent properties, the β-SiAlON:Eu PiG film has great potential for use in high-power and wide color gamut laser displays or projectors.

The development of the LED based light source has been encountering a serious obstacle, which is the difficulty of achieving high luminance. This is mainly caused by the "efficiency drop" problem in LEDs, to which ~~that~~ there is no effective solution for now. A laser diode (LD) does not suffer from the "efficiency drop", which means that it can theoretically ~~can~~ achieve much higher luminance than LEDs. Therefore, laser lighting has ~~been becoming~~become a competitive alternative in the field of high-luminance lighting devices.[1,2,3]

Similar to white LEDs, the most popular approach to realize white light emission for laser lighting is based on pumping phosphor(s) by blue LDs.[4,5] Regarding this, the phosphor is a key component determining the colorimetric and photometric properties of a laser lighting device. Considering the high-power laser irradiation, the "phosphor + resin" encapsulation is not feasible for laser lighting due to the low thermal conductivity and poor thermal stability. Therefore, all-inorganic phosphors (i.e., ceramics, single crystals, phosphor-in-glass) with high thermal conductivity and robust heat resistance are in great demand. In the last five years, a series of all-inorganic phosphors with high saturation threshold for laser lighting were reported, including $(Y/Lu)_3Al_5O_{12}$:Ce, $CaSiAlN_3$:Eu, $La_3Si_6N_{11}$:Ce, etc.[6-12] These phosphors possess wide emission bands, which ~~is~~ are beneficial for~~to~~ high color rendering. In the application of laser displays or projectors, phosphors with narrow emission are superior to those with wide emission bands due the possibility of achieving wide color gamut.[13-15] $Si_{6-z}Al_zO_zN_{8-z}:Eu^{2+}$ (β-SiAlON:Eu) with an intense and narrow emission band is widely used as a green emitter in wLED-based backlights.[13,14,16,17] ~~Aiming to apply~~With the aim of applying β-SiAlON:Eu in laser lighting, several variations of β-SiAlON:Eu based phosphor-in-glass (PiG) were developed. Xie et. al reported a PiG sheet that saturated at 0.7 W/mm^2.[16] To improve the saturation threshold, Xiang et. al reported a PiG sheet using a SiO_2-Na_2CO_3-H_3BO_3-CaO glass.[17] The saturation threshold increased to more than 4 W/mm^2, resulting in a maximum luminous flux of 363 lm. However, there is still room for further improvement.

~~Because~~As the PiG is brittle, it is very difficult to machine it to thinner than 200 μm. Many studies have proved that thinner samples have higher saturation threshold due to the faster heat dissipation. Thus, the saturation thresholds of PiG sheets were limited. A PiG film is much thinner (<100 μm) than a PiG sheet, which is beneficial for achieving a higher saturation threshold. Our previous studies found that the saturation threshold of a $CaSiAlN_3$:Eu based film (>9 W/mm^2) is significantly higher than the corresponding PiG sheet (~0.5 W/mm^2).[18,19] This indicates that a β-SiAlON:Eu based PiG film may have great potential to attain high saturation threshold. Progress was made by Wang et. al, who developed a β-SiAlON:Eu-PiG film in a transmission configuration. The saturation threshold reached ~8 W/mm^2 and a maximum luminous flux of ~680 lm was attained.[13] However, the saturation threshold still needs to be further improved and some important issues, like the luminous exitance or luminance have not been addressed. Previous studies found that a phosphor in reflection configuration possessed better heat dissipation than ~~that~~ in transmission configuration, due to the shorter distance between the laser irradiation spot and heatsink.[8,18] Thus, with above survey on all-inorganic phosphors for laser lighting, β-SiAlON:Eu-PiG film in a reflective configuration is a promising solution.

The aim of this study is to develop a β-SiAlON:Eu-PiG film with high saturation threshold. The PiG film was fabricated in a reflective configuration on a corundum substrate, showing an internal quantum efficiency of 58% and a saturation threshold of over 26.4 W/mm^2. A high lumen density of 1970 lm/mm^2 and a high luminous exitance of 1052 lm/mm^2 were thus attained. A facile and economic blade-coating synthetic route was employed.

The pristine β-SiAlON:Eu powder is commercially available from Nakamura-Yuji Co. Ltd. A ZnO-B_2O_3-SiO_2 ($T_g \approx 390$ °C) glass was used as encapsulation material. The glass/phosphor mass ratio was 150%. The heating temperature was 460 °C and the soaking time was zero. More detailed information of the coating process was presented in our previous work.[18]

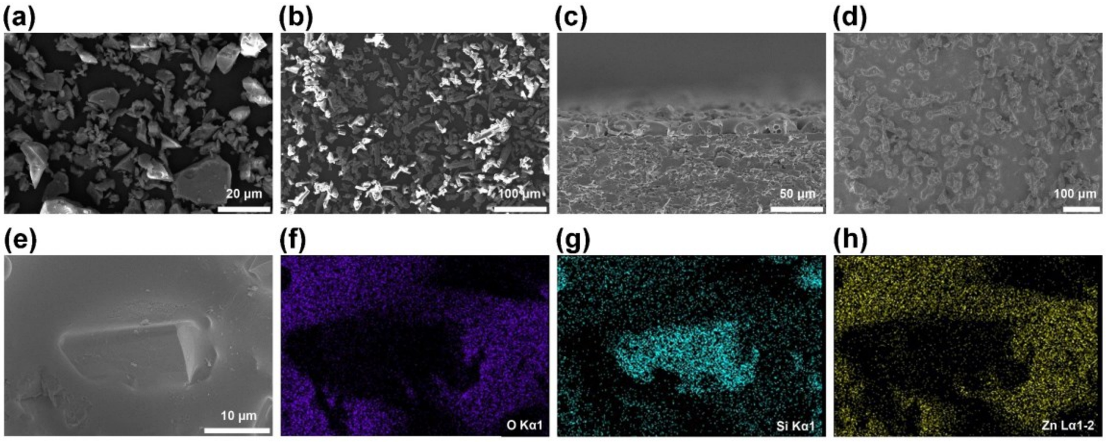

FIG. 1. (a-e) SEM images of the glass power, β-SiAlON:Eu phosphor powder, cross-section of the PiG film, surface of the PiG film and surface of the PiG film with high magnification, respectively; (f-h) corresponding EDS element mapping of O, Si and Zn, respectively.

The morphologies of the starting materials and the PiG film were observed using a scanning electron microscope (SEM, ZEISS, Merlin Compact) equipped with an energy disperse X-ray spectroscope (EDS). The results are presented in Fig. 1. The particle size of the glass frits was in the range of 2–20 μm [Fig. 1(a)]. The β-SiAlON:Eu phosphor powder presented a rod-shaped morphology with an average equivalent particle size of ~20 μm [Fig. 1(b)]. Fig. 1(c) shows the cross-sectional interface of the PiG film. Note that the film was tightly bonded to the corundum substrate, indicating a good adhesion and reliability. Fig. 1(d) shows the surface of the PiG film indicating that the glass completely melted, and the phosphor powder was uniformly dispersed in the glass matrix. EDS mappings were conducted on the surface [Fig. 1(e)] of the PiG film. As shown in Fig. 1(f-h), the β-SiAlON:Eu particles are rich in Si; in contrast, the glass matrix is rich in O and Zn. Note that slight element diffusion occurred at the boundary, indicating that there could be interface reaction between the particle and glass matrix. The internal quantum efficiency (IQE) was measured using a spectrofluorometer

(Edinburgh, Fls-1000) equipped with an integrating sphere (d = 150 mm) and a 450 W Xe lamp. At an excitation wavelength of 450 nm, the PiG film showed an IQE of 58%, which is an acceptable value. Considering the corundum substrate ($Al_2O_3:Cr^{3+}$) could absorb a small part of the luminescence from β-SiAlON:Eu, the IQE could be further improved by selecting more suitable substrate.

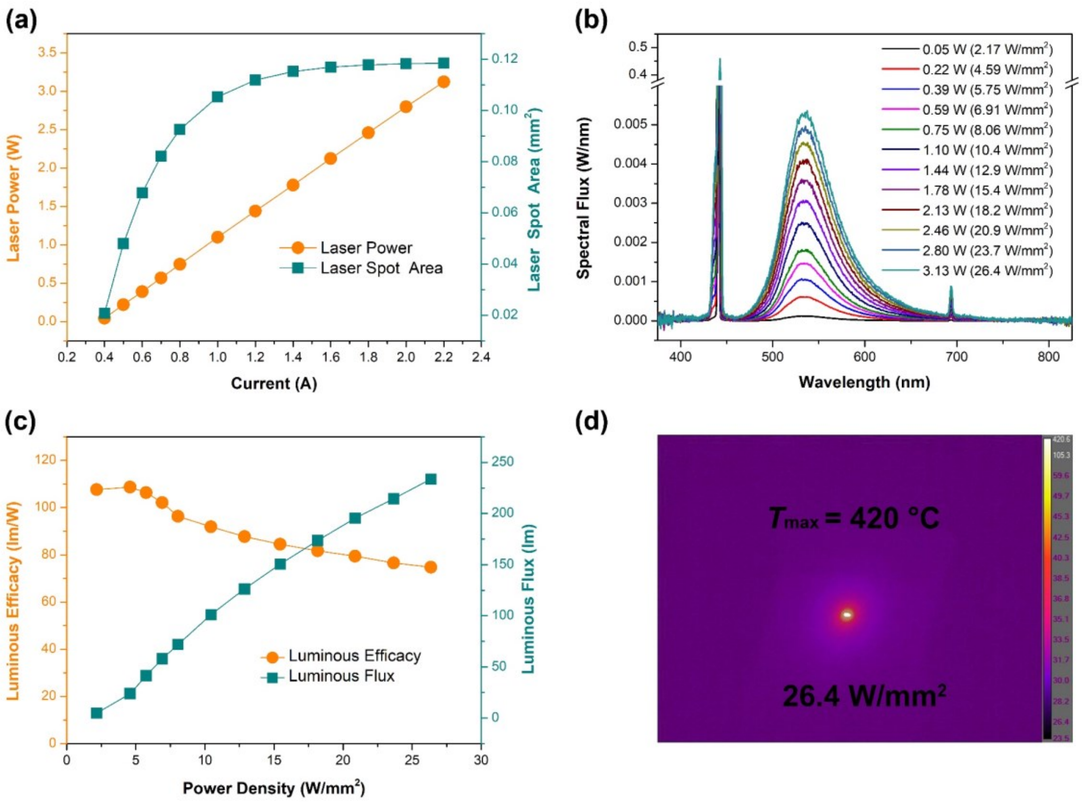

FIG. 2. (a) driving current dependent variation of the laser power and laser spot area of the LD; (b) total spectral flux from the PiG film measured under different laser power/power densities; (c) luminous flux and luminous efficacy of the PiG film as a function of the power density; (d) infrared thermal image of the film under 26.4 W/mm^2 laser irradiation.

The luminescence saturation behavior of the PiG film was investigated in a very wide laser power density range from 2.17 W/mm^2 to 26.4 W/mm^2. A custom-built sphere-spectroradiometer measurement system was used including an integrating sphere (Labsphere, RT-060-SF) fiber-coupled to an array spectrometer (Instrument Systems, CAS-140-CT-151). The detailed information of the measurement setup was presented in our previous studies.[18] The driving current dependent variation of the laser power and laser spot area of the LD are shown in Fig. 2(a). The laser power increased linearly with the current. In contrast, the laser spot area increased almost logarithmically with current. The reason for this increase in spot area is because of the physics of broad-area diode lasers, where more spatial modes appear in the slow axis direction of the output beam, when the current is increased. This underlines the importance of precise measurements of the spot size in order to estimate the laser power density. Here, we have defined the laser spot area at the $1/e^2$ intensity level. The laser power density dependent emission spectra of the PiG film were investigated, and the results are shown in Fig. 2(b). The spectral peak remained stable in center wavelength around 535 nm for all power levels, while the spectral width (FWHM) increased from 55 nm at low power levels to 64 nm at maximum input power. A rise in the temperature intensifies vibrations of the crystal lattice, which results in a change of the Eu-N bond length. This affects the crystalline field and enhances the nephelauxetic effect that ultimately leads to the increase in the FWHM.[20] From 2.17 W/mm^2 to 26.4 W/mm^2, the emission intensity of the PiG film increased almost linearly with increasing incident power density, showing no sign of luminescence saturation. The luminous efficacy and luminous flux of the PiG film as a function of incident laser power density are shown in Fig. 2(c). The emitted luminous flux increases monotonously in the entire power density range, indicating a high saturation threshold of over 26.4 W/mm^2 (3.13 W). The maximum luminous flux reached 235 lm. This value is limited by the maximum laser power. The initial luminous efficacy (at 4.59 W/mm^2) reached 109 lm/W, however, decreased steadily with the increasing laser power density. At 26.4 W/mm^2, the luminous efficacy declined to 75 lm/W. It is worth noting that the luminous efficacy of the PiG film is lower than that in previous report (150 lm/W).[17] The way to improve the luminous efficacy will be systematically investigated later on, including design of microstructure, using more suitable glass bonding material and optimizing the heating process and film thickness. The temperature of the PiG film was monitored with a thermal camera (FLIR A655sc) equipped with a focusing lens (FLIR T198059) to obtain a resolution of approximately 50 µm after removing the integrating sphere. The phosphor temperature increased almost linearly with input laser power and reached a maximum temperature of 420°C. An example thermal camera image at maximum input laser power is given in Fig. 2(d). Here, also the slightly elliptical shape of the laser spot can be seen. The relatively modest decrease in luminous efficacy seen in Fig. 2(c) demonstrates the high stability against thermal quenching of β-SiAlON:Eu also demonstrated previously, while also demonstrating the benefits of using a thin PiG film on a corundum substrate with high thermal conductivity.[16,21]

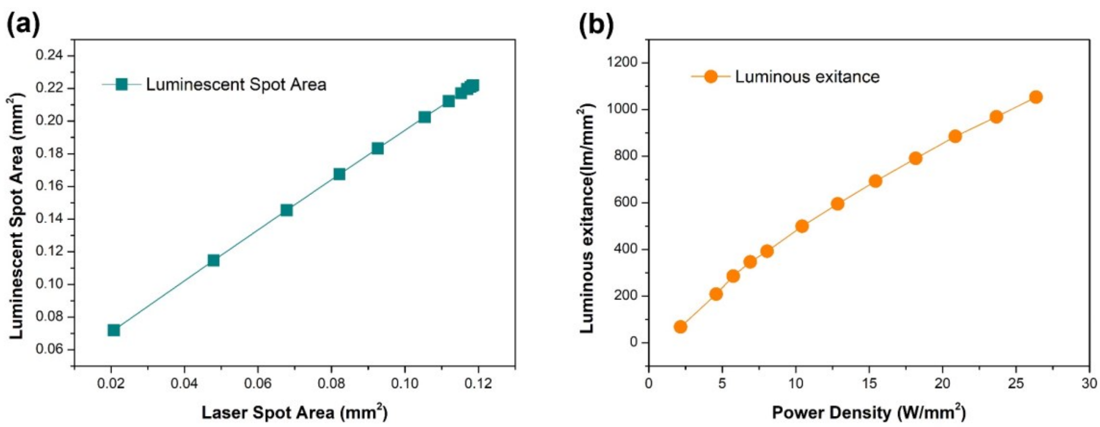

FIG. 3. (a) luminescent spot area of the PiG film as a function of incident laser spot area; (b) luminous exitance of the PiG film as a function of pumping laser power density.

Luminous exitance (luminous flux per emitting area, lm/mm^2) is an important parameter to evaluate a phosphor for laser lighting. However, it ~~has seldom~~ is rarely ~~been~~ studied because of the difficulty of measuring the actual emitting area of a phosphor-converted white laser lighting device. In our previous study, a custom-built setup was used to measure the emitting area and the relationship between the luminescent spot area and the incident blue laser spot area was numerically investigated.[22] This setup was used to investigate the luminescent spot area of the present PiG film. As shown in Fig. 3(a), the luminescent spot area was always larger than the incident laser spot area. Further, the luminescent spot area increased linearly with the incident laser spot area. The maximum so-called lumen density of the PiG film was calculated to 1970 lm/mm^2 for the incident laser spot size. In previous studies, the optimal lumen densities of the β-SiAlON:Eu-based PiGs were lower than 1000 lm/mm^2, indicating the superiority of the PiG film in this study.[16,17]

However, the lumen density only has limited applicability, as the generated luminescent light has a larger spot size than the blue laser. More importantly, the luminous exitance of the PiG film can thus be calculated based on the luminous spot sizes measured in Fig. 3(a). As shown in Fig. 3(b), the luminous exitance of the PiG film increased almost linearly with increasing incident power density reaching a maximum value as high as 1052 lm/mm^2. This compares favorably with the approximately 300 lm/mm^2, which is currently the maximum available from LEDs.[23]

In summary, we reported an efficient and robust β-SiAlON:Eu PiG film. A facile blade-coating synthetic approach was employed to fabricate the PiG film. With blue excitation, the PiG film showed an IQE of 58% and a luminous efficacy of 109 lm/W. More importantly, the PiG film possessed a saturation threshold of over 26.4 W/mm^2 and a high luminous exitance of 1052 lm/mm^2 was thus attained. The robustness of the PiG film against elevated temperatures was demonstrated and up to 420°C was measured locally on the film with only minor thermal quenching effects. The above results verifies~~d~~ that the β-SiAlON:Eu PiG film is a suitable color converter for use in high-luminance and wide-color-gamut laser displays or projectors.

Declaration of Competing Interest

We declare that we have no conflict of interest.

Acknowledgements

This work was supported by National Natural Science Foundation of China (NSFC 51802083 and 51772076), Danish Energy Technology Development and Demonstration program (EUDP 64017-0588), and Key Research Foundation of Henan Provincial Department of Education (20A430018).

Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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