Journal Pre-proof

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PII: S0955-2219(20)30020-0

DOI: https://doi.org/10.1016/j.jeurceramsoc.2020.01.020

Reference: JECS 12997

To appear in: *Journal of the European Ceramic Society*

Received Date: 22 November 2019

Revised Date: 16 December 2019

Accepted Date: 9 January 2020

Please cite this article as: Wu H, Hao Z, Pan G-Hui, Zhang L, Wu H, Zhang X, Zhang L, Zhang J, Phosphor-SiO_2 composite films suitable for white laser lighting with excellent color rendering, *Journal of the European Ceramic Society* (2020), doi: https://doi.org/10.1016/j.jeurceramsoc.2020.01.020

Phosphor-SiO_2 composite films suitable for white laser lighting with excellent color rendering

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Abstract

Currently, phosphor composite films draw much attention in white laser lighting. In this work, we developed a novel phosphor-SiO_2 film via a mild method, which avoiding the phosphor degradation. Commercial colloidal silica was elected as the source of SiO_2 and acted as an inorganic binder, gap filler and a protective coating layer. Composite films comprised of mixed $Lu_3Al_5O_{12}:Ce^{3+}$ and $CaAlSiN_3:Eu^{2+}$ phosphors exhibit a uniform dense strucure and strong adhesion to the substrate. When excited by blue diodes laser, the optimal film exhibits excellent thermal stability (it maintains 89.1% of the room-temperature intensity at 200 ^{o}C), super resistance against laser irradiation (12.9 W/mm^2), a broad emission spectra with a full width at half maximum of 180 nm and a high luminous efficiency (183 lm/W). The color rendering index of the film was improved to 85. These outstanding properties indicate that the derived films are a promising candidate for white laser lighting.

Key words: Laser lighting; solid-state lighting; luminescence

Introduction

With the rapid development of illumination technology, solid state lighting, such as phosphor-conversion white laser lighting and white lighting emitting diode (LED), has attracted much attention due to their superior features, such as high efficiency, long life time and environmental friendliness [1-5]. Commercial white LEDs generate white light through a combination of blue GaN/InGaN chips with various phosphors dispersed in organic silicone resin (phosphors in silicone, PiS) [6-8]. However, the quantum efficiency (QE) drop of LED chips at high input current density makes it difficult to achieve high-power and high-brightness device using single LED. Alternatively, laser diodes (LD) do not suffer from these problems, the output power and the QE of LD increase linear with the operating current [9]. Thus the lighting scheme involving blue LD is more attractive as an excitation source for high-power white light than blue LEDs. However, the great heat, yielded during high-density laser irradiation on phosphor materials, would burn or carbonize the traditional organic PiS materials due to their low thermal conductivity and poor thermal stability [9, 10].

To solve this problem, different kinds of inorganic phosphor conversion such as, phosphor ceramics and phosphors in glass (PIG) have been widely investigated [11-22]. Although the phosphor ceramics have high thermal conductity, the processing difficulty (i.e. vacuum sintering or SPS sintering) and excessively high fabricating costs hinder commercial production [23-25]. The PIG could be fabricated through a relatively simple process, but there are two serious problems for PIG: low endurable laser power density (usually lower than 1 W/mm^2) and poor thermal conductivity of the glass matrix [26-28]. In order to solve these problems, an interesting phosphor-substrate configuration was designed, in which a phosphor film was coated on the substrate with high conductivity [29-33]. Park et.al presented a YAG-PIG film bonded on an aluminum substrate and the composite gave an output of 430 lm under a

4W blue LD [34]. Zheng et. al created a YAG-PiG film on a single crystal sapphire substrate and it exhibit a luminous flux of 1839 lm [29]. However, all these phosphor films were PIG films which were prepared by co-sintering of a mixture of phosphors and mother glass powders at a high melting temperature (600-1000 ^oC). Significant phosphor degradation caused by the sintering process is a serious drawback of this method [10, 35]. It can be considered that the interface reaction between commercial phosphor crystals and melting glass facilitated by Na^+ or K^+ as solvent aid in melting glass, cause the lattice distortions of phosphors and reduce their luminescence activity [36]. Xu et. al prepared a YAG-SiO_2 films without melting process, which exhibit excellent thermal stability and high quantum efficiency. However, their photoluminescence performances in laser lighting have not been investigated systematically [37].

In this study, we developed a novel phosphor-SiO_2 composite film coated on the sapphire substrate without melting process. Sapphire is selected as the substrate because of its high thermal conductivity (~30 W m^{-1} K^{-1}) [30, 38]. Commercial colloidal silica was elected as the source of SiO_2 and acted as an inorganic binder, gap filler and a protective coating layer. The composite films based on different phosphors (YAG or $CaAlSiN_3:Eu^{2+}/Lu_3Al_5O_{12}:Ce^{3+}$) were prepared by a facile blade coating procedure, followed by investigation on their microstructure and thermal stability. Their photoluminescence performances (such as luminous flux, luminous efficiency, CRI, correlated color temperature and quantum efficiency) were also investigated in detail under blue LD excitation.

Experimental

Commercial YAG or $CaAlSiN_3:Eu^{2+}/Lu_3Al_5O_{12}:Ce^{3+}$ (CASN/LuAG) phosphors (Yantai Shield Advanced Materials Co. Ltd, China) were elected as phosphor materials. The red CASN and green LuAG phosphors were mixed together and their weigh ratios (abbreviated as R/G) were chosen to be 1/18, 1/15 and 1/12. Commercially available colloidal silica (35 wt%, Dalian Snow Chemical Co. Ltd, China) was used as inorganic binder and Polyvinyl Pyrrolidone (PVP, Sinopharm

Chemical Reagent Co. Ltd, China) was chosen as organic binder. Firstly, viscous slurries with different content of phosphors, PVP and colloidal silica were prepared by fully stirred and mixed in a beaker for 4h. For an appropriate viscosity and high adhesion force, the weight ratio of phosphors, PVP and colloidal silica was optimized to be 3:0.02:1. Then the resultant slurries were coated on the sapphire substrate by blade coating. The samples were dried at room temperature for 12 h. Finally, the dried samples were sintered at 500 ^{o}C for 30 min in a muffle furnace to burn out the organic. The schematic illustration of the preparation process is shown in Fig. 1.

Characterization

The microstructure of the samples was observed by a scanning electron microscopy (SEM, Model S-4800, Hitachi, Japan). The density was measured by the Archimedes method, using deionized water as the immersion medium. Density was calculated using the formula: $\text{Density} = \frac{m_2 - m_1}{m_2 - m_1 - (m_4 - m_3)} \times \rho$ where m_1, m_2 is the weight of the sapphire substrate without and with film out of water, respectively and m_3, m_4 is the corresponding weight submerged in water, ρ is the density of distilled water at room temperature. The adhesion of the films was assessed by a tape test following a cross-cut, according to the Chinese national stand (GB-9286-1998), which corresponds to the International Organization for Standardization (ISO) 2409, using the Scotch 610 transparent tape (width: 19 mm, adhesion: 10 N/25 mm). The temperature-dependent emission spectra were carried out by using a home-made measurement system, which consists of a 452 nm blue LD, a fluorescence microscopy (BX53M, Olympus, Japan), a cooling/heating stage (THMS-600, Linkam, UK) and a charge-coupled device (CCD) spectrometer (QEPro, Ocean Optics, China). The samples were heated from 25 to 300 °C with a step size of 25 °C at a heating rate of 25 °C/min and maintained at each temperature for 1 min. The electroluminescence (EL) properties of the samples under laser irradiation were measured by an integrated test system (EVERFINE, China) including photoelectric characteristic testing system, a rotating integral ball, a 452 nm LD and high precision fast spectral radiometer (HAAS-2000). The optical power of the blue LD, as determined by the input current, was measured with a laser power meter (PM100D, THORLABS, USA). The laser spot was captured by a beam analyzer (BC106N-VIS, THORLABS, USA) and the beam diameter was determined according to the International Organization for Standardization (ISO) 11146. The laser spot size was calculated to be 0.26 mm^2. The surface temperature of the samples was measured by an infrared thermal imaging system (ETS320, FLIR, USA). The internal/external quantum efficiency (IQE/EQE) was measured with an absolute photoluminescence quantum yield measurement system (Quantaurus-QY Plus C13534-12, Hamamatsu Photonics, Japan). All the measurements were conducted at room temperature unless specially mentioned.

Results and discussion

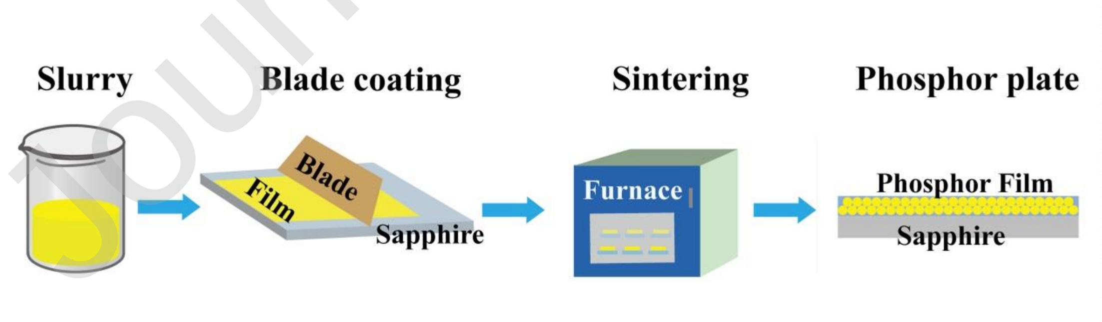

Fig. 1 Schematic illustration of the preparation process for the phosphor-SiO_2 composite films.

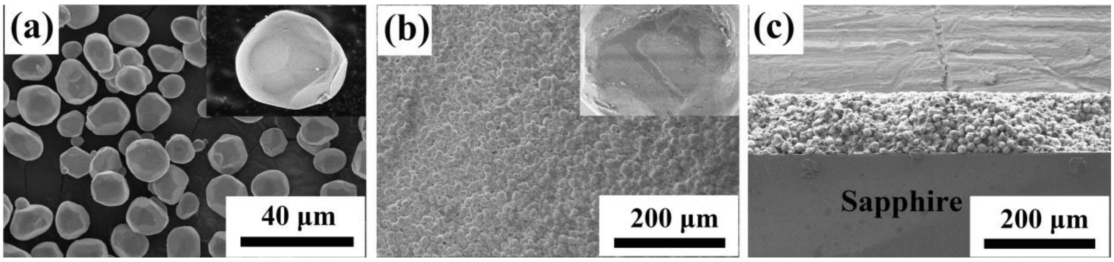

Fig. 2 a) Morphology of the YAG phosphors，inset is the high magnification image of YAG particle; b) top view SEM image, c) cross-sectional view SEM image of YAG based film.

Fig. 2a exhibits the SEM imagine of YAG phosphors. These particles are characteristic of spheroidal morphology with smooth surface and size in range of 8~17 μm. Following the blade coating of such YAG phosphors on a sapphire substrate, dense YAG-SiO_2 composite films were prepared with the assistance of colloidal silica. Fig.2b shows the top view image of the obtained film. Obviously, the film exhibits dense structure and uniform thickness. Phosphor particles in the film accumulate together compactly and only a few small pores can be observed. Based on the densities of YAG (4.56g/cm^3) and SiO_2 (2.32g/cm^3), the theoretical density of the YAG-SiO_2 composite film is estimated to be 4.14 g/cm^3. The density of the film was measured to be 3.99 g/cm^3 and its relative density reaches 96.4%. In contrast to the pristine YAG phosphors, a layer of nano-sized particles (20~60 nm) of SiO_2 was coated on the surface of the phosphors embedded in the film (the inset in Fig.1c). The IQE of the YAG based film and YAG phosphors are 94% and 95%, respectively. The YAG phosphors embedded in the film were perfectly preserved and the oxidation or erosion of phosphors caused by the sintering process did not occur due to the thin SiO_2 protective layer [35]. The cross section image in Fig. 2c demonstrates that the film is bonded to the substrate tightly, which could be ascribed to the adhesion function of the colloidal SiO_2. The adhesion of the film was assessed by a tape pull-off test following a cross-cut. After the test, the interfacial adhesion of the composite film to the substrate seems strong and only a small area of the film (＜5%) peels off from the substrate (Fig.

S1). The structure of colloidal silica (Si-OH) provides strong bonds between phosphors and the substrate [39, 40]. The surface of the film is smooth enough even if without polishing and its thickness is ~135 μm. The gaps of the phosphor particles are also filled up by SiO_2 gel. The colloidal SiO_2 serves as an excellent inorganic binder, gap filler and protective coating layer. High phosphor content and contacted cohesive force between phosphors and the substrate make the composite film could endure high-density laser irradiation without damage. The film did not fall off from the substrate, even under 14.3 W/mm^2 blue LD excitation.

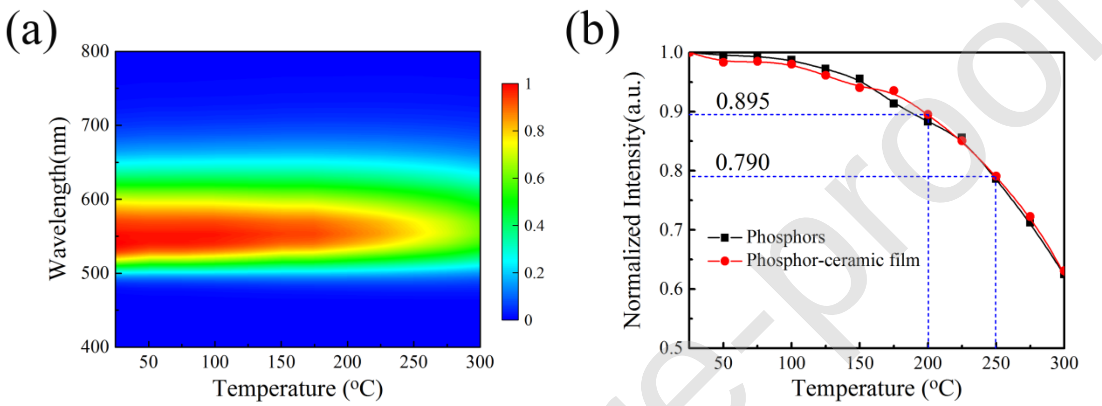

Fig. 3 a) Temperature dependence of the PL spectra for YAG-SiO_2 film under 452 nm blue LD excitation; b) integrated and normalized PL intensity of YAG phosphors and film as a function of temperature.

For laser lighting, thermal stability of the composite film is of great importance. Temperature dependence of the photoluminescence (PL) spectra for YAG-SiO_2 film excited by 0.08 W blue LD is shown in Fig. 3. Overall, as the temperature increases from 25 to 300 °C, PL intensity of the sample shows a continuous drop. The thermal quenching arises from the non-radiative relaxation of excited electrons via the thermally activated trapping of the electrons or the interaction between the electrons and phonons [41, 42]. Simultaneously, we also observe that the emission band slightly red-shifted with the increase of the operating temperature. It can be seen that the thermal stability of the composite film is comparable with that of YAG phosphors. The integrated PL intensity at 200 °C drops to 89.5% of the initial intensity at room temperature. The film maintains 79% of the room-temperature intensity even at 250 °C, which illustrates the excellent thermal stability for laser lighting.

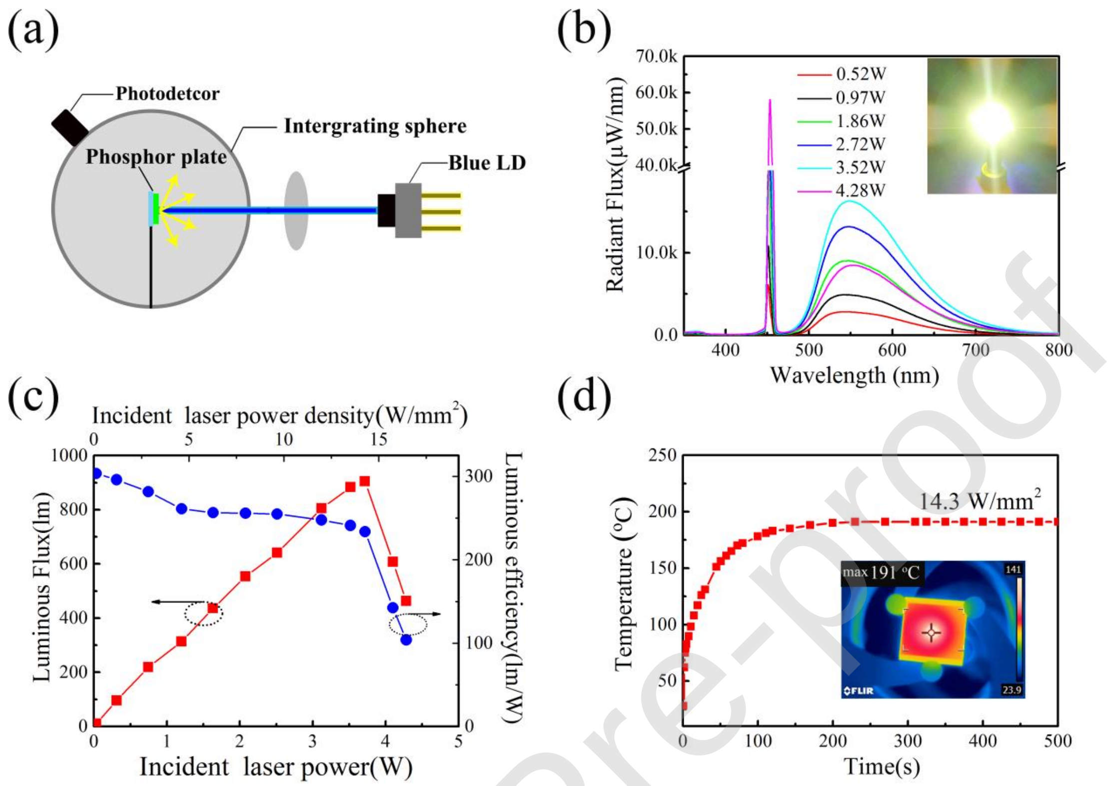

Fig. 4 a) Schematic of laser-driven white light and the photoelectric performance testing device for the ceramics film driven by blue LD; b) EL spectra of the YAG-SiO_2 composite film with different incident laser and the insert is the corresponding lighting photographs under 0.52 W laser excitation; c) luminous flux and luminous efficacy of the film as a function of the incident blue LD; d) surface temperature of the sample without heat sink under 3.52 W laser excitation and the insert is the corresponding thermal infrared image.

Furthermore, the feasibility of the YAG-SiO_2 composite film serving as a LD-pumped phosphor conversion was also studied. Thus, a configuration for white laser lighting was constructed by coupling the composite film with a 452 nm blue LD in a reflection mode [43]. Its optical performance was investigated using the configuration shown in Fig. 4a. The EL spectra exhibit two emission peaks: the sharp peak at 452 nm corresponds to the emission of the laser and the broad emission band between 480 and 720 nm corresponds to the emission of the YAG-SiO_2 composite film ((Fig. 4b). With increasing the incident LD power from 0.52 to 3.52

W, the emission intensity of the sample increases monotonously. But when the incident power increases to 4.17W, luminescence saturation is observed and the emission intensity of the film drops sharply. The maximum luminous flux reaches to 905 lm at an incident power of 3.52 W and the corresponding power density of blue LD is 14.3 W/mm^2 (Fig. 4c). As to the luminous efficiency, it slightly decreases as the incident laser power increases, indicating the thermal quenching due to the increased temperature of the laser spot upon high-power laser excitation. The local heating would quench the luminescence by transforming the excitation light energies into phonons or thermal ionization, which in turn further increases the temperature [29]. When the incident power is higher than a threshold value, the heat cannot be efficiently dissipated but accumulates and increases the temperature of the sample continuously [29, 44]. Meanwhile the luminous saturation would occur and luminous efficiency would drop. It is believed that the thermal quenching dominates the luminous saturation and efficiency drop. As shown in Fig. 4d, the surface temperature change of the laser spot on the sample without heat sink was measured by an infrared thermal image. The temperature incerases quickly and reaches an equilibrium within 150 s. As the laser power increases to 3.52 W, the equilibrium temperature of the laser spot amounts to 191^{o}C. Moreover, the red-shift of the emission band with the increase of incident laser power also indicates the rise of the surface local temperature, which is similar to the red-shift observed in Fig. 3a (Fig. S2).

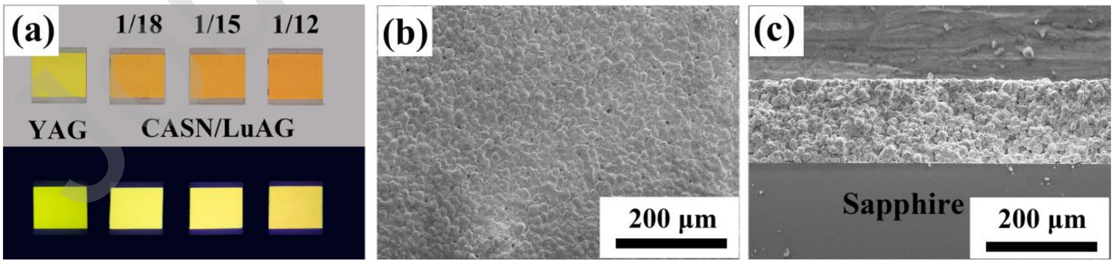

Fig. 5 a) Photograph of the phosphor-SiO_2 with different content phosphors under day light and blue light; b) top view SEM image, c) cross-sectional view SEM image of the film with R/G=1/15.

Color quality is an important consideration for phosphor-converted white lighting systems as general illumination sources [45, 46]. Although the YAG based film exhibits high thermal stability and high efficiency, the CRI of this phosphor converted white light was commonly low (~60) [29, 30]. Recent demand market for laser lighting applications such as automotive headlamps or outdoor lighting also requires a higher CRI ＞ 80. To improve the color rendering, green LuAG and red CASN phosphors were mixed together to enrich the green and red light component in the spectra. The micrograph of the phosphors is shown in Fig. S3. The LuAG phosphors are spheroid-shaped particles (14 μm in size), while the CASN phosphors are cuboid-shaped particles (16 μm in length). Photographs of the composite films with different content phosphors are shown in Fig. 5a. The appearance of the YAG based film shows bright yellow colour under daylight, while the CASN/LuAG based films turn from orange to deep orange as the CASN content increased. Under 450 nm blue light, bright yellow and orange luminescence is generated by the films.Both phosphors have a strong absorption at 452 nm, enabling them to be excitated the 452 nm incident blue LD. When excited by blue LD, the LuAG phosphors yield a broad emission band peaking at 533 nm, whereas the CASN phosphors exhibit an emissions peak at 634 nm (Fig. S4). The micrographs of the composite film (R/G=1/15) with mixed phosphors are exhibited in Fig. 5b and Fig. 5c. The obtained film exhibits dense structure, smooth surface and uniform thickness and its thickness is ~152 μm. The film was bonded to the sapphire substrate tightly and the maximum laser power density it could endure is 12.3 W/mm^2.

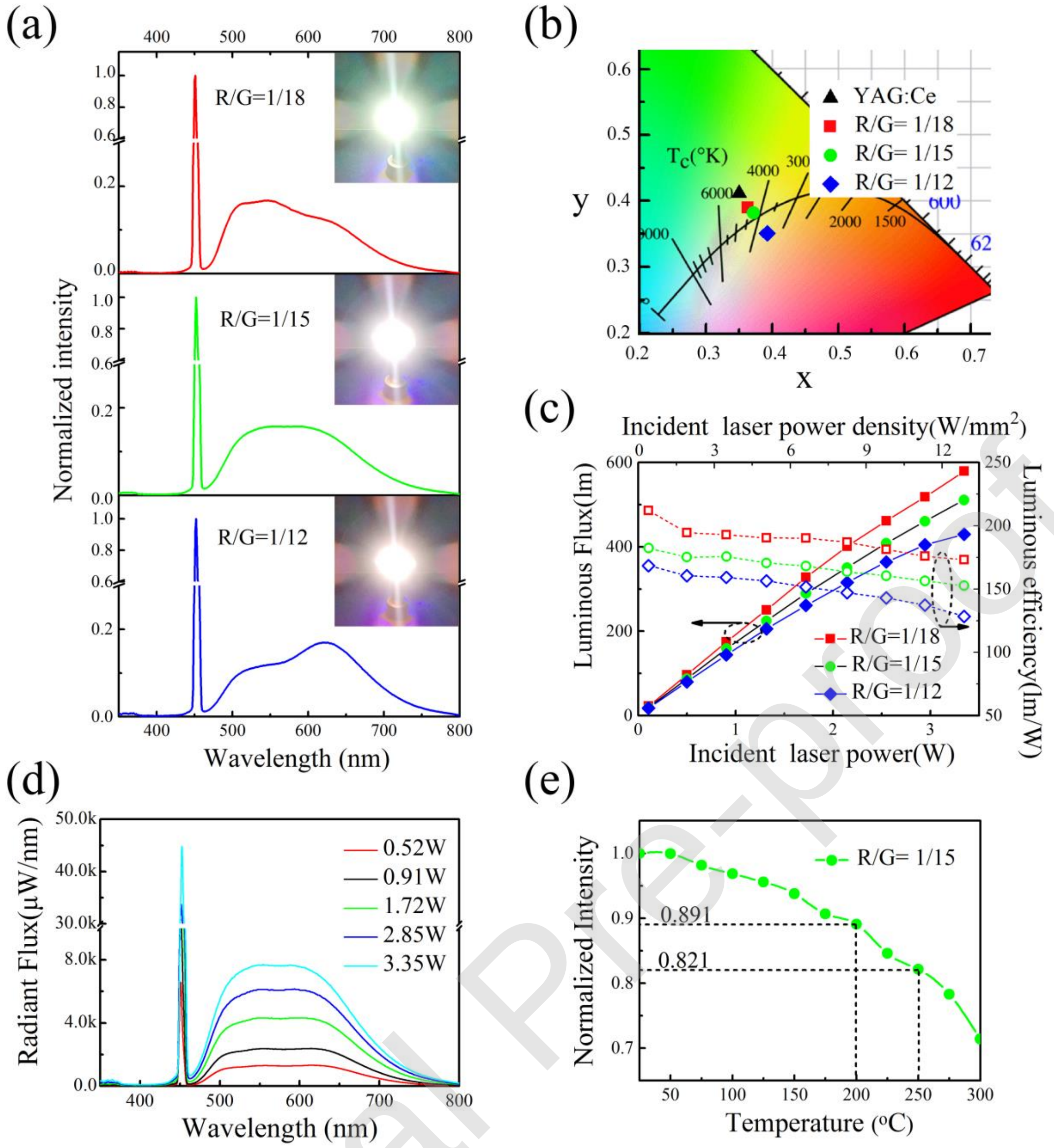

Fig. 6 Optical performance of the phosphor-SiO_2 composite resZszsazffcvxkjninjmmkihjhg nghvgyjtrfilms with different content of LuAG and CASN: a) normalized EL spectra driven by 0.52 W blue laser and the insert is the corresponding lighting photographs; b) the corrsponding CIE color coordinate; c) luminous flux and luminous efficiency with different incident laser; e) integrated and normalized PL intensity of the film with R/G=1/15 as a function of temperature.

By controlling the weight ratio of the red and green phosphors, composite films with different R/G were fabricated and their optical performances were checked. Overall the emission spectra of the films based on CASN/LuAG phosphors were greatly broadened covering the whole green, yellow and red region from 460 to 750 nm (Fig. 6a). The correlated full width at half maxium (FWHM) is ~180 nm, while that based on YAG phosphors is just ~114 nm. Two distinguishable broad emission bands are observed, which correspond to the green emission at ~ 530 nm and the red emission at ~ 630 nm. The varying tendency of the emission intensity between the green and red bands is easily discernable by adjusting the R/G value. As the R/G increases (CASN content increases), the red emission component gradually intensifies while the green emission comonent decreases owing to the more effective absorbtion of blue light by the red phosphors.

Fig. 6b depicts the corresponding chromaticity coordinates of ceramic film under 0.52 W blue LD. As the CIE color coordinates shift from yellow region to red region, the appearance of the white laser light moved from cool to warm white (the insert in Fig. 6a). The movement can be well identified from the correlated color temperature (CCT) value, as summarized in Table 1. Moreever, the CRI is also significantly improved from 61 to 92. In addition, the luminous flux and luminous efficiency of the samples with various R/G were investigated and shown in Fig. 6c. With the increase of incident power, the lumious flux of the three samples increase nonlinearly. The lumious efficiency, howerver, decrease slowly with increasing incident power, which could be induced by thermal quenching. For samples with the R/G of 1/18, 1/15, 1/12, the maximum lumious flux can reach to 579, 511, 404 lm and the corrsesponding lumious efficiency is 173, 152, 137 lm/W, respectively. The lumious efficiency is much lower than the YAG-based film due to the serious reabsorption of the green emission by the red phosphors [47]. To supress the reabsorption effect, the multilayered structure would be developed in the future work.

As shown in Tab. 1, the sample based on YAG phosphors exhibits the highest luminous efficiency of 234 lm/W, but the lowest CRI of 61. When CASN/ LuAG phosphors were used，the CRI increases up to 80. With the increase of CASN content, the IQE/EQE decreases and this is ascribed to the more serious reabsorption effect. Balancing the luminous efficiency, QE and CRI, the sample with R/G =1/15 exhibits the optimal performance. With increasing the incident LD power from 0.51 to 3.35 W, the emission intensity of the sample (R/G =1/15) increases monotonously (Fig. 6d). The sample yields a uniform white light with of a luminous flux of 511 lm, a luminous efficiency of 152 lm/W, a CCT of 4277K and an IQE of 81% under 3.35 W blue LD excitation (with the power density of 12.9 W/mm^2). Its thermal stability was also investigated and the film maintains 89.1 % of the room-temperature intensity even at 200 ^{o}C (Fig. 6e). These results indicate that the phosphor-SiO_2 composite films could be used for high-power white laser lighting with high CRI.

Table. 1 Optical performance of the film with different content of phosphors

Sample	YAG	CASN/LuAG (R/G)		
		1/18	1/15	1/12
IQE	94%	89%	89%	81%
EQE	52%	44%	37%	36%
FWHM(nm)	114	177	183	185
Laser power(W)	3.72	3.35	3.35	3.35
Power density(W/mm^2)	14.3	12.9	12.9	12.9
Luminous Flux(lm)	905	579	511	404
Luminous efficacy (lm/W)	234	173	152	137
CCT(K)	4986	4550	4277	3439
CRI	61	80	85	92
CIE-x	0.3502	0.3636	0.3718	0.3927
CIE-y	0.4113	0.3896	0.3820	0.3509

Conclusion

In summary, a facile procedure was developed to prepare a robust and efficient phosphor conversion by coating the phosphor-SiO_2 composite films on the sapphire substrate for laser-driven white lighting. The YAG based film exhibits high quantum efficiency (94%), high thermal stability (it maintains 89.5% of the room-temperature intensity at 200 ^{o}C) and super resistance against blue laser irradiation (14.3 W/mm2). When drove by 3.72 W blue LD, the film produces a white light with a luminous flux of 905 lm, a luminous efficiency of and a CRI of 61. To improve the color rendering, composite films comprised of mixed green LuAG and red CASN phosphors were prepared and tested. The optimized sample (R/G=1/15) produces the white light with a luminous flux of 511 lm, luminous efficiency of 152 lm/W, a FWHM of 184 nm and CRI of 85. Through the combination of LuAG and CASN phosphors, the CRI of the composite films was largely improved for white laser lighting. These results indicate that the developed phosphor-SiO_2 composite films offer great potential for use in high-power laser-driven white lighting with excellent CRI.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledge

This work was supported by National Key R&D Program of China (Grant No. 2017YFB0403100, 2017YFB0403104), National Natural Science Foundation of China (Grant No. 51772286,11874055, 11974346 and 11604330).

Reference

[1] J.H. Oh, S.J. Yang, Y.R. Do, Healthy, natural, efficient and tunable lighting: four-package white LEDs for optimizing the circadian effect, color quality and vision performance, Light-Science & Applications 3 (2014).

[2] Z.G. Xia, Q.L. Liu, Progress in discovery and structural design of color conversion phosphors for LEDs, Prog Mater Sci 84 (2016) 59-117.

[3] M. Zhao, H.X. Liao, M.S. Molokeev, Y.Y. Zhou, Q.Y. Zhang, Q.L. Liu, Z.G. Xia, Emerging ultra-narrow-band cyan-emitting phosphor for white LEDs with enhanced color rendition, Light-Science & Applications 8 (2019).

[4] X.F. Li, J.D. Budai, F. Liu, J.Y. Howe, J.H. Zhang, X.J. Wang, Z.J. Gu, C.J. Sun, R.S. Meltzer, Z.W. Pan, New yellow $Ba_{0.93}Eu_{0.07}Al_2O_4$ phosphor for warm-white light-emitting diodes through single-emitting-center conversion, Light-Science & Applications 2 (2013).

[5] S.H. Lim, Y.H. Ko, C. Rodriguez, S.H. Gong, Y.H. Cho, Electrically driven, phosphor-free, white light-emitting diodes using gallium nitride-based double concentric truncated pyramid structures, Light-Science & Applications 5 (2016)

[6] L. Wang, R.J. Xie, Y.Q. Li, X.J. Wang, C.G. Ma, D. Luo, T. Takeda, Y.T. Tsai, R.S. Liu, N. Hirosaki, $Ca_{1-x}Li_xAl_{1-x}Si_{1+x}N_3:Eu^{2+}$ solid solutions as broadband, color-tunable and thermally robust red phosphors for superior color rendition white light-emitting diodes, Light-Science & Applications 5 (2016).

[7] P.P. Dai, C. Li, X.T. Zhang, J. Xu, X. Chen, X.L. Wang, Y. Jia, X.J. Wang, Y.C. Liu, A single Eu^{2+}-activated high-color-rendering oxychloride white-light phosphor for white-light-emitting diodes, Light-Science & Applications 5 (2016).

[8] Y.H. Kim, P. Arunkumar, B.Y. Kim, S. Unithrattil, E. Kim, S.-H. Moon, J.Y. Hyun, K.H. Kim, D. Lee, J.-S. Lee, W.B. Im, A zero-thermal-quenching phosphor, Nature Materials 16 (2017) 543.

[9] J.J. Wierer, J.Y. Tsao, D.S. Sizov, Comparison between blue lasers and light-emitting diodes for future solid-state lighting, Laser & Photonics Reviews 7(6) (2013) 963-993.

[10] H. Lin, T. Hu, Y. Cheng, M. Chen, Y. Wang, Glass Ceramic Phosphors: Towards Long-Lifetime High-Power White Light-Emitting-Diode Applications-A Review, Laser & Photonics Reviews (2018) 1700344.

[11] A. Ikesue, Y.L. Aung, Ceramic laser materials, Nat Photon 2(12) (2008) 721-727.

[12] Q. Zhang, R. Zheng, J. Ding, W. Wei, Excellent luminous efficiency and high thermal stability of glass-in-LuAG ceramic for laser-diode-pumped green-emitting phosphor, Opt Lett 43(15) (2018) 3566-3569.

[13] D. Chen, W. Xiang, X. Liang, J. Zhong, H. Yu, M. Ding, H. Lu, Z. Ji, Advances in transparent glass–ceramic phosphors for white light-emitting diodes—A review, Journal of the European Ceramic Society 35(3) (2015) 859-869.

[14] R. Zhang, H. Lin, Y. Yu, D. Chen, J. Xu, Y. Wang, A new-generation color converter for high-power white LED: transparent Ce^{3+}:YAG phosphor-in-glass, Laser & Photonics Reviews 8(1) (2014) 158-164.

[15] S.H. Ahn, Y.H. Nam, K. Han, W.B. Im, K.Y. Cho, W.J. Chung, Phosphor-in-glass thick film formation with low sintering temperature phosphosilicate glass for robust white LED, Journal of the American Ceramic Society 100(4) (2017) 1280-1284.

[16] H. Chen, H. Lin, J. Xu, B. Wang, Z. Lin, J. Zhou, Y. Wang, Chromaticity-tunable phosphor-in-glass for long-lifetime high-power warm w-LEDs, Journal of Materials Chemistry C 3(31) (2015) 8080-8089.
[17] W. Nian, L. Tiecheng, L. Feng, Z. Wei, M. Benyuan, L. Zhongwen, Q. Jianqi, Transparent Ce:$Y_3Al_5O_{12}$ ceramic phosphors for white light-emitting diodes, Applied Physics Letters 101(6) (2012) 061902 (4 pp.)-061902 (4 pp.).
[18] S. Li, Q. Zhu, L. Wang, D. Tang, Y. Cho, X. Liu, N. Hirosaki, T. Nishimura, T. Sekiguchi, Z. Huang, R.-J. Xie, $CaAlSiN_3$:Eu^{2+} translucent ceramic: a promising robust and efficient red color converter for solid state laser displays and lighting, Journal of Materials Chemistry C 4(35) (2016) 8197-8205.
[19] Y. Liu, S. Liu, P. Sun, Y. Du, S. Lin, R.J. Xie, R. Dong, J. Jiang, H. Jiang, Transparent Ceramics Enabling High Luminous Flux and Efficacy for the Next-Generation High-Power LED Light, ACS Appl Mater Interfaces (2019).
[20] J. Xu, J. Wang, Y. Gong, X. Ruan, Z. Liu, B. Hu, B. Liu, H. Li, X. Wang, B. Du, Investigation of an LuAG:Ce translucent ceramic synthesized via spark plasma sintering: Towards a facile synthetic route, robust thermal performance, and high-power solid state laser lighting, Journal of the European Ceramic Society 38(1) (2018) 343-347.
[21] J. Xu, B. Hu, C. Xu, J. Wang, B. Liu, H. Li, X. Wang, B. Du, Y. Gong, Carbon-free synthesis and luminescence saturation in a thick YAG:Ce film for laser-driven white lighting, Journal of the European Ceramic Society 39(2-3) (2019) 631-634.
[22] C. Cozzan, G. Lheureux, N. O'Dea, E.E. Levin, J. Graser, T.D. Sparks, S. Nakamura, S.P. DenBaars, C. Weisbuch, R. Seshadri, Stable, Heat-Conducting Phosphor Composites for High-Power Laser Lighting, Acs Applied Materials & Interfaces 10(6) (2018) 5673-5681.
[23] Q. Du, S. Feng, H. Qin, H. Hua, H. Ding, L. Jia, Z. Zhang, J. Jiang, H. Jiang, Massive red-shifting of Ce^{3+} emission by Mg^{2+} and Si^{4+} doping of YAG:Ce transparent ceramic phosphors, Journal of Materials Chemistry C 6(45) (2018) 12200-12205.
[24] S. Li, Q. Zhu, D. Tang, X. Liu, G. Ouyang, L. Cao, N. Hirosaki, T. Nishimura, Z. Huang, R.-J. Xie, Al_2O_3-YAG:Ce composite phosphor ceramic: a thermally robust and efficient color converter for solid state laser lighting, Journal of Materials Chemistry C 4(37) (2016) 8648-8654.
[25] X. Liu, X. Qian, Z. Hu, X. Chen, Y. Shi, J. Zou, J. Li, Al_2O_3-Ce:GdYAG composite ceramic phosphors for high-power white light-emitting-diode applications, Journal of the European Ceramic Society 39(6) (2019) 2149-2154.
[26] K. Han, S.H. Lee, Y.G. Choi, W.B. Im, W.J. Chung, Improved color rendering index and thermal stability of white LEDs with phosphor-in-glass using the SiO_2-B_2O_3-ZnO-Na_2O glass system, Journal of Non-Crystalline Solids 445–446 (2016) 77-80.
[27] Q.Q. Zhu, X. Xu, L. Wang, Z.F. Tian, Y.Z. Xu, N. Hirosaki, R.J. Xie, A robust red-emitting phosphor-in-glass (PiG) for use in white lighting sources pumped by blue laser diodes, Journal of Alloys and Compounds 702 (2017) 193-198.
[28] X. Zhang, J. Yu, J. Wang, B. Lei, Y. Liu, Y. Cho, R.-J. Xie, H.-W. Zhang, Y. Li, Z. Tian, Y. Li, Q. Su, All-Inorganic Light Convertor Based on Phosphor-in-Glass Engineering for Next-Generation Modular High-Brightness White LEDs/LDs, ACS

Photonics 4(4) (2017) 986-995.

[29] P. Zheng, S. Li, L. Wang, T.L. Zhou, S. You, T. Takeda, N. Hirosaki, R.J. Xie, Unique Color Converter Architecture Enabling Phosphor-in-Glass (PiG) Films Suitable for High-Power and High-Luminance Laser-Driven White Lighting, ACS Appl Mater Interfaces 10(17) (2018) 14930-14940.

[30] R. Wei, L. Wang, P. Zheng, H.T. Zeng, G.M. Pan, H. Zhang, P. Liang, T.L. Zhou, R.J. Xie, On the luminance saturation of phosphor-in-glass (PiG) films for blue-laser driven white lighting: Effects of the phosphor content and the film thickness, Journal of the European Ceramic Society 39(5) (2019) 1909-1917.

[31] Y. Peng, X. Guo, R. Li, H. Cheng, M. Chen, Thermally stable WLEDs with excellent luminous properties by screen-printing a patterned phosphor glass layer on a microstructured glass plate, Appl. Opt. 56(12) (2017) 3270-3276.

[32] E. Kim, S. Unithrattil, I.S. Sohn, S.J. Kim, W.J. Chung, W.B. Im, Facile one-step fabrication of 2-layered and 4-quadrant type phosphor-in-glass plates for white LEDs: an insight into angle dependent luminescence, Opt. Mater. Express 6(3) (2016) 804-814.

[33] R. Xiang, X. Liang, P. Li, X. Di, W. Xiang, A thermally stable warm WLED obtained by screen-printing a red phosphor layer on the LuAG:Ce^{3+} PiG substrate, Chemical Engineering Journal 306 (2016) 858-865.

[34] J. Park, J. Kim, H. Kwon, Phosphor-Aluminum Composite for Energy Recycling with High-Power White Lighting, Advanced Optical Materials 5(19) (2017) 1700347.

[35] J. Xu, D.A. Hassan, R.-J. Zeng, D.-L. Peng, $Lu_3Al_5O_{12}$:Ce@SiO_2 phosphor-in-glass: Its facile synthesis, reduced thermal/chemical degradation and application in high-power white LEDs, Journal of the European Ceramic Society 36(8) (2016) 2017-2025.

[36] L.-Y. Chen, W.-C. Cheng, C.-C. Tsai, Y.-C. Huang, Y.-S. Lin, W.-H. Cheng, High-performance glass phosphor for white-light-emitting diodes via reduction of Si-Ce^{3+}:YAG inter-diffusion, Opt. Mater. Express 4(1) (2014) 121-128.

[37] J. Xu, B. Liu, Z. Liu, Y. Gong, B. Hu, J. Wang, H. Li, X. Wang, B. Du, Design of laser-driven SiO_2-YAG:Ce composite thick film: Facile synthesis, robust thermal performance, and application in solid-state laser lighting, Optical Materials 75 (2018) 508-512.

[38] R. Apetz, M.P.B. Bruggen, Transparent Alumina: A Light-Scattering Model, Journal of the American Ceramic Society 86(3) (2003) 480-486.

[39] D. Kong, H. Yang, S. Wei, D. Li, J. Wang, Gel-casting without de-airing process using silica sol as a binder, Ceramics International 33(2) (2007) 133-139.

[40] L. Chu, M.W. Daniels, L.F. Francis, Use of (Glycidoxypropyl)trimethoxysilane as a Binder in Colloidal Silica Coatings, Chemistry of Materials 9(11) (1997) 2577-2582.

[41] T. Miyakawa, D.L. Dexter, Phonon Sidebands, Multiphonon Relaxation of Excited States, and Phonon-Assisted Energy Transfer between Ions in Solids, Physical Review B 1(7) (1970) 2961-2969.

[42] T.W. Kang, K.W. Park, J.H. Ryu, S.G. Lim, Y.M. Yu, J.S. Kim, Strong thermal stability of $Lu_3Al_5O_{12}$:C^{e3+} single crystal phosphor for laser lighting, Journal of Luminescence 191 (2017) 35-39.

[43] A. Lenef, J. Kelso, M. Tchoul, O. Mehl, J. Sorg, Y. Zheng, Laser-activated remote phosphor conversion with ceramic phosphors, in: M.H. Kane, J. Jiao, N. Dietz, J.J. Huang

(Eds.), Thirteenth International Conference on Solid State Lighting2014.
[44] S. Li, L. Wang, N. Hirosaki, R.-J. Xie, Color Conversion Materials for High-Brightness Laser-Driven Solid-State Lighting, Laser & Photonics Reviews 12(12) (2018) 1800173.
[45] Y. Wei, G.C. Xing, K. Liu, G.G. Li, P.P. Dang, S.S. Liang, M. Liu, Z.Y. Cheng, D.Y. Jin, J. Lin, New strategy for designing orangish-redemitting phosphor via oxygen-vacancy-induced electronic localization, Light-Science & Applications 8 (2019).
[46] T. Senden, R.J.A. van Dijk-Moes, A. Meijerink, Quenching of the red Mn^{4+} luminescence in Mn^{4+}-doped fluoride LED phosphors, Light-Science & Applications 7 (2018).
[47] B.H. Kim, J. Hwang, Y.J. Lee, J.H. Kim, D.W. Jeon, M.J. Lee, Comparison between mixed and spatially separated remote phosphor fabricated via a screen-printing process, Optical Engineering 55(8) (2016).