Science Bulletin 64 (2019) 1649–1651

Contents lists available at ScienceDirect

Science Bulletin

journal homepage: www.elsevier.com/locate/scib

Research Highlight

Cyan phosphors for full-visible-spectrum lighting: shining new light on high-CRI white pc-LEDs

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Nowadays, solid-state lighting sources based on white light-emitting diodes (LEDs) are extensively replacing the traditional illumination devices (incandescent and fluorescent lamps) due to their unique and appealing properties, such as high efficiency, high brightness, long working lifetime, small size, fast response time and eco-friendliness [1–4]. Because of this, white LEDs have permeated deeply into many important sectors, including indoor and outdoor lightings, signals, displays and therapeutic applications. In order to make sure that white LEDs can provide artificial lighting source with good color quality and high vision performance, there are several important figures of merit that are used to characterize white LEDs: color rendering index (CRI), correlated color temperature, luminous efficacy of radiation and luminous efficacy.

The CRI is a tool used to determine a light source's distinctive attributes. It is an international measurement scale that quantifies the ability of a light source to reproduce the colors of various objects faithfully when compared to a familiar basis of reference, either incandescent light or daylight. The scale of CRI varies from 0 to100. The higher the CRI is, the better the color rendering ability is. As a guide, natural light and incandescent light source have a CRI of 100, which can enable to show the true colors of the object. Typically, a light source with a CRI > 70 is acceptable for outdoor lighting, whereas a CRI > 80 is required for indoor lighting. However, light sources with a high CRI > 90 are desirable in color-critical applications such as photography, cinematography, museum and art gallery, jewelry shops, surgery, as well as printing and painting businesses [5,6]. Generally speaking, light sources possessing a wider emission spectrum are more likely to offer higher CRI values. Accordingly, in recent years a new concept called "full-visible-spectrum lighting" has gained considerable attention in the field of white LED lighting technology, which strives to achieve an ultrahigh-CRI light source that mimics natural sunlight. The full-visible-spectrum lighting has several important applications. First, full-visible-spectrum lighting can be employed as light therapy to treat the symptoms of seasonal affective disorder. Second, full-visible-spectrum lighting can be used in indoor plant growth, because it can provide sufficient light that approximates natural sunlight in order to trigger photosynthesis for plants. Third, full-visible-spectrum lighting can help reduce eye fatigue and also provide benefits including better visibility, improved color rendering quality, better health, and greater productivity, thus creating a healthy and productive lifestyle.

At present, the most popular method for fabricating white LEDs is based on phosphor-converted LEDs (pc-LEDs), which utilize the combination of blue-emitting InGaN LED chips (440–470 nm) and YAG:Ce^{3+} yellow-emitting phosphor coating (500–700 nm) to produce white light illumination [7–12]. However, such white LED devices suffer from low CRI values (<75) due to the broken emission spectra in comparison with incandescent light and sunlight: a cyan "gap" (between the primary blue LED and YAG:Ce^{3+} phosphor emission; ~470–510 nm wavelength range) and a lack of red (Fig. 1) (https://www.led-professional.com/resources-1/articles/high-color-rendering-full-visible-spectrum-leds-the-first-wave-of-conventional-white-leds-primarily-focused-on-lumens-per-watt-these-leds-were-by-soraa). Recently, intense research efforts have been made to improve the CRI values of YAG:Ce^{3+}-based white LEDs through carefully employing red phosphors, and a high CRI around 90 has been achieved [13–16]. In sharp contrast, so far, too little attention has been paid to covering the cyan gap by developing cyan-emitting phosphors towards realizing blue-excited white pc-LEDs with ultrahigh-CRI (>95) [17]. In fact, once the problem of cyan gap is solved, full-visible-spectrum lighting could be created. However, there are several exacting requirements for such a cyan phosphor: (1) it has strong absorption in blue spectral range (440–470 nm) and can emit cyan emission around 490 nm in the 470–510 nm wavelength range, along with small Stokes shift; (2) the luminescence efficiency should be high; (3) its emission band should be narrow in order to minimize efficiency loss owing to self-absorption which is frequently occurred in the white pc-LEDs based on multi-component phosphors (the YAG:Ce^{3+} phosphor exhibits intense absorption of blue light in the range of ~420–480 nm). Moreover, narrow emission band may ensure high luminous efficacy because human eyes show very low sensitivity in the blue spectral range; (4) good physicochemical stability; (5) high thermal stability; (6) easy fabrication and low cost; and (7) environmental-friendly. However, it remains challenging to obtain such a perfect cyan-emitting phosphor with

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https://doi.org/10.1016/j.scib.2019.09.008

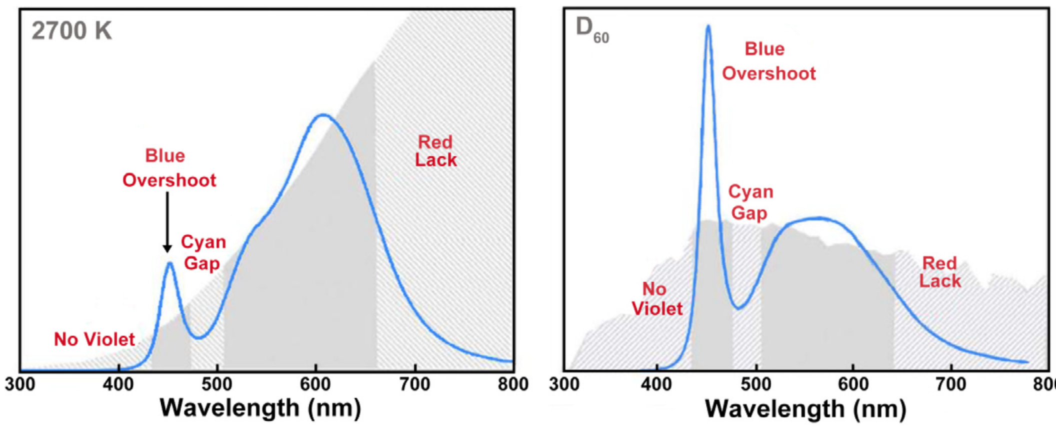

Fig. 1. (Color online) Spectrum comparison of standard blue-based LEDs to incandescence (2700 K) and daylight (6000 K).

high-quality luminescent properties for ultrahigh-CRI white pc-LEDs.

In a recent publication of *Light: Science & Applications*, Xia and co-workers [18] discovered an interesting blue-excitable narrow-band cyan-emitting $Na_{0.5}K_{0.5}Li_3SiO_4:Eu^{2+}$ (abbreviated as: NKLSO: Eu^{2+}) phosphor with emission peak around 486 nm and full width at half-maximum (FWHM) of about 20.7 nm as well as a small Stokes shift of 1069 cm^{-1} (Fig. 2a). By using this cyan phosphor, the authors filled the cyan gap that commonly exists in blue-excited pc-LEDs and thus realized high-CRI white pc-LED device with CRI value up to 95.2.

In their recent work, Xia and co-workers fabricated the NKLSO: Eu^{2+} cyan phosphors through a facile traditional solid-state reaction technique with relatively low calcination temperature (750 °C), which method is suitable for mass-production. Then, the authors examined the photoluminescence properties of NKLSO:Eu^{2+} phosphors, and they surprisingly found that upon excitation with ultraviolet to blue light, the composition-

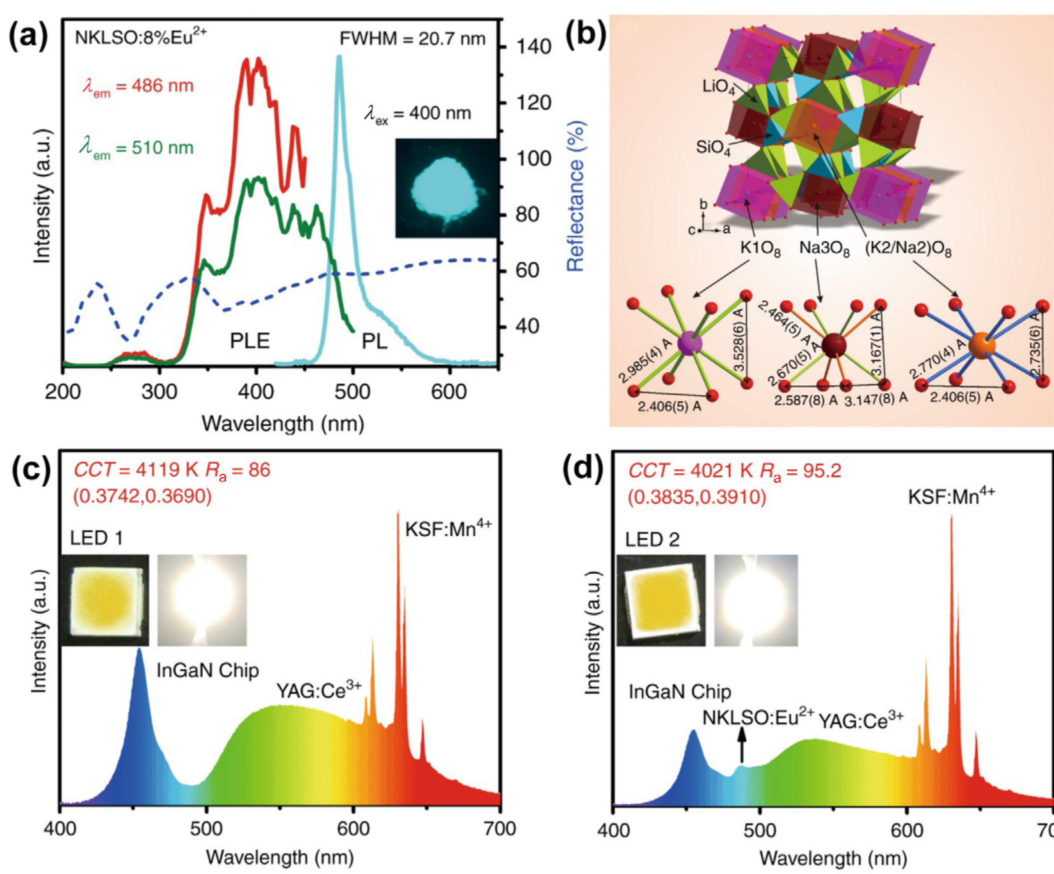

Fig. 2. (Color online) Blue-excitable narrow-band cyan-emitting phosphor NKLSO:Eu^{2+} for full-visible-spectrum white LEDs. (a) Diffuse reflectance, photoluminescence and photoluminescence excitation spectra of NKLSO:8%Eu^{2+} at room temperature. (b) Crystal structure of NKLSO and the coordination spheres of K1, Na3, and K2/Na2. (c, d) Emission spectra of the white LED devices fabricated with the commercial yellow phosphor YAG:Ce^{3+}, the commercial red phosphor KSF:Mn^{4+} and (c) without or (d) with the cyan phosphor NKLSO:8%Eu^{2+} on a blue LED InGaN chip (λ = 455 nm) under 20 mA current. Adapted by permission from Ref. [18], Copyright (2019) Springer Nature.

optimized NKLSO:8%Eu^{2+} phosphors exhibited a bright narrow-band cyan emission (FWHM ≈ 20.7 nm) with a strong peak at 486 nm as well as a weak shoulder peak at 530 nm (Fig. 2a). Moreover, the NKLSO:Eu^{2+} phosphors included a broad excitation band in the 300–500 nm spectral range with a maximum at 400 nm (Fig. 2a). Consequently, the NKLSO:Eu^{2+} phosphors can be used for fabricating white pc-LEDs based on commercial blue-emitting LED chips. The outstanding emission characteristics of NKLSO:Eu^{2+} phosphors are critically dependent on the compound's crystal structure (Fig. 2b). In NKLSO host, the LiO_4 and SiO_4 tetrahedra were connected to each other by corner- and edge-sharing forming a highly condensed three-dimensional framework, and three different cations (K1, K2/Na2, and Na3) provided a highly symmetric dopant site (for Eu^{2+} activators) with an almost ideal cubic coordination (coordination number = 8), resulting in the narrow-band cyan emission. Importantly, the developed NKLSO:Eu^{2+} cyan phosphors also possessed good thermal stability; the integrated emission intensity at 150 °C remained about 93% of that at room temperature.

To further assess the potential application of NKLSO:Eu^{2+} cyan phosphors in high-CRI white LEDs, Xia and co-workers fabricated two blue-excited white pc-LED devices with and without the as-prepared NKLSO:8%Eu^{2+} cyan phosphors and compared their photoelectric properties (Fig. 2c, d). Impressively, the authors found that the addition of NKLSO:8%Eu^{2+} cyan phosphors can compensate the cyan gap and thus effectively enlarged the CRI value from 86 to 95.2, which was beneficial for full-visible-spectrum lighting. Furthermore, the pc-LED device fabricated with NKLSO:8%Eu^{2+} cyan phosphors exhibited superior color stability under different driven currents. Nevertheless, the introduction of NKLSO:8%Eu^{2+} cyan phosphors into white LED resulted in an obvious reduction in luminous efficacy (at 20 mA driven current, 174.98 lm/W for LED device without NKLSO:8%Eu^{2+} cyan phosphors and 119.92 lm/W for LED device with NKLSO:8%Eu^{2+} cyan phosphors), which could be attributed to the following two possible reasons. First, there is a trade-off between luminous efficacy and CRI; thus the CRI value will be boosted inevitably at the expense of luminous efficacy. Second, the as-prepared NKLSO:8%Eu^{2+} cyan phosphors still possessed relatively modest quantum efficiency (internal quantum efficiency = 76% and external quantum efficiency = 30% under 400 nm excitation). Further improvements on the photoluminescence quantum efficiency are required, which may be realized via optimization of the synthesis procedure [19].

In sum, this work by Xia and co-workers provides new insights for rational design of high-performance blue-excited narrow-band cyan-emitting phosphors for high-CRI white LEDs towards full-visible-spectrum lighting. Similar narrow-band cyan photoluminescence might be expected in other Eu^{2+}-activated oxide-based UCr_4C_4-type compounds and also nitride phosphor materials [17]. These findings in this work will promote the development of various novel narrow-band emitting inorganic phosphors for solid-state lighting and displays. Definitely, future research is still needed to search for a desirable efficient and stable cyan-emitting phosphor for practical application in full-visible-spectrum white pc-LEDs. One area for future work might be to investigate and understand the influence of emission peak position and width of cyan phosphor on the values of both CRI and luminous efficacy of white pc-LEDs, which in turn can help in designing and optimizing such a perfect cyan phosphor. Hopefully, in the near future the full-visible-spectrum white pc-LEDs could deliver bright and energy-saving light sources that accurately mimic natural sunlight without producing any harmful ultraviolet light.

Conflict of interest

The author declares that he has no conflict of interest.

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Xiaoyong Huang obtained his Ph.D. degree from South China University of Technology in 2011. Afterwards, he worked as a postdoctoral researcher at the National University of Singapore. His current research mainly focuses on the design, synthesis and characterization of functional materials towards applications in photonic and optoelectronic devices, sensors, and photocatalysis.