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Review article

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Optimizations of luminescent materials for white light emitting diodes toward healthy lighting

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ABSTRACT

White light emitting diodes (wLEDs) have been widely used as the green lighting sources. The commercial wLEDs devices are mainly achieved through the combination of blue emission chips and yellow phosphors, which offer advantages of high efficiency and long lifetime. However, the color rendering index (CRI) of traditional wLEDs is low due to the lack of red components. In recent years, with the improvement of the quality of life, a lot of efforts have been paid to improve the performance of wLEDs devices related to CRI, correlated color temperature, light uniformity, luminous flux, etc. In this article, we summarize the recent advances on the optimization of wLEDs toward healthy lighting. Brief introductions on the fundamentals of healthy effect of lighting are presented, followed by discussions of current methods to realize wLEDs devices. Special overviews on strategies for luminescent materials of wLEDs lighting devices are also discussed.

1. Introduction

White light emitting diodes (wLEDs) are becoming the most popular lighting devices around the world, which are typically fabricated by blue emission chips with rare-earth doped phosphors with yellow emission under the excitation of blue light to produce white light [1–3]. Compared with previously used lighting devices such as incandescent and fluorescent bulbs, wLEDs use less energy with no glass or mercury, negating the risk of glass shards or toxic metal floating which have been regarded as the direction of future lighting [4]. For example, it is forecast that LEDs will penetrate around 86 % of electrical lighting installations by 2035 in the United States [5]. The popularity of electrical lighting has also put people's attention on the environmental impacts of the artificial light at night itself [6]. Human's biological clock controlling circadian rhythms have been changed for increasing reliance on artificial lighting found in existing studies [7]. Apart from that, too much blue light and the lack of red light in the light source will lead to poor color rendering of the light. The original color of the object can't be presented, which can easily lead to visual fatigue, and will also affect people's mood and appetite to a large extent. Besides, human visual perception depends on the regulation of pupil and photoreceptor cells. The pupil adjusts its size according to the strength of the external light to make the luminous flux on the retina remains moderate. Therefore, when the eyes are in uneven light for a long time, it will make the eyes tried, and to a certain extent, it will destroy the photosensitive cells on the retina, so that vision is affected. In view of the impact of light source on human health, there is an urgent

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need to improve light quality. In this paper, we introduce the influence on health of artificial light, current fabrication methods of wLEDs, analyze its advantages and disadvantages, list the performances of healthy light source and summarize the optimization strategies for the most commonly used wLEDs at present, and put forward the prospect of the future lighting.

2. Fundamentals of healthy lighting

2.1. Visual effect of lighting

Ambient lighting is vital for visual health of human eye, including the spectral composition and intensity of light. There is increasing evidence that the spectral composition of ambient lighting can influence normal ocular growth and refractive development and the intensity of ambient lighting has a great influence on the ciliary muscle because it needs to adjust focal distance and light exposure according to luminance of lighting environment. The impact of the spectrum on visual health can't be underestimated. Infant rhesus monkeys were used as experimental subjects to draw a conclusion that the long-wavelength red light greatly reduced the likelihood of developing either form-deprivation myopia or compensating myopic changes in response to imposed hyperopic defocus [8]. Besides, it has been preliminarily demonstrated that 650 nm low-level red light not can slow down myopia progression, but also reverses the myopia progression in human [9,10]. In contrast, short-wavelength blue light may lead to retinal damage and degeneration and the primate threshold of phototoxicity for wavelength between 400 and 450 nm was experimentally fixed at 22 J/cm² [11–13]. At the same time, a lighting environment with suitable luminance is also needed for visual health. More consumption of strength is needed from ciliary muscle adjusting light exposure to obtain clear image in retina in dark ambient lighting contributing to visual fatigue. [2,14] Further experiments imply that luminance above 80 cd/m² would be helpful to prevent the accumulation of visual fatigue [15].

2.2. Non-visual biological effect of lighting

Apart from a visual effect, light entering the human eyes has also an important non-visual biological effect on the human body [16–18], which is mediated by the non-image-forming intrinsically photosensitive retinal ganglion cells (ipRGCs) in the retina [19,20]. These effects are manifested in changes in sleep behavior [21,22], alertness [17,18,21], mood [18–20]. A third type of light-sensitive cells unlike cone and rod photoreceptors, ipRGCs, can affect the circadian phase by reporting light stimuli to the suprachiasmatic nucleus (SCN) to slow down the melatonin secretion, which have spectral sensitivity to light in the blue part around 460–480 nm [19, 22] The circadian stimulus value increases with corneal illumination and decreases with correlated color temperature (CCT) in a certain range and the artificial lighting with the CCT of 2700 K and 4000 K can be selected to reduce the impact on circadian phase [22]. The ipRGC-driven effects and mechanisms of light on mood have been studied [20,23]. The means of positive mood always decreased with an increase in CCT [24]. Multiple studies have proven that lighting environment with CCT of 4000 K is beneficial for both circadian phase and mood (Fig. 1) [19,24].



Fig. 1. The distributions of the subjective perception feedback (Reprinted with permission from Ref. [24], copyright 2022, Elsevier).

3. Product features of wLEDs

There are several parameters to evaluate the characteristics of ambient lighting, including CCT, color rendering index (CRI), CCT uniformity, luminous flux, etc. Among them, CCT and CRI are mainly affected by the spectral composition of light. Luminous flux defines the light emitted by a source such as a lamp or received by a surface, irrespective of direction, which can measure the intensity of light. Light uniformity refers to whether the spectral composition and intensity of light are consistent at different angles. It is generally accepted that the spectral composition and intensity of light have a huge impact on body's health, so a suitable light can be selected according to these parameters to achieve healthy lighting. CRI represents the ability of a light source to accurately reproduce the color of the object it illuminates. When there is little or no main wave reflected by the object under the reference light source in the light source spectrum, the color will produce a significant chromatic difference. The CRI of sunlight is defined as 100, and the larger the CRI, the closer to the natural light source [25]. CCT is an important measurement standard of wLEDs products, which is related to the color characteristics of wLEDs light source display [26]. The more blue-components in the emission spectrum of lighting source, the higher CCT. CCT uniformity is one of the major problems in wLEDs fabrication, which is associated with the different ratios of blue and vellow emissions, leading to different CCTs at various angles. As a result, there is a vellow ring phenomenon in the application of wLEDs bring a poor lighting experiments [27]. Besides, in the process of the light emitted from the chip through the encapsulating material layer into the air, there are total reflection at the interface reduce the light extraction efficiency and the luminous flux because the difference of refractive index (RI) between semiconductor chips (>2.4 for blue chips of gallium nitride) and encapsulating materials (1.5–1.6 for commercial silicone) [28]. From the point of view of human health and energy saving, we need a white lighting source with high CRI, low CCT, uniform light output and high luminous flux.

Commercial wLEDs are usually approached by integrated packaging of a diode generating a short wavelength and phosphors absorbing a few short wavelength photons and converting them into longer wavelength photons, because white light sensation can be caused when two photons of complementary wavelengths arrive simultaneously on the human eye (Fig. 2a) [2,11,29-32]. The advantages of this combination is that it is simple to make and has the lowest cost and highest efficiency among all the combinations of wLEDs. The efficiency of this kind of wLED is affected by both LED chips and phosphors. Low CRI and high CRI is the biggest deficiency of the wLEDs realized by combining the blue LED and YAG:Ce, mainly caused by the weak luminosity of the phosphor in the red region, and more blue light of short wavelengths than a sunset, originating in narrowband blue-light emission from a semiconductor LED. Apart from that, wLEDs can be composed of ultraviolet LED (UV-LED) chip and phosphors with different color (Fig. 2b) [33,34]. The principle of white lighting is that UV-LED pump different phosphors emitting red, green, blue lights respectively, and the white light can be obtained by adjusting the ratio of three phosphors. Because UV light will not participate in the composition of white light, it is much simpler than blue LED to control the color and the higher CRI and lower CCT can be obtained. But the low efficiency of UV-LED which drops dramatically at wavelengths below 400 nm limits its commercialization, and because the energy difference between the excited and emitted photons is a natural energy loss, when UV light is converted to red, its energy loss is 10 %-20 % higher than that when it is converted from blue light. There is color reabsorption between the mixtures of different phosphors, and it is difficult to match the absorption wavelength of the tricolor phosphor with the emission wavelength of its LED at the same time, which will have a greater impact on lumen efficiency and CRI. What's more, the uneven grain size and specific gravity (affecting settling velocity) of phosphors make inhomogeneous color and intensity distribution in the whole system, which also reduces the conversion efficiency of phosphors. Another way of wLEDs implementation is to combine red LED, green LED and blue LED chips into a pixel to achieve white light (Fig. 2c). This way can control the LEDs with different color separately, leading to a high flexibility of color adjustment and can obtain an ideal light source. Besides, the color purity of white light obtained by this way is also high, so this wLED has a huge market potential in large liquid crystal display (LCD) backlight. But the three colors need to be compensated with feedback circuits to keep the light stable, so that the production cost of white light synthesized by this method is the highest. Table 1 summarize the characteristics



Fig. 2. Three approaches to relized wLEDs for lighting. a) wLED composed of blue LED chip and yellow phosphors, b) wLED composed of UV-LED chip and phosphors with different color, c) wLED composed of three-primary color LED chips. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

of three types wLEDs. In general, the most mature method at present is the combination of 460 nm blue InGaN chip and yellow light YAG:Ce phosphor into wLEDs. In view of the problems existing in this combination, optimizing the phosphor to improve the quality of lighting sources and actively exploring new phosphor to replace YAG:Ce is one of the main topics in the current research and development of wLEDs.

4. Optimization Strategies of wLEDs

4.1. Tuning the emission spectrum of phosphors

Typical unit of wLEDs consists a blue emission chip with a certain amount of yellow luminescent particles encapsulated in packaging materials. As key materials for tuning the emission spectra of wLEDs, luminescent particles are supposed to have the following characteristics: a) excitation spectrum exhibits well overlap with emission of blue LED chip in the wavelength region of 420–480 nm, b) high stability under practical application conditions such as oxygen, carbon dioxide, chemical reagents, water and thermal treatment, c) emission spectra with suitable CRI and CCT when combined with blue light of the chip, d) the luminescent materials can be synthesized in mild conditions, including easy control of particle morphology, harmless production and reasonable cost. To date, many kinds of luminescent materials have been studied for wLEDs, including carbon dots [35–40], semiconductor quantum dots [30,31,41], and inorganic phosphors [1,42–44]. Among them, luminescent ions doped inorganic crystals are most commonly used luminescent materials in LED lighting, which can make white light softer, improve the color reproduction and light efficiency of devices. Tuning the emission spectrum of light is an effective method to improve the CRI and reduce CCT, includes doping modification of phosphors to make the original yellow phosphor to improve the CRI [41]. As shown in Table 2, the CRI is significantly improved after the addition of red components, and it can also be seen that, compared with optimizing the existing yellow luminescent materials, directly adding red phosphors has a more obvious effect on reducing the value of CCT, which less than 4000 K beneficial for both circadian phase and mood.

4.1.1. Yellow-emitting phosphors pumped by blue LED

Ordinary products of wLEDs are approached by integrated packaging of blue chips made of GaN and luminescent particles of YAG: Ce.[1] The space group of $Y_3Al_5O_{12}(YAG)$ belonging to cubic crystal system is *Ia3d*, and YAG exhibits excellent optical and thermodynamic performance. Y in the dodecahedral sites has an ionic radius similar to that of most rare earth elements and is easily substituted. In case of doping with Ce^{3+} , the Ce ions occupy Y resulting in a shift of $Ce^{3+}s$ 5d-4f emission corresponding to yellow-green spectral region. People's eyes and brain work together to blend human perception of separate bands of blue color from chips and yellow color from phosphors into a white light that may be illuminating the object viewed [7,51]. In order to improve the light source quality of wLEDs based on YAG:Ce, many efforts have been paid optimize the luminescent center ion and the structure of host. For instance, the YAG, can be written as A₃B₂C₃O₁₂, where A, B, and C represent three different lattice positions, and the intensity crystal field strength in the dodecahedral sites of YAG directly depends on the R^{3+} - O^{2-} cation–anion distances. As the ionic radius of R^{3+} increases, the corresponding cation-anion distances become smaller, and the crystal field strength is larger [52–55]. As shown in Fig. 3a and b, the substitution of Gd^{3+} for Y^{3+} results in red shift by increasing the crystal field splitting effect for the ionic radius of Gd^{3+} (0.1053 nm) is larger than that of Y^{3+} (0.1019 nm) [56,57]. Similarly, the substitution of Tb^{3+} with larger ionic radius (0.104 nm) for Y^{3+} can play a similar role in tuning the emission spectrum of light [54,55]. The above way only substitute cations in single-site, and there are other way to substitute cations is double-site substitution. The existed research has demonstrated that when $Mg^{2+}-Si^{4+}/Ge^{4+}$ as ion pairs substituting for $Al(1)^{3+}-Al(2)^{3+}$ are synchronously incorporated into the YAG:Ce host lattice, the 5d level is lowered, and the emission peak shifts to longer wavelength [50,58,59]. However, the sites of substitution are disputed, and there are another theory that the Y^{3+} -Al³⁺ are substituted by M^{2+} -Si⁴⁺ (M = Ba, Sr, Ca, Mg), and red-shift increases with the decrease of M^{2+} radius [60]. But one thing is certain, that is, the $Ce^{3+}-O^{2-}$ bond length is decreased, resulting in redshift according to the following equation:

$$D_q = \frac{1}{6} Z e^2 \frac{r^4}{R^5} \tag{1}$$

where Dq is a measure of the energy level separation, Z is the anion charge, e is the electron charge, r is the radius of the d wavelength, and R is the bond length [58]. Interestingly, interstitial defects caused by carbon atoms also can shorten the $Ce^{3+}-O^{2-}$ bond length,

Table 1	
Characteristics of three types wLEDs.	

Feature	Blue LED + yellow phosphor	UV-LED + RGB phosphor	RGB LED
CRI	Moderate	High	High
CCT	High	Low	Low
CCT uniformity	Low	High	Low
Structure	Simple	Simple	Complicated
Cost	Low	Low	High
Efficiency	High	Low	High

Table 2

Typical luminescent materials and its devices.

Strategy	Materials	Emission/nm	wLED performance		Reference
			CCT/K	CRI	
Adding red-emitting materials	Cs ₂ NbOF ₅ :Mn ⁴⁺	631.5	3168	90	[46]
	Li ₃ Na ₃ Al ₂ F ₁₂ :Mn ⁴⁺	629	3874	90.6	[2]
	K ₂ TiF ₆ :Mn ⁴⁺	580-660	2736	87.3	[43]
	K ₃ TaO ₂ F ₄ :Mn ⁴⁺	630	3488	90.0	[47]
	SrLiAl ₃ N ₄ :Eu ²⁺	650	2875	91.1	[48]
	CD	620	3827	92.7	[29]
Optimizing yellow-emitting materials	Ca4SiAl3N7:Ce3+	568	4068	89.6	[49]
	Ce:Y _{2.99} (Mg _x Al _{5-2x} Si _x)O ₁₂	571	4384	81	[50]
	CaY ₂ Al ₄ SiO ₁₂ :Ce ³⁺ ,Mn ²⁺	543, 616, 750	5460	90.5	[45]

leading to a larger crystal field splitting of 5d orbital of the Ce³⁺ atom and bigger 5d centroid shift [61].

Based on the energy transfer from Ce^{3+} to red light emitting activators, such as Mn^{2+} [45,62,63], Pr^{3+} [64], Cr^{3+} [65], etc., can also supplement the red component of the spectrum. Mn^{2+} is a common red-light emitting activator, the energy transfer process is briefly described with the example of Mn^{2+} . As shown in Fig. 3c and d, the energy transfer process can achieve from Ce^{3+} to neighboring Mn^{2+} via non-radiation transition because the lowest 5 d¹ energy level of Ce^{3+} is a little higher than the ${}^{4}T_{1}$ of Mn^{2+} . And then the ${}^{4}T_{1}$ excited electrons transfer back to ${}^{6}A_{1}$ ground state, which emits three bands at 528, 590, and 745 nm supplementing the red light, and the CCT of wLED can decrease from 4044 K to 3152 K [63].

In addition, phosphors with nitride as hosts are promising candidates for wLED [42,49,66]. These materials mainly are obtained by introducing N into the crystal structure of silicates or aluminosilicates, which can meet the needs of wLED for color conversion, because their coordination environment with strong rigidity, diverse structure and high covalency can reach strong absorption of blue light and long wavelength emission and high-quantum efficiency [49]. As reported, some phosphors belonging to nitridosilicate and nitridoaluminosilicate, such as $Ca_{8-x}Mg_7Si_9N_{22}:xCe^{3+}$ [42], $(La,Ca)_3Si_6N_{11}:Ce^{3+}$ [67], $Ca_4SiAl_3N_7:Ce^{3+}$ [49], $Ba_5Si_{11}Al_7N_{25}: Eu^{2+}$ [66], can be effectively excited by the blue chip and emit a strong broadband yellow light. $Ca_4SiAl_3N_7:Ce^{3+}$ has more red light components than YAG:Ce³⁺ accounting for 64 %, and wLED based on it has a CRI of 89.6 and a CCT of 4068 K (Fig. 3e and f) [49]. Compared with the PL spectrum of YAG:Ce³⁺, the Li₂SrSi₂N₄:Ce³⁺ spectrum shows a broader emission band due to the two sites in Li₂SrSi₂N₄ for Ce^{3+} to occupy reflected in Fig. 3g and h [68]. Apart from excellent optical performances, the thermal stability of these phosphors can meet requirement of wLEDs due to the highly condensed three-dimensional network structure based on Si(Al)N₄ tetrahedra and the stable chemical bonding between the atoms. $Ba_5Si_{11}Al_7N_{25}: Eu^{2+}$ phosphor, as an example, can remain >90 % of the initial intensity at 83 K at 473 K and the quenching temperature is about 700 K [66]. But the harsh synthesis condition requiring high temperature and pressure, hinders its development progress. It is key to explore phosphors with nitride as hosts phosphors that can be synthesized at lower temperature and atmospheric pressure with high efficiency and stable emission.

4.1.2. Red-emitting phosphors pumped by blue LED

Adding red phosphor excited by blue light is a direct and effective way to supplement the red component of wLED light, which can also reduce CCT and improve CRI [2,71]. Therefore, the study of red phosphor for wLED has attracted extensive attention in recent years. Eu^{2+} and Mn^{4+} are the most commonly used red luminous centers. A comparison of some recently reported red-emitting phosphors is shown in Table 3. The first generation of red phosphors are Eu^{2+} activated sulfides represented by CaS and SrS [72]. Although they can make up for the deficiency of red light, these sulfide phosphors cannot be commercialized because they are hygroscopic and will produce toxic gases during the hydrolysis process. At present Eu²⁺ doped nitride phosphors are the most mature red materials with high quantum efficiency and high quenching temperature [73]. Compared with the traditional oxide matrix, the nitride matrix has a more excellent effect in rearranging electron cloud, which can split the 5d energy level, ultimately leading to a smaller 4f-5d energy level difference to achieve a better match between phosphors and blue LED chips (Fig. 4a-c) [74]. However, the preparations of nitride phosphors are in harsh conditions, usually requiring high temperature >1700 °C and high pressure >0.9 MPa [75]. Mn⁴⁺, a transition metal ion, has been regarded as a promising red emission luminescence center, which is doped in oxides [76], fluorides [2,44,77] and oxyfluoride [46,47] matrix to obtain red phosphors showing good luminescence properties and playing an important role in improving the CRI of wLEDs. The ground state energy level of Mn^{4+} is ${}^{4}A_{2}$. When it is excited, ${}^{4}A_{2}$ transitions to excited states including ${}^{4}T_{1}$ and ${}^{4}T_{2}$, and then produces ${}^{2}E \rightarrow {}^{4}A_{2}$ radiation transitions, which lead to red emission (Fig. 4d–f). Fluoride matrix with Mn⁴⁺ doping has the characteristics of mild synthesis conditions, low crystallization temperature, and strong absorption of blue light. And K₂SiF₆:Mn⁴⁺ and K₂TiF₆:Mn⁴⁺ as the typical example, have been commercialized [32,46]. However, toxic hydrofluoric acid is used in the preparation process, and the phosphor is unstable in high humidity environment. After water absorption, decomposition reaction will generate MnO₂ covering phosphors and toxic HF gas as follows, which reduces the color stability of wLEDs and pollute the environment [44]. Some researchers focused on improving the water resistance of phosphors, including surface coating to isolate water [43,77], or optimizing the crystal field of matrix to improve the stability of Mn⁴⁺ [44]. As shown in Fig. 4g, a Mn^{4+} -activated fluoride red phosphor, $K_2TiF_6:Mn^{4+}(KTF)$, moisture resistance of which became better after superhydrophobic surface modification with octadecyltrimethoxysilane, can maintain at 83.9 % or 84.3 % of luminous efficiency after being dispersed in water for 2 h or aged at high temperature (358 K) and high humidity (85 %) atmosphere (HTHH) for 240 h, respectively. The moisture resistance can also be optimized from the perspective of crystal field of matrix. For example, the moisture and thermal stability of



Fig. 3. a) Schematic energy diagram of Ce^{3+} in YAG, TAG, and GAG (Reprinted with permission from Ref. [69], copyright 2017, Royal Society of Chemistry), b) emission spectra of Ce:(Y,Gd)AG ceramics with different Gd³⁺ ions substitution (Reprinted with permission from Ref. [70], copyright 2019, Elsevier), c) schematic illustration of the energy transfer process in YAG:Ce,Mn, d) PL spectra of YAG:Ce,Mn under 460 nm excitation (Reprinted with permission from Ref. [63], copyright 2020, Junrong Ling et al.), e,f) EL spectrum of Ca₄SiAl₃N₇:Ce³⁺ based WLED(e) and YAG:Ce³⁺ based WLED(f) with 460 nm blue LED chip as excitation source (Reprinted with permission from Ref. [49], copyright 2023, Elsevier), g,h) the energy level diagram(g) and the Gaussian peaks fitting for PL spectrum(h) of Ce³⁺-doped Li₂SrSi₂N₄ phosphor (Reprinted with permission from ref. [66], copyright 2018, Royal Society of Chemistry). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

 $K_2Ge_{0.6}Ti_{0.4}F_6$: Mn^{4+} under cation substitution can keep 71 % or 83 % of the original luminescence intensity after immersing in deionized water for 180 min or treated under 423 K, and a wLED with high CRI of 92.4, low CCT of 3113 K and high luminous efficiency of 123.1 lm/W can be obtained by encapsulating it with commercial yellow phosphors on blue chip [44]. In that work, the mechanism was explained that the moisture stability of phosphors are significantly enhanced under the synergistic effects of the two following reasons: the expanded unit cell caused elongation of Mn–F bond by substituting the Ge⁴⁺ (r = 0.53 Å) site with Ti⁴⁺(r = 0.60 Å) promotes the dissociation of [MnF₆]²⁻ and then enhance the reaction between [MnF₆]²⁻ and H₂O (Fig. 4h), meanwhile, the complex with inner orbital hybridization [TiF₆]²⁻ based on the six empty p^3d^2 hybridization orbitals. In recent years, developing novel green

Table 3

Typical red phosphors.

Light Emitting Activators	Host	Materials	Excitation /nm	Emission /nm	Reference
Mn^{4+}	Oxyfluoride	Cs ₂ NbOF ₅ :Mn ⁴⁺	467	631.5	[46]
		$K_3 TaO_2 F_4: Mn^{4+}$	360, 460	630	[47]
		LiAl ₄ O ₆ F:Mn ⁴⁺	150-550	662	[81]
	Fluoride	Li ₃ Na ₃ Al ₂ F ₁₂ :Mn ⁴⁺	467	629	[2]
		K ₂ TiF ₆ :Mn ⁴⁺	462	580-660	[43]
		$K_2Si_{1-x}F_6:xMn^{4+}$	450	630	[78]
		Na ₃ GaF ₆ :Mn ⁴⁺	467	626	[82]
		K ₂ LiAlF ₆ :Mn ⁴⁺	300-500	635	[83]
	Oxides	CaAl ₂ Si ₂ O ₈ :Mn ⁴⁺ , Mg ²⁺	330	652	[76]
		SrMgAl _{10-v} Ga _v O ₁₇ :Mn ⁴⁺	340	661	[80]
		Sr ₄ Al _{14-x} Ga _x O ₂₅ :Mn ⁴⁺	450	665	[84]
		Li ₂ MgTiO ₄ :Mn ⁴⁺	476	676	[85]
Eu ²⁺	Nitride	SrLiAl ₃ N ₄ :Eu ²⁺	400-600	650	[48]
		CaAlSiN ₃ :Eu ²⁺	460	550-800	[86]
		Sr ₂ Si ₅ N ₈ :Eu ²⁺	421	616	[87]
	Sulfide	(Sr,Zn)S:Eu ²⁺	400–500	606	[88]
		SrS:Eu ²⁺	430–500	609	[89]



Fig. 4. a-c) The crystal structure(a), configurational coordinate diagram of the ground state of Eu^{2+} and the excited states of Eu^{2+} (b), PL and PLE spectrum(c) of $Ca_5Si_2Al_2N_8$: Eu^{2+} (Reprinted with permission from Ref. [74], copyright 2016, Elsevier), d-f) crystal structure(d), Tanabe–Sugano diagram for the 3 d³ electron configuration of Mn^{4+} (e), PLE and PL spectra(f) of $CaAl_2Si_2O_8:Mn^{4+}$, Mg^{2+} (Reprinted with permission from Ref. [76], copyright 2023, John Wiley and Sons), g) TEM images of KTF modified and images of a drop of water on the modified KTF phosphors (Reprinted with permission from Ref. [43], copyright 2018, American Chemical Society), h) crystal structure of $K_2Ge_{1-x}Ti_xF_6:Mn^{4+}$ phosphors(Reprinted with permission from Ref. [44], copyright 2023, Elsevier).

synthetic routes for the highly efficient fluorides with Mn^{4+} doping red phosphors is also a hot spot, and the main method is to reduce the use of toxic HF by replacing it with low-toxic NH_4F/HCl or H_3PO_4/KHF_2 [32,78]. When the oxide exhibiting better chemical stability is used as matrix, the luminous color of Mn^{4+} is located in the deep red emission region, which does not match the sensitive range of human eyes. Besides, compared to fluoride phosphors, the absorption of blue light of is less effective, which can't perfectly satisfy the application for blue-excited wLEDs [79,80]. Red phosphors based on oxyfluorides not only have higher chemical stability than fluoride phosphors, but also has stronger blue light absorption than oxide phosphors [46].

4.1.3. Other frontier luminescent materials for wLEDs

In recent years, semiconductor quantum dots, metal halide perovskites, and luminescent carbon dots with high quantum efficiency, adjustable luminous color and high color saturation are also becoming substitutes for rare earth doped phosphors [30,31,41,90–96]. Semiconductor quantum dots are composed of core material enclosed within a shell of another semiconductor material with a diameter of 2–10 nm as shown in Fig. 5a and b [97]. The size of QDs reflects the properties like optical property, absorbance and photo-luminescence in dependent manner. In addition, the PL emission wavelength of the ternary alloyed QDs can be tuned via adjusting the chemical composition. For example, by means of altering chemical composition, the AIS/ZnS QDs synthesized via a microwave-assisted aqueous method exhibited tunable emission ranging from 540 to 622 nm (Fig. 5c) and yellow AIS/ZnS quantum dots with high PL QY of 58.27 % were applied on commercial blue InGaN chips for wLED with a CRI of 75.6 and a CCT of 4393 K (Fig. 5d) [91].

Perovskite are organic–inorganic or all-inorganic materials with the form of ABX₃, and their electronic and optoelectronic properties are tightly related to and thus can be tuned to be ideal materials for wLED by the size, shape, surface, structure and component [98–100]. For example, CsGeBr₃ demonstrated the capability to emit white light under UV radiation and the device fabricated by



Fig. 5. a) Schematic illustration for the synthesis of thick shell InP/ZnS/ZnS QDs, b) TEM image of InP/ZnS and InP/ZnS/ZnS QDs (Reprinted with permission from Ref. [97], copyright 2020, John Wiley and Sons) c) PL spectra ($\lambda_{ex} = 460$ nm) of AIS/ZnS QDs, d) Emission spectrum of the LED based on AIS/ZnS QDs (Reprinted with permission from Ref. [91], copyright 2020, Elsevier).

CsGeBr₃ combined with red-emitting MOF:Eu³⁺ phosphors can exhibit a commendable CRI of 92 and a CCT of 3020 K [96]. In another work, Zhang et al. develop the synthesis of heterostructural CsPbX₃-PbS (X = Cl, Br, I) quantum dots via a room temperature in situ transformation route [98]. The CsPbX₃-PbS quantum dots show uniform size distribution as the TEM images shown in Fig. 6a and exhibit tunable dual emission feature with the visible and near-infrared photoluminescence (PL) as the PL spectra shown in Fig. 6b [98]. As one of rising stars in luminescent materials, the shortcomings of the traditional perovskites that limiting their practical applications, including the poor environmental, thermal, and optical stability, are being overcome gradually by controlling the composition between cations and halides or by replacing/doping cations at A- and B-sites with other metal cations, and so on [94]. Besides, more environmentally friendly lead-free halide perovskites make the practical application more likely [101].

The emerging carbon dots that can be easily prepared by green procedures have recently been demonstrated to be a superior color converter for realizing wLEDs excited by blue chips, which not only maintains the excellent luminescence performance of quantum dots, but also has the advantages of low toxicity, low cost and wide source of raw materials [102,103]. For instance, Shen et al. develop an in situ solvothermal process for synthesizing CDs using reaction mixture of urea, trisodium citrate and DMF, as shown in Fig. 7a [92]. The CDs show uniform distribution with an average particle diameter of 4.5 nm (Fig. 7b) and wLEDs devices with CCT of 4492 K can be achieved in the CDs based LEDs approaching to the coordinate (0.33, 0.33) of pure white light (Fig. 7c) [92]. In another work, a kind of CD powder with high PL QY of 65 % in the range of 500–620 nm was synthesized by a simple one-pot microwave heating method, and wLED with high luminous efficiencies (>90 $\text{Im} \cdot \text{W}^{-1}$) were realized using the CDs [93]. However, considering the high cost and difficulty to largely produce of these frontier luminescent materials, rare earth doped phosphor is still the main material to achieve wLEDs. Therefore, producing high-quality luminescent materials on a large scale remains challenging [104].

Although adding red phosphors has a more obvious effect on reducing the value of CCT compared to optimizing the existing yellow luminescent materials, the uneven grain size and specific gravity of phosphors make inhomogeneous color and intensity distribution in the whole system, which also reduces the conversion efficiency of phosphors. As a result, among these Optimization Strategies of tuning the emission spectrum of phosphors, optimizing the luminescent center ion or the structure of host of YAG:Ce is the simplest because this kind of phosphor with good performance can be obtained only by using cheap oxides as raw materials for high temperature calcination. While the high temperature and pressure environment required for the preparation of nitride phosphor requires higher performance of the equipment. Frontier luminescent materials with excellent performance, including semiconductor quantum dots, metal halide perovskites, and luminescent carbon dots, are prepared by colloidal and hydrothermal synthesis mainly, large-scale preparation of which is difficult and costly.

4.2. Tuning the scattering processes of light

In commercial wLEDs, the yellow-emitting phosphors were mixed with transparent encapsulating material and then dispersed in a cup reflector or directly coated on the LED chip surface. Due to the simplicity of preparation, the latter direct coating method can achieve mass production, but it suffers from propagation energy losses for the large difference of RI between semiconductor chips and encapsulating materials and it has been shown that nearly 60 % re-emitted yellow rays are backscattered to the LED chip, which seriously decreases the luminous flux [105]. In addition, this direct coating method results in poor quality in angular CCT and the unwanted phenomenon such as "yellow ring". The scattering effect can strongly influence the optical path and weaken the CCT deviation in wLEDs. More importantly, it can increase the amount of received photons passing through layers of packages, which is crucial for the luminous efficiency of wLEDs [106]. There mainly are two methods to tuning the scattering of light, and adjusting structure of the wLEDs is an effective method to enhance the uniformity of CCT [27]. Besides, it has been discovered that scattering capability of nanoparticles not only enable better utilization of blue light which increased the luminous flux, but help to reduce angle-dependent CCT deviation in recent years [106–108].



Fig. 6. a) TEM images and b) PL spectra of the CsPbX₃-PbS (Reprinted with permission from Ref. [98], copyright 2020, American Chemical Society).



Fig. 7. a) Proposed in situ formation process of fluorescent CDs phosphors, b) TEM image of the NCDs in aqueous solution c) the emission spectrum of blue chip and LED based on CDs (Reprinted with permission from Ref. [92], copyright 2018, Elsevier). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

4.2.1. Adjusting structure of the wLED

The remote phosphor coating method, separating the LED chip from the phosphor layer, can improve luminous efficacy by reducing the phosphor layer backscattering light reabsorbed by the blue LED chip [27,109]. Kuo et al. use Monte Carlo method to simulate both of conventional remote phosphor and patterned remote phosphor structures as shown in Fig. 8 [105]. Most of large angle of blue rays and backscattering yellow rays exit the package via the window region, ensuring the intensity ratio of yellow to blue rays with good constancy as compared to the conventional remote structure in larger divergent angle as shown in Fig. 8 and it can also concluded



Fig. 8. a) The ray tracing simulation results, b) angular-dependent CCT(b) of two phosphor distribution structures (Reprinted with permission from Ref. [105], copyright 2011, Optical Society of America).

from the measurement data that the angular-dependent CCT of patterned remote phosphor structure was more uniform in larger angle distribution (Fig. 8b) [105]. For devices with textured sidewalls, the scattering probability of photons at the GaN/air interface is increased and the light extraction efficiency is enhanced since photons are allowed to find escape cones in the horizontal direction [110–112]. Compared with conventional LED without specific designs, LED with 45° sidewalls and a backside reflector can achieve significant improvement of 55.8 %, 49.3 %, 47.2 %, and 55.4 % in the light output power, luminous flux, external quantum efficiency, and wall-plug efficiency respectively [110]. Besides, LED with a hybrid structure incorporating a microhole array, 45° sidewalls, and an appropriate SiO₂ nanoparticle/microsphere passivation layer, shows 50.6 %, 50.9 %, 48.4 %, and 49.9 % enhancements in light output power, luminous flux, luminous efficacy, and wall-plug efficiency. The passivation layer play an important role in reducing reverse-biased leakage current, leading to substantial enhancements in optical properties while LED without any degradation in electrical performance [112]. In general, adjusting structure of the wLED can achieve a higher luminous flux and more uniform CCT by the increased scattering probability and opportunity to find photons escape cones.

4.2.2. Adding non-luminous nanoparticles to the encapsulant

Adding a certain amount of non-luminous nanoparticles to the encapsulant also can improve CCT uniformity and luminous flux of wLEDs, such as ZrO₂ [28,106,113], TiO₂ [114], cellulose nanocrystals (CNC) [107], as summarized in Table 4, since the use of scattering additive in the encapsulant can significantly influence the optical path. As the angular-dependent emission intensity, CNC-embedded wLED structures can exhibit superior performance and produce high-quality white light at different viewing angles. It was found that adding 6 wt% of CNCs to wLED increases the luminous flux by around 33 %, reduces the CCT from 4220 to 3985 K, and reduces angular-dependent CCT variation from 270 to 224 K [107]. Besides, scattering additive with high refractive index (RI) can improve the light extraction efficiency of LEDs because it can bridge the gap of RI between the chips through encapsulant to the air as shown in Fig. 9a and b [113]. Fig. 9a shows schematic diagram of LED encapsulation and the light extraction models at the interface, which indicating that the value of refractive index is one of the most important parameter while evaluating the performance of the encapsulation material to make devices with high light extraction efficiency [113]. Our group have developed a kind of wLED exhibiting significant enhanced luminous flux (form 13.28 to 31.34) and decreased proportion of blue light(3.1 %-1.3 %), resulting in low correlated color temperature was obtain by introducing ultrasmall ZrO₂ nanoparticles in commercial-grade silicone without any additional red phosphors (Fig. 9b) [28]. However, the commercial encapsulant are generally less compatible with nanoparticles leading to mass nanoparticles aggregate, which results in low transparency of light. Surface modification is an effective way to improve the dispersibility of particles [28,113,114]. As reported, the dispersibility of modified TiO₂ particles was greater than that of the as-prepared TiO₂ particles (1.6–2.5 times). Except that the luminous fluxes could be increased by 2.95%–11.79 % by adding TiO₂ particles in the encapsulant, which consistent with the above, the color uniformity will improve with the increasing dispersibility [114]. As a result, controlling the dispersion for nanoparticles in polymeric hosts to avoid the scattering of large aggregated particles and obtain optically transparent hybrid nanocomposites is crucial. Among these non-luminous nanoparticles, ZrO₂ exhibits a remarkable light scattering ability with high refractive (n: 2.18 at 500 nm) and without any visible light absorption due to its large bandgap energy, improving the CCT uniformity and luminous flux of wLEDs [115].

5. Conclusion and outlook

WLEDs has been widely used in daily lives and the property studies and preparation of the most commonly used phosphor YAG:Ce are nearly mature. Many scientists are devoted to exploring new materials or promoting the performance of existing luminescence materials to improve the performance of wLEDs toward energy-efficient, healthy and comfortable lighting. At present, a high CRI of more than 90 has been achieved by doping modification of YAG:Ce to make the original yellow phosphor emission wavelength redshift, and/or adding an appropriate amount of red phosphor into packaging materials. And the CCT uniformity and luminous flux are significantly improved by tuning the scattering of light, such as adjusting structure of the wLEDs, Adding a certain amount of nonluminous nanoparticles to the encapsulant. There are still the following points worth studying to optimize wLEDs: In view of tuning emission spectra, luminescent materials should be improved further, including a) improve the thermal stability of luminescent materials to ensure their stable performance in high power LED devices, b) enhance absorption of the material in the blue light region, that is, improve the light conversion efficiency of wLEDs pumped by blue chips, c) reduce the particle size of phosphors, which affect it's deposition in the packaging materials, to improve the light uniformity, d) optimize the phosphor production process, reduce energy consumption, and reduce the use of toxic raw materials, e) develop green preparation methods, expand the production scale of frontier luminescent materials, and reduce costs. On the other hand, it is also necessary to make full use of non-luminous particles for better utilization of blue light to increase the luminous flux, reducing angle-dependent CCT deviation. To solve the problem of transparency reducing caused by nanoparticles aggregation, the dispersion for nanoparticles in polymeric hosts should be controlled to avoid the scattering of large aggregated particles and obtain optically transparent hybrid nanocomposites.

Data availability statement

Data availability statement Data sharing is not applicable. No data was used for the research described in the article.

CRediT authorship contribution statement

Fen Wang: Writing - original draft, Conceptualization. Hao Pan: Writing - review & editing, Investigation, Conceptualization.

Table 4

Typical non-luminous nanoparticles tuning the scattering processes.

Nanoparticles	Luminous flux	CCT/K	CCT deviation/K	Reference
ZrO ₂	31.34 (100 mA)	4573		[28]
ZrO ₂	> 30 (120 mA)	< 4500	7	[106]
TiO ₂	34.14 (121 mA)	7651.1	124.2	[114]
ZnO	70 (350 mA)		40	[108]
CNCs	328 (2.6 V)	< 4300	59	[107]



Fig. 9. a) schematic diagram of LED encapsulation and the light extraction models at the interface (Reprinted with permission from Ref. [113], copyright 2019, Elsevier), b) CIE color coordinates and optical photograph of the wLEDs (Reprinted with permission from Ref. [28], copyright 2019, Elsevier). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Wei Mao: Writing – review & editing, Resources. Dan Wang: Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Dan Wang is an associate editor of Heliyon Chemical Engineering Section. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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