

Article

Effect of Amorphous Photonic Structure Surface Mounted on Luminous Performances of White LED

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Abstract: We fabricated amorphous photonic structures (APSs) with different periods and hole diameters. The GaN-based white light emitting diodes (LEDs) at nominal correlated color temperatures (CCTs) of 5000 and 6000 K were surface mounted by these APSs. The electroluminescence (EL) measurements showed less luminous efficiency (LE) and higher CCT than the ones of the virginal white LEDs. However, the LEs of many APS-mounted white LEDs increased compared to white the LEDs without APSs at the same CCTs. A finite-difference time-domain (FDTD) simulation was carried out on the APSs surface-mounted white LEDs and bidirectional scattering distribution functions (BSDFs) of different emissions were transferred to a Monte Carlo ray tracing simulation. The simulated LEs and CCTs conformed well to the experimental ones. The effects of the blue emission transmission and phosphor concentration were simulated to predict the absolute LE enhancement methods for white LEDs. At last, the hopeful APSs for high Les' general lighting were discussed.

Keywords: white light emitting diode (WLED); amorphous photonic structure (APS); luminous efficiency (LE); correlated color temperature (CCT); Fresnel loss; finite-difference time-domain (FDTD)



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1. Introduction

Phosphor-based white light-emitting diodes (LEDs) are low-cost and popular lighting devices because of their high luminous efficiency, long lifetime, and environmental protection. Although the luminous efficiency of white LEDs with an InGaN active layer in the laboratory have been achieved more than 300 lm/W [1], the commercial ones usually showed a lower efficiency below 200 lm/W. In a phosphor-coated white LED, the interplay between multiple scatterings, emissions, and absorptions of light would be concerned carefully [2]. To reduce the back-scattering and inelastic scattering of blue and yellow lights, some techniques, such as scattered photon extraction (SPE), remote phosphor, multi-layered phosphor, and enhanced light extraction by internal reflection (ELiXIR) are developed [3–6]. The phosphor layer features include the encapsulant and phosphor materials, volume or mass concentration and size distribution of particles, matrix blending, and so on, which can adjust the global performances of the phosphor layer [7–9]. The efficient matching of refractive indexes among the encapsulant, package structure, chip, and phosphor is the key issue in light extraction [10,11]. The light extraction efficiency (LEE) nearly approaches 100% when the resin with a refractive index above 1.8 encapsulated the GaN-based LED [11]. Such high index resin is not available due to its reliability. To improve the LEE of white LEDs is still important and significant in the fields of general lighting and backlight of liquid crystal display. With the maturation of the material and processes of the white LED encapsulation, even several percent enhancement of LEE becomes very difficult [12].

Recently, some nanostructures were applied on the white LED packaging to improve the color conversion efficiency, directionality, and spontaneous emission rate (SER) [13–22]. Quantum dot (QD) material is a type of good phosphor for white LEDs because of its high quantum yield, narrow spectral width, and high SER [13–15]. However, the agglomeration, self-absorption of luminescence, and surface state of the QDs lead to low efficiency in the encapsulant. Dang et al. presented a nonporous (NP) GaN by a simple electrochemical (EC) etching technique, where the QDs do not agglomerate and Förster resonance energy transfer may occur by being immediately proximal to the active layer [13]. The metallic nanostructures cause the surface plasmon coupling to both QD and quantum wells to enhance color conversion [15,22]. The ZnO nanorod is fabricated on the AZO seed layer using the aqueous solution method, which increases the luminous flux of white LED by 60% [16]. The directionality of light emission can be determined by dielectric Vogel spiral arrays or rare-earth phosphor-patterned films [17,18]. The directional emissions are beneficial to imaging and projecting devices. Due to the scattering of the phosphor, the total internal reflection (TIR) at the light extraction surface is not paid much attention. Many people find that flat devices show similar LEEs compared to the structured devices [2,10,23,24]. However, the TIR and Fresnel losses still remain on the light extraction surface [11,16,25–27], which should be alleviated to enhance the LEE of white LEDs.

There are some reports on mounting photonic structures onto the light extraction surface of monochromatic LED packages [28–30]. It is found that the LEE of blue LEDs can be improved even if it has reached over 90% [29]. The moth-eye technique can eliminate the Fresnel reflection effectively [31]. Some moth-eye structures are amorphous photonic structures (APSs) that show short-range order only. So, it is expected that the LEE will be improved when the white LED package surface is mounted with APSs. Moreover, APSs show a strong light extraction ability for a broad spectrum because of its antireflective structures and coherent scattering [32–34]. However, it has not been used in the white LED yet, which consists of a narrow blue emission and a broad yellow emission. In APS optimization, the finite-difference time-domain (FDTD) is used [35]. However, the FDTD simulation is limited to be operated on several microns by the computation capacity. It should combine with other simulators, such as Monte Carlo ray tracing to complete the large-size white LED package simulation [36]. In this work, we proposed an easy and feasible method that nanoscale intermediate polymer stamp (IPS) APSs were transferred to the silicone surface of white the LED package by the surface-mounting method, which was well used in the white LED package by Lumileds Co. [37]. Combined with the nanoimprint technique, it will open a door to apply the nanostructures in the white LED structures. The different periods and hole diameters of APSs were used on the different correlated color temperatures (CCTs). Although all the APS-mounted white LEDs showed less luminous efficiency (LE) than the virgin white LEDs, the LEs of some APS-mounted white LEDs were enhanced when compared to the same CCT white LEDs without APSs. Moreover, some light output powers (LOPs) were enhanced by surface-mounted APSs. At last, the FDTD combined with ray tracing simulations were carried out, which explained the LE reduction with the APSs and predicted the LE enhancement with appropriate APS structures.

2. Material and Methods

APS structures were fabricated from commercial nanoporous anodic aluminum oxide (AAO) templates, typically as shown in Figure 1a. The templates were the silicon replicas of the original AAO structures on aluminum sheets. In this work, the quasi-periods of AAO templates were about 450 and 800 nm. The hole diameters were 400, 300, and 200 nm for the 450 nm-period AAO templates and the depths were about 500 nm, which corresponded to APS-1, APS-2, and APS-3. The hole diameters were 600 and 700 nm for the 800 nm-period AAO templates and the depths were about 600 and 700 nm, respectively, which corresponded to APS-4 and APS-5. Figure 1b shows the schematic diagram of APS transferring and surface-mounting process. First, the APS was transferred to the intermediate poly-

mer stamp (IPS) using nanoimprint technology. The detailed processes of nanoimprint and demolding would be found in [29]. Subsequently, PDMS (Sylgard184, Dow Corning 10:1 ratio with the curing agent) acting as the adhesive coating was spin-coated onto the commercial white LED packages (length = 5.6 mm, width = 3.0 mm, height = 0.77 mm, model: HV56301ZMQ20DC from Hualian Electronics Corp., Ltd., Xiamen, China). The schematic diagram and feature size of the white LED package is shown in Figure 1c. The IPS and PDMS covered the surface of white LED and the thickness of IPS and PDMS was about 0.19 mm. The nominal CCTs of the white LEDs were 5000 and 6000 K, respectively. Finally, the IPS was bonded onto the white LED using PDMS adhesive. The whole device was then heated for 30 min at 100 °C to cure the PDMS. The top-view photo of the completed device is shown in Figure 1d.

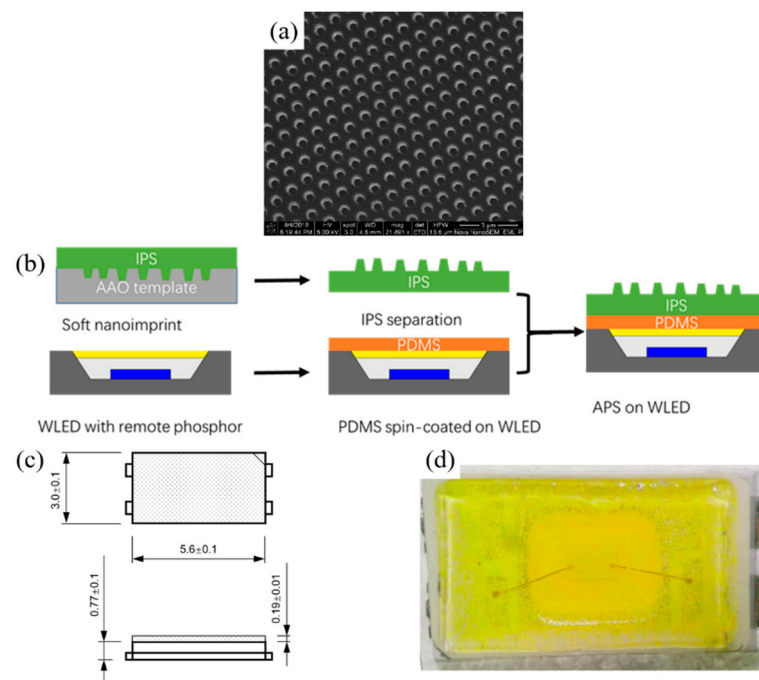


Figure 1. (a) SEM images of typical APS imprinted using AAO template. (b) The schematic diagram of APS transferring and surface-mounting process. (c) The schematic diagram and feature size of the white LED with and without package. (d) The top-view photo of the completed device.

The surface morphology of the APSs was examined using a scanning electron microscope (SEM, Nova Nano SEM 430, FEI, New York, USA). The electroluminescence (EL) spectra of the LEDs with and without APSs were measured using an Everfine HAAS-2000 spectroradiometer under the constant mode by using an integrating sphere (Everfine Co., Huangzhou, China). The integral duration is set as the constant of 100 ms. The white LED was located on the copper support. In the measurement, the increase in the junction temperature was less than 2 °C and could be ignored. Three-dimensional FDTD simulations were carried out to study the effect of APSs on the light extraction using Lumerical software (FDTD Solutions v8.21, Vancouver, BC, Canada) [35]. However, FDTD simulation dealt with light propagation in a small area due to the computation capacity and cannot work well in the phosphor layer. So, the Monte Carlo ray-tracing method was combined with an asymmetric bidirectional scattering distribution function (BSDF) [36]. After obtaining a BSDF for certain APSs using FDTD simulation, the corresponding APS surface was defined by the BSDF in a ray-tracing simulation.

3. Results and Discussion

Figure 2 shows the EL results of white LEDs at 5000 K with and without APSs mounting. The nominal CCTs of the virginal white LEDs are 5000 K. The APSs include APS-1,

APS-2, and APS-3, which correspond to the patterns of AAO400, AAO 300, and AAO200, respectively, in [29]. The periods of the APSs are about 450 nm. Figure 2a shows that all the integral intensities of blue emissions increase, while the ones of yellow emissions decrease with the APSs mounted. The white LEDs with APS-1 show more yellow emission reductions and more blue emission increase than those with APS-2 and APS-3. It is found that the spectral shapes are also modified by APSs. The EL intensities at the short-wavelength side of the yellow emissions are higher than the virginal ones. Figure 2b,c show the LOPs and LEs ratios to the virgins with different currents. It can be seen that the LOPs of the white LED with APS-2 are enhanced about 0.5% and the ones with APS-3 are similar to the virginal ones. However, the white LED with APS-1 shows that its LOP reduces significantly. Additionally, the LOP ratios increase with current monotonously. In Figure 2c, all the LEs of white LEDs decrease when the APSs are mounted. The white LEDs with APS-2 and APS-3 show about a 1% reduction in LEs, while APS-1 shows a more than 3% reduction.

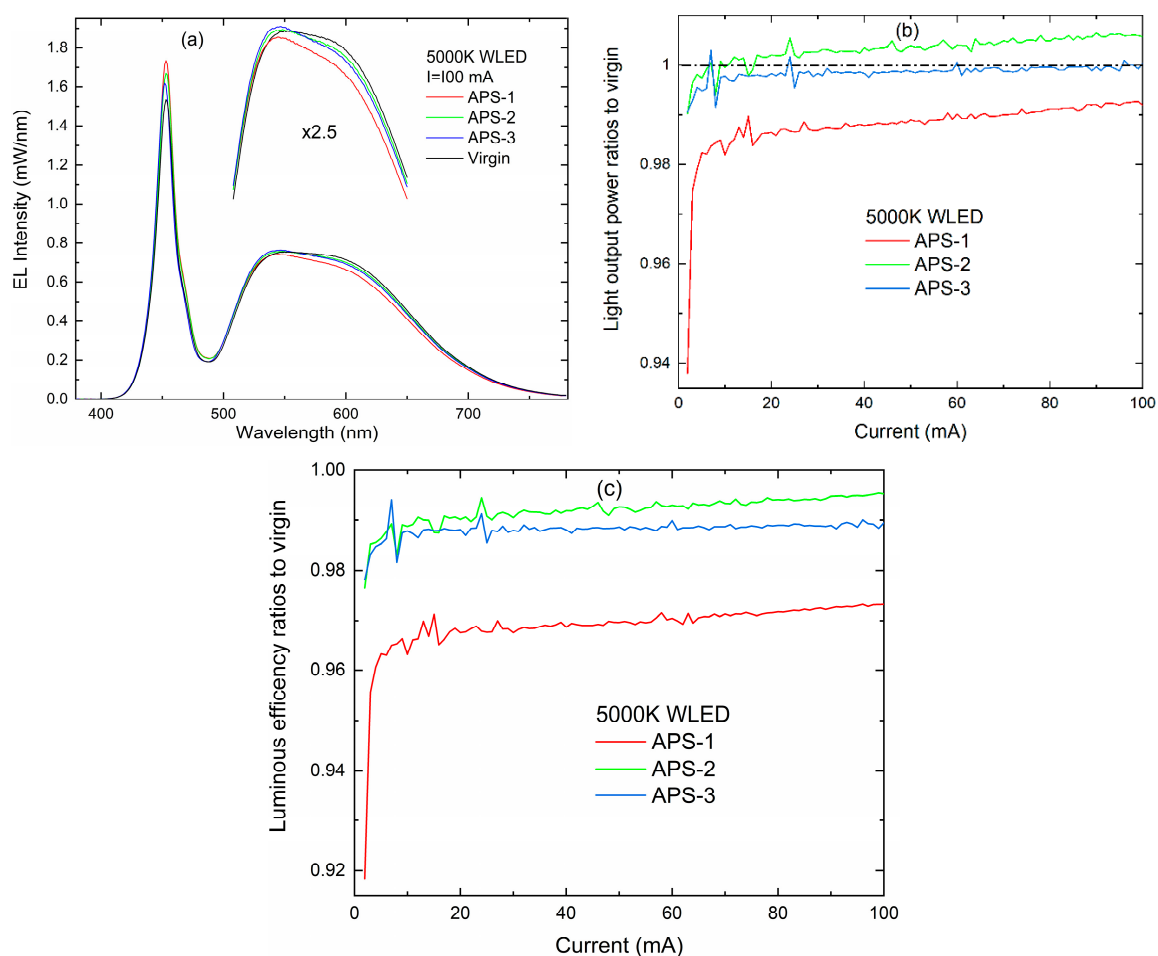


Figure 2. EL results of white LEDs of 5000 K mounted with and without APSs. (a) EL spectra at current of 100 mA. Current dependencies of (b) light output power ratios to virgin and (c) luminous efficiency ratios to virgin for different APSs. The periods of the APSs are about 450 nm. In the inset of (a), four magnified segments are shown as 2.5 times of those in the original ones.

The blue emissions are enhanced by APSs mounting, which is due to the Fourier power spectra of APSs within a certain spatial frequency, as described in our previous work [29]. As for the decreasing yellow emissions, it may be due to the quenching of yellow emission by APSs or the less blue emissions to excite the yellow phosphor. Using the multistage FDTD simulations [29], the LEE enhancements of the monochromatic LEDs with wavelengths of 450, 525, and 580 nm were calculated as 3.7%, 1.2%, and 1.6% by APS-1, respectively. So, the yellow emissions decreasing is caused by less blue emission

excitation. It also indicates APS-1 may be a candidate nanostructure for broadband LEE enhancement. The LEE enhancements of blue LEDs mounted with APS-1, APS-2, and APS-3 are 3.0%, 2.3%, and 1.3%, respectively [29], which demonstrates the reduction in yellow emission. As for the increasing yellow emission at the short-wavelength side, it may be due to the smaller hole diameters of APS-2 and APS-3 than those of APS-1. Although the blue emission excitation for yellow phosphor reduces with APS-2 and APS-3, the more yellow emission at short-wavelength side is extracted from the package. The enhancement of blue emission and part yellow emission lead to the LOPs with APS-2 and APS-3 not decreasing, as shown in Figure 2b. However, because the photopic vision function of blue light is much lower than that of the yellow one, all the LEs in Figure 2c show the reduction of 1–3%. With the current increasing, the CCT increases monotonously (not shown here), which means an increase in the blue emission content in the spectrum of white LED. For the virginal LED and LEDs with APSs, the increasing current would result in the droop effects on LOPs and LEs. However, considering the ratios to the virginal LEDs, the ratios of LOPs and LEs increase monotonously because there are more blue emissions with the increasing current and the APSs would further enhance the blue emission.

The EL measurements were also carried out on the white LEDs at 5000 K with APS-4 and APS-5 and 6000 K with APS-1. The LEs, LOPs, and CCTs of white LEDs with and without different APSs at the current of 100 mA are obtained and calculated, as listed in Table 1. All the LEs of white LEDs decrease when the APSs are mounted. The low CCTs of white LEDs with suitable APS seemly correspond to small LE variations. The LOPs of white LEDs with APSs change little compared to the flat ones. With APS-2 and APS-3, they show the LOP enhanced or unchanged compared to the virginal LEDs. The CCT increases when the APSs are mounted. APS-1 causes more CCT increase than APS-2 and APS-3. It is also seen that the nominal 6000 K white LED with APS-1 shows more CCT increase than the 5000 K one with the same APS. Combined with LEs and LOPs results, the blue emission increase seems to overlap the yellow emission decrease in LOPs, which agrees with the early report when the color conversion efficiency is high [38]. It is reasonable that the CCT increases with APSs because the blue emission contents increase in the white LEDs' spectra. Similarly, the smaller loss of yellow emission LOP will cause the smaller CCT increase. Lower nominal CCTs correlate to the smaller blue emission content in the white LED spectrum, which means a smaller effect of APS-1 on LE and CCT. It can be supposed that the smaller blue emission contents and appropriate APSs would enhance the LEs and decrease the CCT increment after APS is mounted on the white LED packages.

Table 1. LEs, LOPs, and CCTs of white LEDs with and without different APSs at the current of 100 mA.

Structures		LE (lm/W)	LOP (mW)	CCT (K)
APS-1	without	153.66	154.1	6304
	with	148.06	153.0	7354
	without	164.74	158.5	4940
	with	160.08	157.2	5615
APS-2	without	165.47	159.6	4823
	with	165.07	160.5	5240
APS-3	without	167.84	158.4	4779
	with	166.12	158.4	5198
APS-4	without	166.40	158.7	4712
	with	163.55	155.9	4942
APS-5	without	164.54	157.4	4820
	with	160.62	154.4	5119

Figure 3 shows the EL spectra of white LEDs with and without APSs at CCTs of 5000 K. The periods of the APSs are 800 nm. The hole diameters are 600 and 700 nm for APS-4 and APS-5, respectively. The operating current is 100 mA. Compared to the EL spectra in Figure 2, the peak intensities of blue emissions increase insignificantly, while the ones of yellow emissions decrease similarly to with APS-2 and APS-3 after the APSs are mounted. So, both LOPs and LEs decrease for the white LEDs with APS-4 and APS-5. The CCTs also increase, whose changes are lower than 300 K. It indicates that the APSs with 800 nm period cannot enhance the blue light extraction significantly. The small blue light extraction enhancement would not cause significant yellow emission reduction. The yellow emission reduction may be due to the destructive interference. The non-iridescent structural colors in an APS come as a result of coherent scattering [34], while the other wavelength lights may not be extracted from the semiconductors due to the destructive interference. As for the small CCT changes of white LEDs by APS-4 and APS-5, it is due to the very small blue emission increase. If the APSs cause the yellow emission coherent scattering and the blue emission decreasing, the LOPs and LEs will be enhanced.

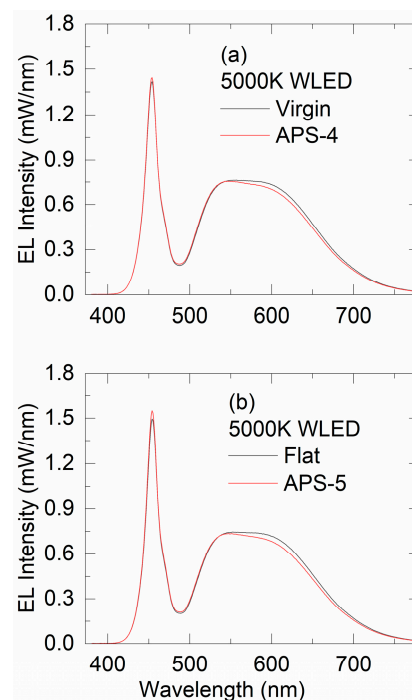


Figure 3. EL spectra of white LEDs mounted with and without (a) APS-4 and (b) APS-5 at CCTs of 5000 K. The periods of the APSs are about 800 nm.

The commercial white LEDs at the nominal CCTs of 5000 and 6000 K show a standard deviation of 200 K. So, ten typical white LEDs with APSs and their virgins are chosen to inspect the effects of the APSs on luminous performances. Their LEs and CCTs are plotted in Figure 4. It is obvious that all the white LEDs with APSs show lower LEs and higher CCTs than their virgins. With the CCT increase from 5000 K, the LEs seem to decrease monotonously. The record LE of white LEDs is 303 lm/W at about 5000 K [1]. More yellow emissions show higher LEs of white LEDs at the same LOPs. However, the red phosphor is less efficient than the yellow one, so the LEs will decrease when the CCT is lower than about 4500 K. Figure 4 shows that, at the same CCTs, many white LEDs show higher LEs with APSs than those without APSs, such as red points 2 and 4. The red points 9 and 10 also show higher LEs if the CCTs of the virginal white LEDs extrapolate to 8500 K. According to the linear fitting results, when the CCT is higher than 6500 K, the white LEDs with these APSs will show LE enhancements to the virgins at the same CCT. It indicates that the blue light emission enhancement by APSs can benefit the LE increases in white LEDs at the same CCTs. However, the absolute LE increases in white LEDs are impossible with the

above APSs because the CCT increases compared to the virgins. The experimental data points fluctuate along the fitting lines, which indicates the different effects of APSs and phosphor contents on the blue and yellow emissions. More blue and yellow emissions than the virginal ones originate from the emission enhancement by APSs. The yellow emission coherent scattering APSs may be concerned in light extraction.

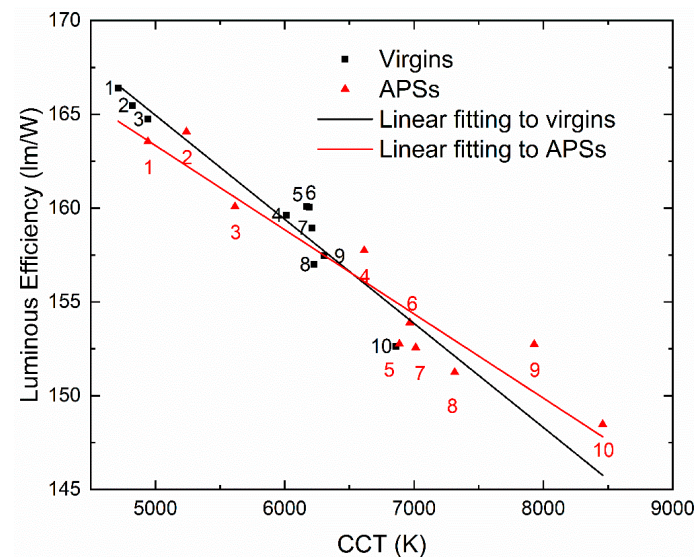


Figure 4. Relationship between luminous efficiencies and CCTs for white LEDs with and without APSs. The data are chosen with APS-1, APS-2, APS-3, APS-4, and APS-5 at the nominal CCTs of 5000 and 6000 K. The same numbers of black and red data points correspond to the same white LEDs with and without APSs. The fitting lines show same color as the experimental data.

In general, the LEs can be improved with appropriate APSs and corresponding virginal white LED spectra. At the same CCTs, the white LEDs with APSs can be easily achieved with higher LEs than those without APSs. However, the absolute enhancement of LE does not compare the same white LEDs with and without APSs. Here, we carried out the FDTD and ray tracing simulations to explore the LEs enhancement method. Because FDTD cannot simulate the phosphor effects and ray tracing cannot deal with the nanostructures [35,36], the BSDF of APS-1 was calculated by FDTD, which was then used in a ray tracing simulation. Figure 5a shows the schematic diagram of FDTD model according to our previous study [29]. The light source emitted the plane wave and it propagated from IPS through APS and then into the air. The refractive index values of IPS and air were set as 1.41 and 1.00, respectively. The lateral dimensions of the computational domain were set as $6\ \mu\text{m} \times 6\ \mu\text{m}$, considering the limitation of computer memory and computation capacity. The boundary condition in the top simulation area was set as a perfectly matched layer boundary condition and the four lateral boundaries were set as periodic boundary conditions. Figure 5b presents the LED model in the ray tracing simulator. The blue rectangular represents the LED chip and the yellow layer above represents the phosphor. In the ray tracing simulator, we figured out the emission of phosphor from the SPD of white LED without APS. In Figure 5c, the EL spectra of white LEDs are simulated, where the virginal and APS-1 white LEDs are simulated according to the experimental ones. The percentages of 100%, 90%, 80%, and 60% correspond to the blue emission transmission ratios to the real one with APS-1 by FDTD. The yellow emission transmissions remain unchanged. Then, the modified BSDFs are transferred to a ray tracing simulator. The blue emission reductions using nanostructures cause significantly yellow emission enhancements. It is observed that the simulated EL spectra of virginal and APS-1 (100% transmission) white LEDs are similar to the experimental ones. With the whole effects of APS-1, the LE decreases about 4% and the CCT increases from 6336 to 7589 K compared to the virginal ones. These results also agree with the experiment ones, whose LE decreases about 3.6% and the CCT increases

from 6304 to 7354 K, as listed in Table 1. It means the FDTD combined with ray tracing with the BSDFs is suitable for the white LEDs with and without APSs.

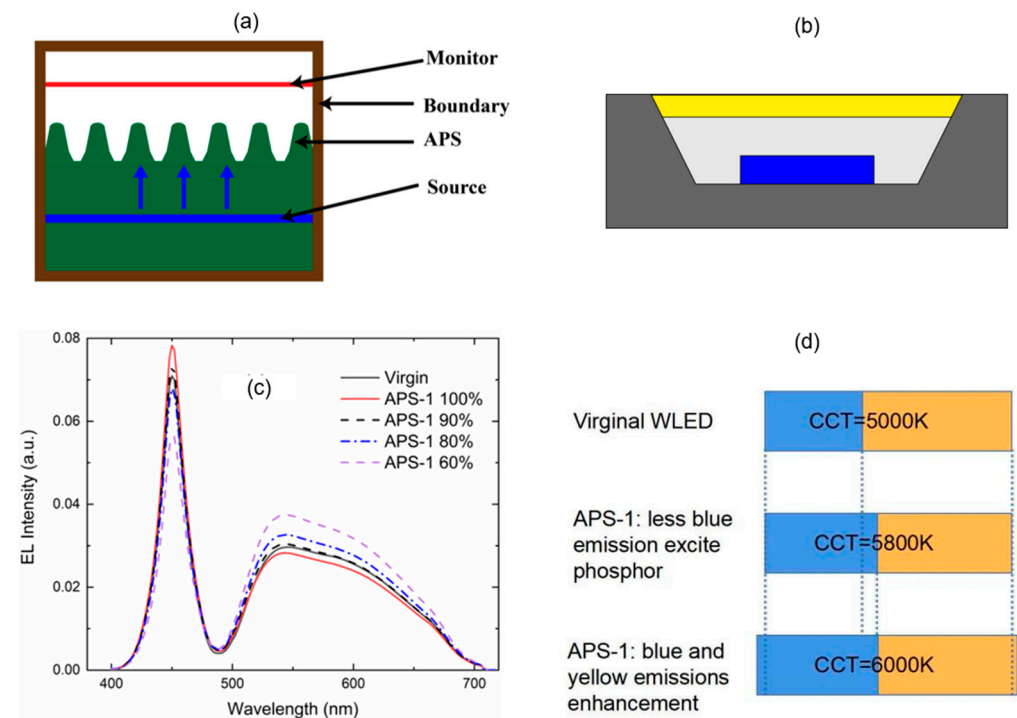


Figure 5. (a) The schematic diagram of white LED with APS in the FDTD simulation. (b) The white LED model in the ray tracing simulator. (c) Simulated EL spectra of white LEDs with APS with different blue emission transmission ratios. (d) The diagrammatic sketch of LE enhancement of white LED by APS-1 mounted.

APS-1 90% means a 10% reduction in blue emissions transmission of APS-1, which leads to a small increase in blue and yellow emission LOPs compared to the flat ones, as shown in Figure 5c. With a further decrease in blue emissions transmission for APS-1 80%, blue emission LOP reduces a bit and the yellow one increases significantly. As for APS-1 60%, both blue emission reduction and yellow emission enhancement are significant. The LEs of APS-1 80% and APS-1 60% samples are 10% and 25% enhancements compared to the virginal ones. Moreover, the CCTs decrease to 5868 and 4978 K, respectively. The absolute LEs enhancement requires more blue emissions to excite the yellow phosphor instead of light extraction from APS-1 directly. It is notable that the blue emission transmission reductions are set artificially, which is also difficult to find such APSs in practice. However, it can be found that the new APSs should change its period and size to decrease the blue emission transmission.

At last, the LE enhancement with APSs at the same CCTs can be discussed by a diagrammatic sketch, as shown in Figure 5d. It is supposed that the blue emission is 40% of LOP for 6000 K white LED, while 30% for 5000 K white LED. If their virginal LOPs are same, the virginal LE for 5000 K white LED is higher than that for 6000 K when APS-1 is mounted to the 5000 K LED package, more blue emission will be extracted directly. Less emission blue excitation led to yellow emission reduction. In the semiconductor, APS-1 reassigns the blue emission to extraction and excitation for phosphor. The total LOP does not change in the semiconductor with APS-1. Because the blue emission increases, the CCT will increase, which is supposed as 5800 K. Furthermore, the blue and yellow extracted emissions to air will be enhanced by APS-1. As mentioned above, the blue emission enhanced more than the yellow one as the CCT increased to 6000 K. Both the LOP and LE increase compared to those of the virginal white LEDs at 6000 K.

With the phosphor concentration increasing further, the scattering in the phosphor is more significant. So, the blue emissions reaching the surfaces of light extraction become less. The luminous flux will be enhanced absolutely compared to the virginal one, which is similar to the small transmission ratios of the blue emissions through APS-1 in Figure 5c. However, the quenching effect will appear when the phosphor concentration becomes high, because the self-absorption of phosphor and back scattering to chip will be significant. In the further research, the higher CCT white LEDs will be tried by sophisticated phosphor assembly. On the other hand, the yellow emission coherent scattering APSs may be hopeful to enhance the LEs for white LEDs. The broadband transmission of most yellow emissions and some blue emissions will be better for the absolute LE enhancements. The effective APSs need more design and experiments.

4. Conclusions

In summary, the APSs with different periods and hole diameters were fabricated and surface mounted on the white LEDs at nominal CCTs of 5000 and 6000 K. The EL measurements on these white LEDs were performed. All the white LEDs with APSs show less LE and higher CCT than the ones of the virginal white LEDs. However, the LEs of many APS-mounted white LEDs enhanced compared to the white LEDs without APSs at the same CCTs. Some LOPs of the white LEDs are enhanced by APSs surface mounted. At last, FDTD combined with Monte Carlo ray tracing simulations were carried out on the APSs surface-mounted white LEDs using BSDF transferring. The simulation results of EL spectra, LEs, and CCTs conform well to the experimental ones. The simulations on the effects of the blue emission transmission and phosphor concentration predict that the absolute LE enhancement for white LEDs requires high phosphor concentrations for the existing APSs beneficial to blue light extraction or the optimal APSs for yellow emission coherent scattering. The APSs with the structural color are more hopeful for high LE general lighting.

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