CCT- and CRI-tuning of white light-emitting diodes using three-dimensional non-close-packed colloidal photonic crystals with photonic stop-bands

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Abstract: This study exhibited the correlated color temperature (CCT)- and color-rendering index (CRI)-tuning behavior of light emission from white light-emitting diodes (WLEDs) using three-dimensional non-close-packed (3D NCP) colloidal photonic crystals (CPhCs). The CCT of approximately 5300 K (characteristic of cold WLEDs) of white light propagated through the NCP CPhCs dropped to 3000 K (characteristic of warm WLEDs) because of the photonic stop-bands based on the photonic band structures of NCP CPhCs. This study successfully developed a novel technique that introduces lower-cost CCT- and CRI-tuning cold WLEDs with a CRI of over 90 that of warm WLEDs by using 3D NCP CPhCs.

References and links

1. Introduction

White light-emitting diodes (WLEDs) have attracted considerable attention because of their brightness, reliability, low electrical consumption, and long lifetime [1]. The use of WLEDs for general illumination has recently grown rapidly. The color of white light sources is usually evaluated by two optical properties: the correlated color temperature (CCT) describes how colors are falling close to the blackbody curve, and the color-rendering index (CRI) describes how well colors are reproduced by the light source. For general illumination, incandescent bulbs with a CCT of 3000 K have a yellow appearance and are described as warm light sources, whereas fluorescent bulbs have a CCT of 5500 K and are described as cold light sources. Several approaches for fabricating WLEDs have been developed [2,3]. Cold-WLEDs (c-WLEDs, CCT of approximately 5000–5500 K) are ideal for general illumination, and warm-WLEDs (w-WLEDs, CCT of approximately 2700–3000 K) create a relaxing atmosphere and a feeling of wellbeing for indoor illumination [4]. Commercially available WLEDs widely use a GaN-based blue LED to pump yellow phosphor (cerium-doped yttrium-aluminum-garnet, YAG:Ce). This is the most efficient method because YAG:Ce WLEDs demonstrate high luminous efficiency at low cost [5]. The YAG:Ce WLEDs have other advantages, such as ease of fabrication, low cost, and a high phosphor conversion efficiency of over 70%. However, some problems remain with YAG:Ce WLEDs, such as high CCT, low CRI, and the absence of a red spectrum [6]. WLEDs must have a broadband spectrum to achieve a CRI of over 80, which is required for use in general illumination applications. Therefore, phosphors of two or more colors are required. An excellent CRI of over 90 has been achieved by WLEDs with a larger weight percent (wt%) concentration of red-emitting phosphor, or by multicolored WLEDs [7]. However, nitride- and borate-based red-emitting phosphors cost 10 times more per gram than yellow-emitting phosphors [8]. WLED lighting systems are as bright as traditional incandescent bulbs. Many people still do not appreciate that the higher initial outlay for WLED bulbs yields long-term energy and cost savings. Expanding the WLED lighting market is made difficult by the high cost of WLEDs.

This study develops a novel technique for fabricating low cost w-WLEDs. Three-dimensional (3D) non-close-packed (NCP) colloidal photonic crystals (CPhCs) using polystyrene (PS) nanospheres deposited onto the c-WLEDs are CCT- and CRI-tuning because of the photon stop-bands of 3D NCP CPhC nanostructures. The 3D PhC nanostructures are able to confine and control photons along designated channels known as photonic band gaps [9]. Among the 3D CPhC structures, face-centered-cubic (fcc)-based opals and inverse opals of CPhCs are of great interest because they can be produced with methods based on colloidal solutions [10]. This study used the 3D NCP colloidal crystals. These crystals have attracted extensive interest in recent years because of their fundamental nature and technological importance [11]. Compared to close-packed (CP) CPhCs, the NCP CPhCs have a tunable inter-nanosphere gap and wider photonic band gaps, making them attractive materials for photonic devices [12]. To date, the deposition of 3D NCP CPhCs onto c-WLEDs to obtain w-WLEDs has not been explored. In c-WLEDs with 3D NCP CPhCs, the white light emission spectrum significantly modified based on the photonic stop-bands and phosphor reemission, which cause the CCT of the c-WLEDs, of approximately 5500 K, to drop to the CCT of w-WLEDs, of approximately 3000 K, without increasing the concentration of yellow- and red-emitting phosphor. Lowering the cost of w-WLEDs does not only hasten the transition from conventional bulbs to WLEDs, but also leads to new applications for optical devices.
2. Experimental

The c-WLEDs and w-WLEDs with CCTs of approximately 5500 K and 3000 K, respectively, were fabricated with GaN-based blue LED chips and varying amounts of yellow and red phosphor mixed with a silicone encapsulant. Figure 1(a) shows the 3D NCP CPhCs deposited onto the c-WLEDs. The c-WLEDs and w-WLEDs were made as follows: First, we bonded a 22 mil × 35 mil chip of GaN-based pattern-sapphire substrate (PSS) LEDs (emission wavelength 455 nm) into a commercial plastic lead frame package (5630 package) with silver paste and gold wire. The layout of the GaN-based PSS LED chip is shown in Fig. 1(a). Second, we used Y₃Al₅O₁₂:Ce³⁺ (yellow-emitting) and Ba₂Si₅N₈:Eu²⁺ (red-emitting) phosphors to obtain the c-WLEDs and w-WLEDs. The yellow and red phosphors (of various wt% concentrations) were uniformly mixed with the silicone, and then filled into the leadframe by using traditional phosphor-dispensing techniques. The WLEDs that were denoted as reference c-WLEDs showed a CCT of 5283 K, and those denoted as reference w-WLEDs showed a CCT of 3130 K, at a driving current of 120 mA. The c-WLEDs had a yellow and red phosphor concentration of 16.8 wt% and 1.2 wt%, respectively, and the w-WLEDs had a yellow and red phosphor concentration of 21.0 wt% and 4.0 wt%, respectively. The yellow- and red-emitting phosphors were used in the WLEDs to obtain a broadband spectrum with a CRI of over 80. In this experiment, increasing the concentration of the yellow phosphor in the c-WLEDs by 1.25 times and the red phosphor concentration by 3.33 times caused the high CCT of the c-WLEDs to drop to the low CCT of the w-WLEDs.

Fig. 1. (a) Schematic diagram of the 3D NCP CPhCs deposited onto the 5630 WLED package. The inset shows the OM image of c-WLEDs with 3D NCP CPhCs. (b) The fcc Brillouin zone with symmetry points. The z-axis is along the [111] direction.

Then, we prepared two sizes of the NCP PS nanosphere to get the high CRI, with diameters (D) of 225 nm and 250 nm. The 10-µL NCP CPhCs were deposited onto the entire emission region of the c-WLEDs with a micro-pipette. 3D NCP CPhCs were deposited onto the c-WLEDs with NCP CPhCs of a lattice constant (a) of 349 nm and 385 nm. The optimal NCP CPhC regions were approximately 17-µm-thick to obtain the CRI of over 80 and CCT about 3000 K, as determined using a field-emission scanning electron microscope (FESEM). This study used the FESEM to study the crystalline structure of the resulting NCP CPhCs on the c-WLEDs. 3D NCP CPhCs with fcc structures were grown by natural sedimentation, with the surface parallel to the (111) crystallographic plane. Figures 2(a) and 2(b) show high-magnification FESEM images of the NCP CPhCs fabricated using PS nanospheres with D of 225 nm and 250 nm, respectively. Figure 2(c) shows low-magnification FESEM images of the NCP CPhCs with a D of 225 nm. These images indicate that NCP CPhCs can be fabricated in a relatively large domain without cracks. In addition, the structure of the NCP CPhC consists of an fcc array interconnected by narrow tubular necks [11], as shown in the inset in Fig. 2(c). Figures 2(a) to 2(c) clearly show the NCP CPhC structure in which the
nanospheres are no longer in contact, but are joined to their nearest neighbors in a cross-section by tubular necks. A closer view of the NCP CPHCs shows that the PS nanospheres are arranged with a non-uniform inter-nanosphere gap (x). The tubular neck length and uniformity inter-nanosphere gap of 3D NCP CPHCs could be controlled by solvent species and solvent concentration.

![FESEM images of the 3D NCP CPHCs](image)

**Fig. 2.** FESEM images of the 3D NCP CPHCs prepared using PS nanospheres with D of (a) 225 nm, and (b) 250 nm. (c) The FESEM surface images of the NCP CPHCs showing that the nanospheres stack with a well-organized (111) plane and fcc structures. The first Brillouin zone with the symmetry points indicated. Inset: cross-section of the NCP CPHCs into domains along the (110) plane. (d) Schematic showing the NCP CPHC of the fcc structure viewed along the (100) plane.

### 3. Results and discussion

This study measured the luminous flux, luminous efficiency, CRI, CCT, and CIE color chromaticity coordinates (x, y) versus current characteristics by using a 20 in. integration sphere from Labosphere with a radiometer and photometer (SC-610). Figure 3(a) shows that the luminous flux values of the c-WLEDs, the c-WLEDs with an NCP CPHC of a = 349 nm, the c-WLEDs with an NCP CPHC of a = 385 nm, and the w-WLEDs were 38.7, 34.7, 31.2, and 35.7 lm, respectively, and the luminous efficiencies were 100.8, 90.4, 81.3, and 93.0 lm/W, at a current of 120 mA and a voltage of 3.2 V. The luminous flux of the c-WLEDs with NCP CPHCs decreased by approximately 10% to 19% compared to the reference c-WLEDs. However, the CCT of these two types of WLEDs can drop from approximately 5500 to approximately 3000 K without increasing the concentration of yellow and red phosphor. In addition, the c-WLEDs with an NCP CPHC of a = 349 nm showed a luminous flux, CRI, and CCT almost equal to that of the reference w-WLEDs. Table 1 provides a list of the optical properties of the four types of WLEDs at a current of 120 mA. The c-WLEDs with an NCP CPHC of a = 385 nm shows the highest CRI value of over 90, which indicates that the light source would render the color of an object accurately. The CIE (x, y) coordinates of the four types of WLEDs fall in the white region of the CIE chromaticity diagram, and almost fall on the Planckian locus. This novel technique would reduce the overall cost of w-WLEDs.
Table 1. The optical characteristics of the four types of WLEDs.

<table>
<thead>
<tr>
<th>Devices</th>
<th>Luminous flux (lm)</th>
<th>Luminous Efficiency (lm/W)</th>
<th>CRI (Ra)</th>
<th>CCT (K)</th>
<th>CIE (x, y) coordinate</th>
</tr>
</thead>
<tbody>
<tr>
<td>c-WLEDs</td>
<td>38.7</td>
<td>100.8</td>
<td>85.2</td>
<td>5283</td>
<td>(0.3372, 0.3382)</td>
</tr>
<tr>
<td>c-WLEDs with NCP CPhCs of (a = 349) nm</td>
<td>34.7</td>
<td>90.4</td>
<td>82.2</td>
<td>3111</td>
<td>(0.4306, 0.4016)</td>
</tr>
<tr>
<td>c-WLEDs with NCP CPhCs of (a = 385) nm</td>
<td>31.2</td>
<td>81.3</td>
<td>91.4</td>
<td>3076</td>
<td>(0.4311, 0.4013)</td>
</tr>
<tr>
<td>w-WLEDs</td>
<td>35.7</td>
<td>93.0</td>
<td>83.5</td>
<td>3130</td>
<td>(0.4281, 0.3992)</td>
</tr>
</tbody>
</table>

The optical properties of close-packed (CP) CPhCs have been studied by several research groups [13,14]. These studies have revealed a lower-order photonic stop-band at the \(L\) symmetry point of the fcc Brillouin zone along the [111] direction. The photonic stop-bands of 3D NCP CPhCs can be altered by controlling the size of the nanospheres [11]. This phenomenon was demonstrated using the reflection spectra measurements. The reflection spectra measurements of the NCP CPhCs were performed using a micro-reflectometer with a Y-type optical fiber of white light from a Xe lamp. The reflection spectra of NCP CPhCs at a normal incidence are the same in \(\theta_{\text{out}} = 0^\circ\) of Figs. 4(a) and 4(c). The reflection peak positions of NCP CPhCs at a normal incidence configuration are 550 and 605 nm, respectively, for NCP CPhCs with a \(D\) of 225 nm and 250 nm. The full width at half maximum (FWHM) of the reflection spectra was 51 and 39 nm, respectively. These results demonstrate that the NCP CPhCs have wider photonic stop-bands compared to the CP CPhCs [14]. There was an obvious redshift of the reflection peak position of the reflection spectra, which resulted in a variation of the lattice constant of the NCP CPhCs. The redshift of the reflection peak positions of the NCP CPhCs can be explained by Bragg’s law [11]. For the reflection spectra measurements, the direction of incident light was perpendicular to the (111) planes \(\theta_{111} = 0\). The reflection wavelength \(\lambda_R\) of Bragg’s law is thus given using Eq. (1):

\[
\lambda_R (D) = 2 d_{111} \sqrt{n_{\text{eff}}^2 - \sin^2 \theta_{111}}
\]  

where \(\lambda_R\) is the wavelength of the reflection peak, \(d_{111}\) is the inter-planar spacing between the (111) planes, and \(n_{\text{eff}}\) is the effective refractive index of the crystalline lattice. In the (111) surface of the fcc structure, the inter-planar spacing \(d_{111}\) is given using Eq. (2):
\[ d_{111} = \frac{a}{\sqrt{3}} = \frac{\sqrt{2}}{\sqrt{3}}(D + x) \] (2)

where \( a \) is the lattice constant of the NCP CPhCs, and \( x \) is the NCP inter-nanosphere gap. For the NCP CPhC structure, \( n_{\text{eff}} \) is given using Eq. (3) [11]:

\[ n_{\text{eff}} = n_{\text{ps}}^2 f_{\text{ps}} + n_{\text{air}}^2 (1-f_{\text{ps}}) \] (3)

where \( n_{\text{ps}} \) and \( n_{\text{air}} \) are the refractive indices of the PS nanospheres and air, respectively, and \( f_{\text{ps}} \) is the volume fraction of the nanospheres. For the NCP CPhC structure, \( f_{\text{ps}} \) is given by Eq. (4):

\[ f_{\text{ps}} = \frac{4V_{\text{sphere}}}{V_{\text{cubic}}} = \frac{4\sqrt{2}}{\sqrt{3}} \left[ \frac{D}{2(D + x)} \right]^3 \] (4)

where \( V_{\text{sphere}} \) and \( V_{\text{cubic}} \) are the volumes of the PS nanospheres, as indicated in Fig. 2(d). The wavelength of the reflection peak as a function of the diameter of the NCP CPhCs according to Eq. (1) to (4) is consistent with the measured reflection results, where the average inter-nanosphere gap \( x \) is 22 nm and \( n_{\text{ps}} = 1.59 \). In this experimental results, the luminous flux, CRI, CCT, and CIE (x, y) coordinates of the NCP CPhCs deposited onto the tunable WLEDs was affected significantly by photonic band structures that depend on the CPhCs lattice constant and CPhCs film thickness.

To confirm the light emission distribution from the \( c \)-WLEDs propagated through the NCP CPhCs. The luminescence spectra of the four types of WLEDs were measured by the 20 in. integration sphere at a current of 120 mA, as displayed in Fig. 3(b). The maximum emission peak positions of the \( c \)-WLEDs with differently NCP CPhCs are 605 nm, and 615 nm, respectively, due to the photonic stop-bands and phosphor reemission affect the luminescence spectrum. The light emission of the \( c \)-WLEDs through the NCP CPhCs was propagated according to the photonic band structures of the 3D NCP CPhCs. Measurement of the angular-resolved transmission and reflection spectra is a powerful method of investigating the guided modes of 3D CPhCs. This study measured the angular-resolved transmission spectra to study the light emission distribution of the \( c \)-WLEDs with NCP CPhCs under electrical current. In [15], Lai et al. have explained the apparatus for the angular-resolved transmission spectra measurement setup. A continuous current of 120 mA was applied to the \( c \)-WLEDs with NCP CPhC of \( a = 349 \) nm and 385 nm at room temperature. The angular-resolved transmission spectra as a function of detection angle \( \theta_{\text{out}} \) were obtained with a fiber probe coupled to an optical spectrometer, which was rotated from \( \theta_{\text{out}} = 0^\circ \) (normal direction) to \( \pm 90^\circ \) in increments of 0.5\(^\circ\), and the fiber-to-device distance was maintained at 25 cm. The measuring plane of the \( c \)-WLEDs with NCP CPhCs was fixed along the \( \Gamma-L-U \) path of the fcc Brillouin zone, as indicated in Fig. 1(b). The transmission spectra were then displayed on a wavelength versus detection angle plot, with the color representing the intensity according to a log scale bar. Figures 4(a) and 4(c) show the transmission spectra collected along the \( \Gamma-L-U \) directions in unpolarized light for the \( c \)-WLEDs with an NCP CPhC of \( a = 349 \) nm and 385 nm, respectively. The transmission measurement results showed that the photonic stop-bands for the guided modes shifted toward shorter wavelengths with increasing detection angles \( \theta_{\text{out}} \), which can be attributed to the photonic band structure of the 3D NCP CPhCs. Figures 4(a) and 4(c) demonstrate the first photonic pseudogap of the photonic band structures along the (111) surface, which appear along \( \Gamma-L \) at approximately \( a/\lambda = 0.65 \). The photonic stop-bands at low-energy values correspond to the \( c \)-WLEDs with an NCP CPhC of \( a = 349 \) nm and 385 nm. Additionally, due to the photonic stop-bands caused the light diffracted back into the phosphor layer of \( c \)-WLEDs, which could excite the phosphor again and induce additional fluorescence that will give an extra contribution to the output, as shown in Fig. 3(b). A full
theoretical model of the interaction between the two phosphors and the 3D NCP CPhCs are indeed very complex, that will carefully discussion on elsewhere.

The $c$-WLEDs with 3D NCP CPhCs exhibited photonic stop-bands in the zenithal ($\theta$) and azimuthal ($\phi$) direction that have measured and discuss in the context of the photonic band structures. Figures 4(a) and 4(c) can be represented as the photonic dispersion curves with $k_{//} = a \cdot \sin(\theta_{\text{out}})/\lambda$ for comparison with the calculated photonic band structures [15, 18], as shown in Figs. 4(b) and 4(d). Figures 4(b) and 4(d) also shows the photonic band structures for the infinite NCP CPhCs with fcc structures calculated by plane-wave expansion (PWE) method [16]. Experimental results exhibited a photonic stop-band that consisted of the lowest bands of the photonic band structure in the $L-U$ direction. In addition, the lower energy photonic stop-band appeared at $a/\lambda = 0.65$ in the normal direction, and shifted to higher energies with an increasing detection angle $\theta_{\text{out}}$. The photonic stop-band corresponded to the $L-U$ of the photonic band structures associated with Bragg diffraction by the (111) planes. We took the $F-L-U$ triangle as representative, and assumed that the incident light with wavevector $k$ was in that diffraction plane; with the tip of $k$ lying on the $L-U$ segment [Fig. 1(b)]. This permitted us to classify the bands according to their behavior under mirror reflection, as either symmetric (second and third, red dashed line) or asymmetric (first and fourth, blue continuous line) bands [17], as shown in Figs. 4(b) and 4(d). The azimuthal of the far-field emission distribution has measured as a function of the azimuthal angle $\phi$ by using the angular-resolved transmission-measuring apparatus [18]. In azimuthal measurement results also showed that the photonic stop-bands which attributed to the photonic band structure of the 3D NCP CPhCs, as seen in Fig. 4(a) (Media 1).

![Fig. 4.](image)

Fig. 4. (a) and (c) Unpolarized angular-resolved transmission measurements of the $c$-WLEDs with NCP CPhC of $a = 349$ nm (Media 1) and 385 nm. (b) and (d) The photonic dispersion curves determined from the data shown in (a) and (c) and compared with the photonic band structures. Inset: The photonic band structure of the infinite NCP CPhCs of the fcc structure calculated using the PWE method. With respect to mirror symmetry, the continuous blue lines represent asymmetric bands, and the dashed red lines represent symmetric bands.
The optical properties of the c-WLEDs with 3D NCP CPhCs, which possess wider photonic stop-bands along the main crystallographic directions, were noticeably different to the CP CPhCs. In addition, the photonic stop-bands of NCP CPhCs can be reflected the shorter wavelength of light emission from the c-WLEDs back into the phosphor layer that could excite the red and green phosphor, which caused the CCT of the c-WLEDs to drop to that of the w-WLEDs. A comparison of the angular-resolved transmission spectra and the corresponding photonic band structures shows good agreement between experiment and theory. This study fitted the lowest-order mode with the photonic band structures accurately. The consistency between experiment and theory was acceptable, although small differences were found in the width and intensity in larger detection angles, which is probably associated with the boundary conditions. In the c-WLEDs with 3D NCP CPhCs, the CCT, CRI, and CIE (x, y) coordinates could be significantly modified by the 3D NCP CPhC parameters, such as CPhC lattice constant and CPhC film thickness.

4. Conclusions
In conclusion, this study designed and fabricated c-WLEDs with 3D NCP CPhCs that had CCT, CRI, and CIE (x, y) coordinates that were tunable according to the photonic band structures of 3D NCP CPhCs. The c-WLEDs with an NCP CPhC of $a = 349$ nm shows a luminous flux, CRI, and CCT almost equal to that of the reference w-WLEDs, and with an NCP CPhC of $a = 385$ nm shows the highest CRI value of over 90. The angular-resolved transmission measurement showed the photonic stop-bands that attributed to the photonic band structures of the 3D NCP CPhCs. The c-WLEDs with NCP CPhCs caused the red light emission increase because of the photonic stop-bands and phosphor reemission, which lowered the c-WLED CCT of approximately 5500 K to the w-WLED CCT of approximately 3000 K, without increasing the concentration of yellow or red phosphor. The experimental transmission spectra agreed favorably with the simulated photonic band structures of the 3D NCP CPhCs. This novel technique applied to c-WLEDs would reduce the overall cost of w-WLEDs.

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