Manuscript Details

Manuscript number	LUMIN_2019_1968_R1
Title	Multi-Colour Light Emission based on Pixel-Array Phosphor Layer in LEDs
Article type	Research Paper

Abstract

Phosphor layers are of vital importance for the development of advanced phosphor-converted light-emitting diodes (LEDs). However, owing to the fixed ratio of red-green-blue (RGB) phosphors, it has been difficult for the RGB phosphor layers with conventional structure to tune light emission colours. Herein, we have experimentally fabricated nine types of phosphor layers with patterned RGB pixel array, which consist of tuneable RGB ratio in a planar configuration. Moreover, we carried out optical simulation based on Monte-Carlo theory to assist in adjusting the light-emission colours and the corresponding chromaticity coordinates. The simulations were further verified by the experimental results via samples fabricated by the stencil printing technique. In accordance with the nine types of phosphor layers with patterned pixel arrays in various RGB ratios, we have finally obtained corresponding nine light-emission colours for the applications of LED light emission decoration. These designed advanced pixel-array phosphor layers demonstrate great potential for applications in decorated light emission and display devices with significant implications for industrial improvement in these research areas.

Keywords	phosphor layer, stencil printing, multi-colour light emission, pixel array.
Manuscript category	LED phosphors+phosphors +scintillators/thermoluminescence/dosimetry/afterglow

Corresponding Author	Renli Fu
Corresponding Author's Institution	Nanjing University of Areonautics and Astronautics
Order of Authors	haitao zhu, Renli Fu, Yahui Shi, Qinjiang He, He Wang, Xuhai Liu
Suggested reviewers	Sheng-Yuan Chu, Xiaojuan Liang

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Research Data Related to this Submission

There are no linked research data sets for this submission. The following reason is given:
Data will be made available on request

Dear Editors,

We thank you and the reviewer very much for the positive feedback on our work (No.: LUMIN_2019_1968) as well as the decision on this manuscript. Please find attached our revised manuscript. Below, we have given detailed answers to each of the points raised by the reviewer and revised the manuscript accordingly with the changes highlighted in the text.

Although light-emitting diodes (LEDs) packaged with red–green–blue (RGB) phosphors are widely used in the white lighting industry, they are few methods to be used in multi-colours LEDs product, especially by modifying configuration of phosphor layer. In this article, we designed several configurations of phosphor layers to realize multi-colours emission by simulation and experiment. The experiment results including output light colours and CIE coordinate were compared with the simulated ones and showed that these samples can indeed realize multi-colours emissions.

We sincerely hope that this revision can provide proper corrections to ensure an acceptance by Journal of Luminescence. We are looking forward to your reply!

Best regards

Prof. RenliFu

College of Materials Science and Technology,

Nanjing University of Aeronautics and Astronautics

Response to Reviewer:

1. For the optical simulations, more details should be provided, such as the numerical model, optical properties and light efficiency of each phosphor pixel and layer.

[Response]: We thank the reviewer for this helpful suggestion, and accordingly we have added the following discussion on Page 6 and Page 7 in the revised manuscript regarding the numerical model:

"In this simulation model,

………..

Similarly, the output energy of green and red light can be expressed as:

$$W_G = q_G(A_{G-emission} + A_{G-B}) - A_{R-G} \quad (5)$$

$$W_R = q_R(A_{R-emission} + A_{R-B} + A_{R-G}) \quad (6)$$

"

Regarding the optical properties and light efficiency of each phosphor pixel and layer, we have modified Table 1 as follows:

Tab.1 Parameters applied in the optical simulation [22].

Component	Dimension	Parameter
Chip	1×1×0.1 mm	1 W, 360 nm, ultraviolet chip
Blue phosphor	Pixel size:1.0×1.0×0.1 mm, square, total: 4.0 mm. H 0.1 mm,	S:11.5, g:0.9, ε_{uv}:0.7, n:1.6, q_B:0.8, c:200 mol/L
Green phosphor		S:16.5, g:0.89, ε_{uv}:0.7, ε_B:0.6, n:1.6, q_G:0.9, c:200 mol/L
Red phosphor		S:16.5, g:0.89, ε_{uv}:0.3, ε_B:0.3, ε_G:0.3, n:1.6, q_R:0.9, c:200 mol/L
Transparent substrate	R:3.0, H:0.1 mm	PMMA, Transparent, no absorption
Reflective cup	R:3.0, H:1.0 mm	Perfect mirror reflection

* Abbreviations in Table 1: *H*- thickness, *R*- radius, *S*- Scattering coefficient, *n*- refractive index

It should be noted that we have replaced the symbol of absorption coefficient 'a' with 'ε'. Moreover, the quantum conversion efficiency and concentration of RGB phosphors used in the simulation have been added in this table. Additionally, it would be better to use the quantum conversion efficiency rather than light efficiency to characterize phosphors property, because light (luminous) efficiency (lm/W) is mainly used for white LED, which is not suitable for multi-colours emission of phosphor layers.

2. For experiments, more details should be provided, such as the concentration of the phosphor pixel and layer, the CCT and light efficiency of the LED samples.

[Response]: The reviewer is correct in pointing out that more details should be provided. As for each pixel, the ratio of the organic glue and the phosphor was set to be 25%:75%. Afterwards, the phosphor paste was printed on PMMA to form phosphor pixel array layer by screen printing method, where the ratio of RGB phosphors in the total amount of pixels are in accordance with the pixel-array configurations (as shown in Fig. 1). Additionally, the ratio of RGB phosphors in the designed pixel-array configurations should be 1:0:0, 0:1:0, ..., 1:0:3, 0:1:3 for pattern1-15.

The CCT of the WLEDs can be calculated by the equation (in CIE 1931xy system) as follow:

$$CCT = 437 \times \left(\frac{x - 0.332}{0.1858 - y}\right)^3 + 3601 \times \left(\frac{x - 0.332}{0.1858 - y}\right)^2 + 6861 \times \left(\frac{x - 0.332}{0.1858 - y}\right)^1 + 5517$$

where (x, y) is the correlated colour coordinate in CIE 1931xy system. CCT and luminous efficiency are of vital importance in characterizing the white LED lighting. However, regarding some light, *e.g.*, 450 nm pure blue light and 660 nm pure red light, we could not get the CCT value when the wavelength of a specific light exceeds the white lighting wavelength range.

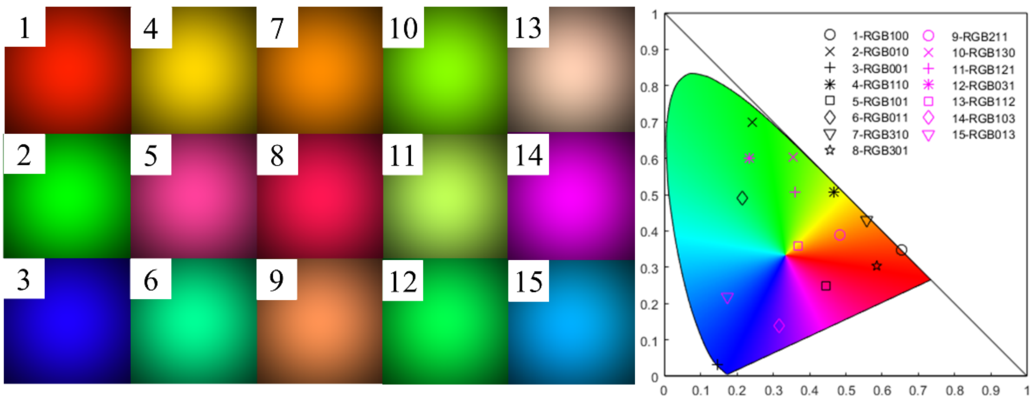
Luminous efficiency is an essential criterion for white LEDs but it has limited meanings here for multi-colour LEDs. The definition of the luminous efficiency is ‘lumens of optical power output per watt of electric power input (lm/W). Lumen is a value related to the spectrum sensitivity of human eyes. Although several articles use luminous efficiency with other units (cd/W, %, etc.) to measure the property of common light colours (blue, yellow, etc.) as well [1-4], unfortunately there is no other luminous efficiency of corresponding light colours as we did to compare. Additionally, luminous efficiency is usually used to measure packaged LED product rather than a single phosphor layer.

1. Jiang, Yang, et al. "Realization of high-luminous-efficiency InGaN light-emitting diodes in the “green gap” range." Scientific reports 5 (2015): 10883.

2. Liang, Junfei, et al. "Improved efficiency of blue polymer light-emitting diodes using a hole transport material." Journal of Materials Chemistry C 5.21 (2017): 5096-5101.
3. Huang, Fei, et al. "High-Efficiency and Color Stable Blue-Light-Emitting Polymers and Devices." Advanced Functional Materials 17.18 (2007): 3808-3815.
4. Xu, X. J., et al. "High-Efficiency Blue Light-Emitting Diodes Based on a Polyphenylphenyl Compound with Strong Electron-Accepting Groups." Advanced Materials 19.9 (2007): 1281-1285.

3. Please exam the light colour shown in Figs. 1, 2 and 4, especially the type 13-15.

[Response]: After carefully checking the light colours presented in Fig.1, 2 and 4, we found the colours of type 13-15 were much different mainly because we made a wrong sequence of type 14 and 15 in Fig.2. Hereby we have corrected it in the revised manuscript.



(a) (b)

Fig.2 (a) Simulated emission colour and (b) corresponding CIE of the pixel-array phosphor layers.

Other reasons responsible for the slight colour difference are as follows:

(1) Colours in Fig.1 were mixed based on colour mixing principal, which do not have light absorption.

(2) Colours in Fig. 2 obtained by simulation took light absorption into consideration, but at this step the shape of phosphor pixels keeps regularly cubic

(3) Colours in Fig. 4 obtained by experimental samples existed behaviour of light absorption and phenomenon of irregular cubic shape (and blank regions).

Where the reasons (1) and (2) can explain why colour of type 13 looks different in Fig.1 and 2, that the designed one is light blue but the simulated one is light orange. To be specific, the ratio of RGB phosphors pixels in type 13 is 1:1:2, which responses for the blue light emission in designing process. However, based on CIE diagram in Fig.2 (b), simulated light colours of type 13 (the purple square) will move from blue region (colour in designed process) to yellow region due to the re-absorption of blue light, leading to the colour difference between designed one and simulated one.

Other colours differences in Fig.1, 2 and 4 are also mainly caused by the three reasons.

4. The phosphor configuration shown in Fig. 4(a) is much different from that shown in Fig. 1, please add more description.

[Response]: The phosphor configuration in Fig. 1 (left-bottom) was set in powerpoint, where the red, green and blue square represent red, green and blue phosphor pixels, respectively, and all pixels are regular cubic structure. In Fig. 4, the orange region represents red phosphor pixels, light green region represents green phosphor pixels, and white one is blue phosphor pixel. Because of manual operation and the viscosity of the phosphor paste, pixels are not regular cubic (such as pattern 9, 14), and there are some blank areas on the substrate (such as pattern7, 15). These factors make them visually not quite similar with each other in Fig.1 and Fig. 4(a). On page 9 in the revised manuscript, we have added detailed description about the configuration in Fig.1 and Fig.4.

'There are 400 pixels for each pixel-array phosphor layer (20 mm×20 mm) of the sample, where the orange, light green and white region (visually light green and white colours might be difficult to distinguish in pattern13) represent red, green and blue phosphor pixels, respectively. These positions correspond to that in Fig.1 (red, green and blue square), although there are still slight differences on cubic shape (pattern9 and 14) and blank area (pattern7 and 15), which is difficult to avoid in manual operation.'

5. From Fig. 4, it can be seen the light colour obtained from experiments and simulations are not so similar in some types of phosphor layer, such as types 9, 11 and 13, please add more results and discussions to make the results more credible.

[Response]: We totally agree with the reviewer that the light colours obtained seem not so similar in some types of phosphor layer. The experiment light colours of type 9, 11 and 13 appear to be white colour, but not pure white light ((0.33, 0.33) in CIE 1931xy). The white colours might be a little orange or green, which can be seen from the margin area of the experimental light colour (seen in the first figure). These colours correspond to the simulation result. Additionally, because there is no environmental light assisted in simulation, the light colours in simulation and experiment can be somehow distinctive visually. However, according to the chromaticity coordinate, the difference between simulation and experiment results is not severe. We have added explanations about the difference on Page 10 in the revised manuscript as follows:

'However, the slight colour differences among design (Fig.1), simulation (Fig.2) and experiment results (Fig.4) can be caused by several factors. 1) The colours in Fig.1 are in an ideal situation where no light inter-absorption behaviour occur, and mixed lights were generated by RGB255 system. 2) The colours in Fig.2 are in semi-ideal conditions where the regular cubic pixel structure remained, but light re-absorption was also taken into consideration. 3) Real structure in experiment (Fig.4) shows irregular pixel shape and some blank areas on the substrate. 4) There is no environmental light assisted in simulation, therefore light colours in simulation and experiment will be somehow distinctive visually. '

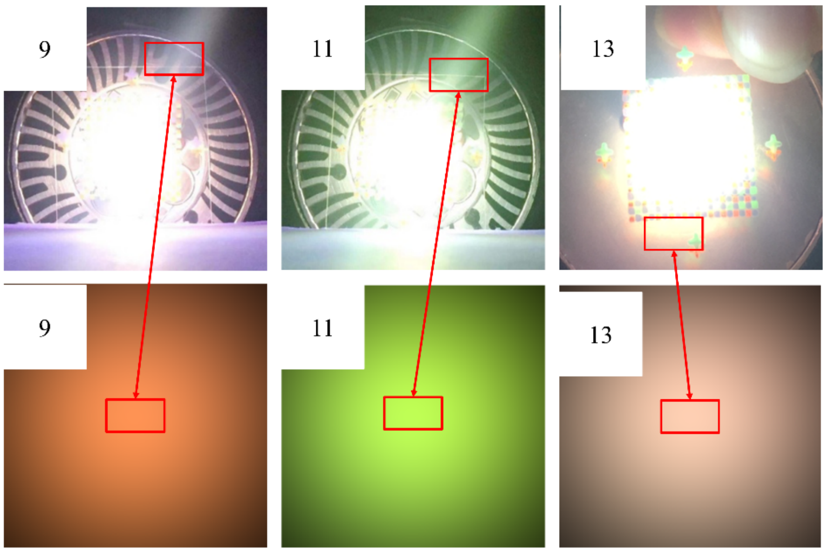


Figure. Comparison between simulation results and the margin area of experiment results

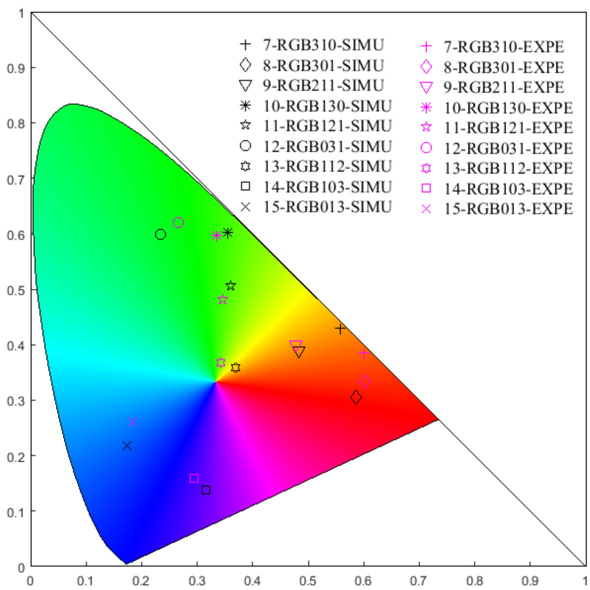


Figure. CIE comparison of simulated and experimental results of phosphor layers labelled from 7 to 15

Multi-Colour Light Emission based on Pixel-Array Phosphor Layer in LEDs

Haitao Zhu,[a] Renli Fu,[a*] Yahui Shi,[a] Qinjiang He,[a] He Wang,[a] Xuhai Liu[b*]

[a]College of Materials Science and Technology, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China

[b]College of Microtechnology & Nanotechnology, Qingdao University, Qingdao 266071, China

*Author to whom correspondence should be addressed

E-mail: renlifu@nuaa.edu.cn (Prof. R. Fu), xuhai@qdu.edu.cn (Prof. X. Liu)

Abstract

Phosphor layers are of vital importance for the development of advanced phosphor-converted light-emitting diodes (LEDs). However, owing to the fixed ratio of red-green-blue (RGB) phosphors, it has been difficult for the RGB phosphor layers with conventional structure to tune light emission colours. Herein, we have experimentally fabricated nine types of phosphor layers with patterned RGB pixel array, which consist of tuneable RGB ratio in a planar configuration. Moreover, we carried out optical simulation based on Monte-Carlo theory to assist in adjusting the light-emission colours and the corresponding chromaticity coordinates. The simulations were further verified by the experimental results via samples fabricated by the stencil printing technique. In accordance with the nine types of phosphor layers with patterned pixel arrays in various RGB ratios, we have finally obtained corresponding nine light-emission colours for the applications of LED light emission decoration. These designed advanced pixel-array phosphor layers demonstrate great potential for applications in decorated light emission and display devices with significant implications for industrial improvement in these research areas.

Keyword: phosphor layer, stencil printing, multi-colour light emission, pixel array.

1. Introduction

Owing to the high luminous efficiency, light-emitting diodes (LEDs) have gradually replaced fluorescent and incandescent lamps in commercial lighting applications [1-2]. To tune LED colour is so far usually obtained by three manners, namely the direct implementation of RGB chips, via the combination of blue chip and yellow phosphor, and through the combination of near-ultraviolet chip and red-green-blue (RGB) phosphors [3-4]. Among them, the method to directly apply RGB chips requires complex driving circuits, which results in complicated cost-ineffective fabrication process [5-7]. In addition, the blue chip combined with yellow phosphor is a traditional method to tune the light emission, however, with limited device performance. In contrast, combining different colour conversion phosphors with ultraviolet LED chip can lead to tuneable light-emission colours with superior device performance [8-11].

Specifically, the ultraviolet LEDs combined with RGB phosphors can achieve multi-colour emission via low-cost dispensing coating technique. Nevertheless, the red phosphor can partially absorb the converted green and blue light emission, and also the green phosphor can partially absorb the converted blue light emission [12]. This enables inferior green and blue light emission as well as low luminous efficiency (LE) of multi-colour LED devices [13]. Therefore, intensive research efforts have been devoted to modifying the configuration of RGB phosphor layers and to develop new RGB phosphors, in an attempt to simultaneously achieve light emission with high luminous efficiency and tuneable colours [14-16].

The vertical configuration of RGB phosphor layers were up to now often applied to tune the light-emission colours [17-19]. In order to avoid the blue light and green light re-absorption by green and red phosphors, the red phosphor was usually coated on the LED chips firstly, with the subsequent coating of green and blue phosphors sequentially. The red and green light can spread through green and blue phosphor layers, respectively. Therefore, only a small quantity of backscattering blue light can be re-absorbed by red and green phosphors in this case, and the corresponding output LED luminescence efficiency can be greatly improved. In addition, by adjusting the content of RGB phosphors in each dispensing layer, specific light-emission colours can be realized by the multi-layer RGB phosphors with ultraviolet LED. Unfortunately, this stacking configuration of RGB phosphor layers can increase the total thickness of the devices, with facile backscattering of the emitting light. Also, according to Beer-Lambert Law, the transmittance of light from inner phosphor layer can be reduced due to the adoption of upper phosphor layer, which renders the stacking configuration as unsuitable candidate for the most optimized luminous efficiency. Additionally, no red and green phosphors have been reported in literature that only allows absorption of near-ultraviolet rather than green and blue light emission. Besides, developing single phosphor with multi-emission peak is cost-ineffective [20-21]. Therefore, to optimize the phosphor layer in a planar rather than vertical configuration could be a promising route for more advanced phosphor-converted multi-colour LEDs.

We have previously developed a new LED phosphor layer with patterned RGB pixel array in a planar configuration [22]. This pattern RGB phosphor layer can avoid the problem of reabsorption by RGB phosphors in multiple layers, thereby improving the luminous efficiency of the LEDs. In this study, we have applied Monte Carlo method to simulate 15 types of patterned RGB pixel-array phosphor layers. Among them, the chromaticity and luminous efficiency of the nine "Secondary Mixing Colours" were also experimentally evaluated. It has been revealed that these patterns of phosphor layers can successfully realize excellent output light colours, opening route for applications in decorated light emission and display devices with significant implications for industrial improvement.

2. Optical simulation of pixel-array phosphor layer

In our previous work, we have developed a phosphor layer with planar pixel-array configuration, and then obtained uniform white light emission with improved luminous efficiency [22]. In this work, based on the same pixel-array configuration strategy, firstly we try to tune the pixel-array configuration of RGB phosphors to obtain multi-colour light emission in LEDs. Via the optical simulations based on Monte-Carlo theory, nine pixel-array configurations of RGB phosphors have been proposed via the primary and secondary mixing method. The design process of the pixel-array configurations of RGB phosphors is provided in Fig.1.

As shown in Fig.1, the red, green and blue colours are labelled as colour1, colour2 and colour3, respectively, which can be used as primary colours to further obtain other colours, according to the colour mix principle. Via selecting and mixing two of the three primary colours, the three new colours are further obtained, namely the "First Mixing Colours" in Fig.1 and labelled as colour4, colour5 and colour6, respectively.

Afterwards, by repeating the selection and mixing process, another nine colours can be obtained that are named as “Secondary Mixing Colours” in Fig.1 and labelled as colour7 to colour15 in sequence. Next, fifteen configurations of pixel-array phosphor layers can be obtained in accordance with the aforementioned fifteen colours, with 400 pixels for each, as illustrated in Fig.1. Finally, fifteen simulation models have been set up based on these pixel-array configurations, and each includes the light source, the pixel-array phosphor layer, and the transparent plate as well as the receiver.

We have previously verified that the LEDs packaged with the planar-patterned RGB phosphors can exhibit good uniformity of correlated colour temperature and high luminous efficiency, when the pixel size was 1.0×1.0×0.1 mm [22]. Therefore, in this study, for the fifteen configurations of phosphor layers, we have adopted the same device layout and pixel size to carry out optical simulation and corresponding experimental studies.

In this simulation model, a great number of light rays emit from the upper surface of the blue chip, and they are randomly distributed with certain energy. The total energy W can be expressed as follows:

$$W = \sum W_i \tag{1}$$

where W_i is the energy of the n-UV ray with label i.

Next, the lambert-beer law can be used to describe the process in which the ultraviolet light rays interact with the phosphor layer:

$$A = \varepsilon c l \tag{2}$$

where A is the absorbance of the phosphor layer, ε and c are the molar absorptivity and the concentration of phosphors, respectively, and l is the thickness of the phosphor layer. The scattering energy and direction can be described by Henyey-Greenstein function (Equation 3). Scattering coefficient and anisotropy coefficient can be calculated according to the Mie theory with this model.

$$p(\theta) = \frac{1 - g^2}{4\pi(1 - 2g\cos\theta + g^2)^{\frac{3}{2}}} \quad (3)$$

where θ is the exiting angle of scattering, g is anisotropy coefficient.

Overall output energy of blue light in this process can be expressed as:

$$W_B = q_B A_{B-emission} - A_{R-B} - A_{G-B} \quad (4)$$

where W_B is the final output energy of blue light, $A_{B\text{-}emission}$, $A_{R\text{-}B}$ and $A_{G\text{-}B}$ are energy absorbance of blue light to n-UV light, by red and green phosphors, q_B is the quantum conversion efficiency of blue phosphor.

Similarly, the output energy of green and red light can be expressed as:

$$W_G = q_G(A_{G-emission} + A_{G-B}) - A_{R-G} \quad (5)$$

$$W_R = q_R(A_{R-emission} + A_{R-B} + A_{R-G}) \quad (6)$$

Table.1 provides the detailed dimensions and optical parameters for the simulation models.

Fig.2 demonstrates the simulated results of light colours and chromaticity coordinate of the fifteen phosphor layers patterns. Among them, the light colours of pattern 1, 2, 3 are the primary colours of RGB phosphors, whereas the other eleven colours are obtained via mixing three primary colours according to the CIE diagram.

To be specific, light colours of patterns 4, 5, 6 are obtained by mixing two of RGB colours, therefore their chromaticity coordinates locate at the line connected by the CIE points of two primary colours.

3. Preparation of the phosphor pixel-array layer

RGB phosphors (*i.e.*, red phosphor N630, green phosphor S525, and blue phosphor BAM460) were purchased from Shenzhen Prospect Technology Co., Ltd. The home-made organic vehicle was prepared by mixing terpineol, dibutyl phthalate, butyl carbitol acetate, castor oil and ethocel. Colourless and transparent organic glass substrates polymethyl methacrylate (PMMA) were applied in the experiments. The preparation process of RGB phosphor layer mainly consists of two steps: (1) Preparation of RGB phosphor paste, and (2) Stencil printing process. Regarding the preparation of phosphor paste, a 150-mesh sieve was used to obtain phosphor with similar grain size. Afterwards, the phosphor and organic vehicle were weighed on an analytical balance. Around 25 wt% organic vehicle was added in phosphor to make the paste. Then, the mixture of phosphor and organic vehicle was magnetically stirred for 30 minutes at 60°C, and subsequently put in a sealed cabinet with 10^{-1} Pa negative pressure for 60 minutes, which can fully mix phosphor and organic vehicle without bubbles. The phosphor paste was then printed onto the surface of PMMA plate, which was fixed onto a stencil printing device using rotary vacuum pump, and the distance between the stencil bottom and the upper-surface of the PMMA plate was about 1.0 mm, as demonstrated in Fig.3 (a). After stencil printing the blue phosphor paste, the sample was moved into an electric-heating vacuum drying oven and dried at 60°C for

20 minutes. Fig.3 (b) illustrates the distribution of RGB phosphors pixels, and the three stencils are labelled as location 1, 2, 3. The RGB phosphor paste were printed at three locations (labelled as 1, 2, 3) to obtain the designed pixel-array phosphor layers.

According to the simulation, the output light efficiency can be more inferior with increasing the phosphors in phosphor layer, and the light emission of pattern 7 to 15 can be inhomogeneous than that of pattern 1-6. Therefore, we have selected and fabricated the phosphor layers labelled from 7 to 15 by stencil-printing technique, to verify the light emission effect of the phosphor pixel array layer. The optical performance of the pattern 7 to 15 phosphor pixel array layers, including the emitted spectra, the output light colours and the chromaticity coordinate, were measured by a Chameleon-QY system (Zolix Instruments Co., Ltd), as shown in Fig.3 (c).

4. Optical characteristics of the pixel-array phosphor layer

The RGB phosphor pixel array of patterns 7 to 15 were fabricated on the transparent PMMA film with thickness of 0.2 mm through screen printing technique, as shown in Fig.4 (a). There are 400 pixels for each pixel-array phosphor layer (20 mm×20 mm) of the sample, where the orange, light green and white region (visually light green and white colours might be difficult to distinguish in pattern13) represent red, green and blue phosphor pixels, respectively. These positions correspond to that in Fig.1 (red, green and blue square), although there are still slight differences on cubic shape (pattern9 and 14) and blank area (pattern7 and 15), which is difficult to avoid in manual operation. The optical simulation and experimental results of the patterns 7 to 15 RGB pixel-array phosphor layer are demonstrated in Fig. 4 (b) to 4 (e).

To be specific, the optical simulation and experimental light colours of RGB pixel-array phosphor pattern 7 to 9 are provided in Fig. 4 (b). The designed colours of pattern 7 to 9 were approx. international orange, razzmatazz and light coral based on the principle of mixing light, respectively. Furthermore, the optical simulation and experimental light colours of RGB pixel-array phosphor pattern 10 to 12 and 13 to15 are illustrated in Fig. 4 (c) and Fig. 4 (d), respectively. The designed colours of pattern 10 to 12 were approx. bright green, light green and malachite, respectively. Furthermore, the designed colours of pattern 13 to 15 were approx. light slate blue, electric indigo and navy blue, respectively [23].

The nine different light colours of the RGB pixel-array phosphor patterns 7 to 15 layers were in conformity with optical simulation and experimental results. However, the slight colour differences among design (Fig.1), simulation (Fig.2) and experiment results (Fig.4) can be caused by several factors. 1) The colours in Fig.1 are in an ideal situation where no light inter-absorption behaviour occur, and mixed lights were generated by RGB255 system. 2) The colours in Fig.2 are in semi-ideal conditions where the regular cubic pixel structure remained, but light re-absorption was also taken into consideration. 3) Real structure in experiment (Fig.4) shows irregular pixel shape and some blank areas on the substrate. 4) There is no environmental light assisted in simulation, therefore light colours in simulation and experiment will be somehow distinctive visually. Basically the chromaticity coordinates results in Fig.4 (e) showed a slight difference between optical simulation and measurement results can be observed for the RGB pixel-array phosphor patterns 7 to 15 layers. Considering the simulation accuracy, this difference can be neglected. The designed 15 phosphors pixel array layer can realize 15 output light colours, and more light colours can be obtained by additive colour methodology.

5. Conclusion

LEDs packaged with RGB phosphors are widely adopted in the current lighting industry, not only in white light illumination but also in decorated illumination. It has been difficult to modify output light colours once the phosphor layers have been solidified. In this work, by designing pixel-array phosphor layers with different RGB phosphors, in combination with remote phosphor-converted lighting method, LEDs can realize multi-colour light emission. The samples fabricated in this paper can achieve 15 output colours, and based on additive colour methodology more light colours can be obtained in future studies. This significant improvement extends the application route for the pixel-array phosphor layers in high-performance lighting industry.

Acknowledgements

This work was financially supported by thePriority Academic Program Development of Jiangsu Higher Education Institution (PAPD), Nanjing, China, and the Fundamental Research Funds for the Central Universities (No. kfjj20180616). Xuhai Liu gratefully acknowledges support from The Startup Foundation for Introducing Talent of Qingdao University (DC1900013518).

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Figure Captions

Fig.1 Design process of the multi-colour pixel-array phosphor layers.

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Tables

Tab.1 Parameters applied in the optical simulation [22].

Component	Dimension	Parameter
Chip	1×1×0.1 mm	1 W, 360 nm, ultraviolet chip
Blue phosphor	Pixel size:1.0×1.0×0.1 mm, square, total: 4.0 mm. H 0.1 mm,	S:11.5, g:0.9, ε_{uv}:0.7, n:1.6, q_B:0.8, c:200 mol/L
Green phosphor		S:16.5, g:0.89, ε_{uv}:0.7, ε_B:0.6, n:1.6, q_G:0.9, c:200 mol/L
Red phosphor		S:16.5, g:0.89, ε_{uv}:0.3, ε_B:0.3, ε_G:0.3, n:1.6, q_R:0.9, c:200 mol/L
Transparent substrate	R:3.0, H:0.1 mm	PMMA, Transparent, no absorption
Reflective cup	R:3.0, H:1.0 mm	Perfect mirror reflection

* Abbreviations in Table 1: *H*- thickness, *R*- radius, *S*- Scattering coefficient, *n*-refractive index

Highlight

1. Nine patterned phosphor layers can realize nine output light colours
2. With increasing mixing time of RGB phosphors, more output light colours of phosphor layers can be obtained.
3. The pixel-array phosphor layers was prepared via stencil printing method which is suitable for a large-scale production.

Multi-Colour Light Emission based on Pixel-Array Phosphor Layer in LEDs

Haitao Zhu,[a] Renli Fu,[a*] Yahui Shi,[a] Qinjiang He,[a] He Wang,[a] Xuhai Liu[b*]

[a]College of Materials Science and Technology, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China

[b]College of Microtechnology & Nanotechnology, Qingdao University, Qingdao 266071, China

*Author to whom correspondence should be addressed

E-mail: renlifu@nuaa.edu.cn (Prof. R. Fu), xuhai@qdu.edu.cn (Prof. X. Liu)

Abstract

Phosphor layers are of vital importance for the development of advanced phosphor-converted light-emitting diodes (LEDs). However, owing to the fixed ratio of red-green-blue (RGB) phosphors, it has been difficult for the RGB phosphor layers with conventional structure to tune light emission colours. Herein, we have experimentally fabricated nine types of phosphor layers with patterned RGB pixel array, which consist of tuneable RGB ratio in a planar configuration. Moreover, we carried out optical simulation based on Monte-Carlo theory to assist in adjusting the light-emission colours and the corresponding chromaticity coordinates. The simulations were further verified by the experimental results via samples fabricated by the stencil printing technique. In accordance with the nine types of phosphor layers with patterned pixel arrays in various RGB ratios, we have finally obtained corresponding nine light-emission colours for the applications of LED light emission decoration. These designed advanced pixel-array phosphor layers demonstrate great potential for applications in decorated light emission and display devices with significant implications for industrial improvement in these research areas.

Keyword: phosphor layer, stencil printing, multi-colour light emission, pixel array.

1. Introduction

Owing to the high luminous efficiency, light-emitting diodes (LEDs) have gradually replaced fluorescent and incandescent lamps in commercial lighting applications [1-2]. To tune LED colour is so far usually obtained by three manners, namely the direct implementation of RGB chips, via the combination of blue chip and yellow phosphor, and through the combination of near-ultraviolet chip and red-green-blue (RGB) phosphors [3-4]. Among them, the method to directly apply RGB chips requires complex driving circuits, which results in complicated cost-ineffective fabrication process [5-7]. In addition, the blue chip combined with yellow phosphor is a traditional method to tune the light emission, however, with limited device performance. In contrast, combining different colour conversion phosphors with ultraviolet LED chip can lead to tuneable light-emission colours with superior device performance [8-11].

Specifically, the ultraviolet LEDs combined with RGB phosphors can achieve multi-colour emission via low-cost dispensing coating technique. Nevertheless, the red phosphor can partially absorb the converted green and blue light emission, and also the green phosphor can partially absorb the converted blue light emission [12]. This enables inferior green and blue light emission as well as low luminous efficiency (LE) of multi-colour LED devices [13]. Therefore, intensive research efforts have been devoted to modifying the configuration of RGB phosphor layers and to develop new RGB phosphors, in an attempt to simultaneously achieve light emission with high luminous efficiency and tuneable colours [14-16].

The vertical configuration of RGB phosphor layers were up to now often applied to tune the light-emission colours [17-19]. In order to avoid the blue light and green light re-absorption by green and red phosphors, the red phosphor was usually coated on the LED chips firstly, with the subsequent coating of green and blue phosphors sequentially. The red and green light can spread through green and blue phosphor layers, respectively. Therefore, only a small quantity of backscattering blue light can be re-absorbed by red and green phosphors in this case, and the corresponding output LED luminescence efficiency can be greatly improved. In addition, by adjusting the content of RGB phosphors in each dispensing layer, specific light-emission colours can be realized by the multi-layer RGB phosphors with ultraviolet LED. Unfortunately, this stacking configuration of RGB phosphor layers can increase the total thickness of the devices, with facile backscattering of the emitting light. Also, according to Beer-Lambert Law, the transmittance of light from inner phosphor layer can be reduced due to the adoption of upper phosphor layer, which renders the stacking configuration as unsuitable candidate for the most optimized luminous efficiency. Additionally, no red and green phosphors have been reported in literature that only allows absorption of near-ultraviolet rather than green and blue light emission. Besides, developing single phosphor with multi-emission peak is cost-ineffective [20-21]. Therefore, to optimize the phosphor layer in a planar rather than vertical configuration could be a promising route for more advanced phosphor-converted multi-colour LEDs.

We have previously developed a new LED phosphor layer with patterned RGB pixel array in a planar configuration [22]. This pattern RGB phosphor layer can avoid the problem of reabsorption by RGB phosphors in multiple layers, thereby improving the luminous efficiency of the LEDs. In this study, we have applied Monte Carlo method to simulate 15 types of patterned RGB pixel-array phosphor layers. Among them, the chromaticity and luminous efficiency of the nine "Secondary Mixing Colours" were also experimentally evaluated. It has been revealed that these patterns of phosphor layers can successfully realize excellent output light colours, opening route for applications in decorated light emission and display devices with significant implications for industrial improvement.

2. Optical simulation of pixel-array phosphor layer

In our previous work, we have developed a phosphor layer with planar pixel-array configuration, and then obtained uniform white light emission with improved luminous efficiency [22]. In this work, based on the same pixel-array configuration strategy, firstly we try to tune the pixel-array configuration of RGB phosphors to obtain multi-colour light emission in LEDs. Via the optical simulations based on Monte-Carlo theory, nine pixel-array configurations of RGB phosphors have been proposed via the primary and secondary mixing method. The design process of the pixel-array configurations of RGB phosphors is provided in Fig.1.

As shown in Fig.1, the red, green and blue colours are labelled as colour1, colour2 and colour3, respectively, which can be used as primary colours to further obtain other colours, according to the colour mix principle. Via selecting and mixing two of the three primary colours, the three new colours are further obtained, namely the "First Mixing Colours" in Fig.1 and labelled as colour4, colour5 and colour6, respectively.

Afterwards, by repeating the selection and mixing process, another nine colours can be obtained that are named as "Secondary Mixing Colours" in Fig.1 and labelled as colour7 to colour15 in sequence. Next, fifteen configurations of pixel-array phosphor layers can be obtained in accordance with the aforementioned fifteen colours, with 400 pixels for each, as illustrated in Fig.1. Finally, fifteen simulation models have been set up based on these pixel-array configurations, and each includes the light source, the pixel-array phosphor layer, and the transparent plate as well as the receiver.

We have previously verified that the LEDs packaged with the planar-patterned RGB phosphors can exhibit good uniformity of correlated colour temperature and high luminous efficiency, when the pixel size was 1.0×1.0×0.1 mm [22]. Therefore, in this study, for the fifteen configurations of phosphor layers, we have adopted the same device layout and pixel size to carry out optical simulation and corresponding experimental studies.

In this simulation model, a great number of light rays emit from the upper surface of the blue chip, and they are randomly distributed with certain energy. The total energy W can be expressed as follows:

$$W = \sum W_i \tag{1}$$

where W_i is the energy of the n-UV ray with label i.

Next, the lambert-beer law can be used to describe the process in which the ultraviolet light rays interact with the phosphor layer:

$$A = \varepsilon c l \tag{2}$$

where A is the absorbance of the phosphor layer, ε and c are the molar absorptivity and the concentration of phosphors, respectively, and l is the thickness of the phosphor layer. The scattering energy and direction can be described by Henyey-Greenstein function (Equation 3). Scattering coefficient and anisotropy coefficient can be calculated according to the Mie theory with this model.

$$p(\theta) = \frac{1 - g^2}{4\pi(1 - 2g\cos\theta + g^2)^{\frac{3}{2}}} \tag{3}$$

where θ is the exiting angle of scattering, g is anisotropy coefficient.

Overall output energy of blue light in this process can be expressed as:

$$W_B = q_B A_{B-emission} - A_{R-B} - A_{G-B} \tag{4}$$

where W_B is the final output energy of blue light, $A_{B\text{-}emission}$, $A_{R\text{-}B}$ and $A_{G\text{-}B}$ are energy absorbance of blue light to n-UV light, by red and green phosphors, q_B is the quantum conversion efficiency of blue phosphor.

Similarly, the output energy of green and red light can be expressed as:

$$W_G = q_G(A_{G-emission} + A_{G-B}) - A_{R-G} \tag{5}$$

$$W_R = q_R(A_{R-emission} + A_{R-B} + A_{R-G}) \tag{6}$$

Table.1 provides the detailed dimensions and optical parameters for the simulation models.

Fig.2 demonstrates the simulated results of light colours and chromaticity coordinate of the fifteen phosphor layers patterns. Among them, the light colours of pattern 1, 2, 3 are the primary colours of RGB phosphors, whereas the other eleven colours are obtained via mixing three primary colours according to the CIE diagram.

To be specific, light colours of patterns 4, 5, 6 are obtained by mixing two of RGB colours, therefore their chromaticity coordinates locate at the line connected by the CIE points of two primary colours.

3. Preparation of the phosphor pixel-array layer

RGB phosphors (*i.e.*, red phosphor N630, green phosphor S525, and blue phosphor BAM460) were purchased from Shenzhen Prospect Technology Co., Ltd. The home-made organic vehicle was prepared by mixing terpineol, dibutyl phthalate, butyl carbitol acetate, castor oil and ethocel. Colourless and transparent organic glass substrates polymethyl methacrylate (PMMA) were applied in the experiments. The preparation process of RGB phosphor layer mainly consists of two steps: (1) Preparation of RGB phosphor paste, and (2) Stencil printing process. Regarding the preparation of phosphor paste, a 150-mesh sieve was used to obtain phosphor with similar grain size. Afterwards, the phosphor and organic vehicle were weighed on an analytical balance. Around 25 wt% organic vehicle was added in phosphor to make the paste. Then, the mixture of phosphor and organic vehicle was magnetically stirred for 30 minutes at 60°C, and subsequently put in a sealed cabinet with 10^{-1} Pa negative pressure for 60 minutes, which can fully mix phosphor and organic vehicle without bubbles. The phosphor paste was then printed onto the surface of PMMA plate, which was fixed onto a stencil printing device using rotary vacuum pump, and the distance between the stencil bottom and the upper-surface of the PMMA plate was about 1.0 mm, as demonstrated in Fig.3 (a). After stencil printing the blue phosphor paste, the sample was moved into an electric-heating vacuum drying oven and dried at 60°C for

20 minutes. Fig.3 (b) illustrates the distribution of RGB phosphors pixels, and the three stencils are labelled as location 1, 2, 3. The RGB phosphor paste were printed at three locations (labelled as 1, 2, 3) to obtain the designed pixel-array phosphor layers.

According to the simulation, the output light efficiency can be more inferior with increasing the phosphors in phosphor layer, and the light emission of pattern 7 to 15 can be inhomogeneous than that of pattern 1-6. Therefore, we have selected and fabricated the phosphor layers labelled from 7 to 15 by stencil-printing technique, to verify the light emission effect of the phosphor pixel array layer. The optical performance of the pattern 7 to 15 phosphor pixel array layers, including the emitted spectra, the output light colours and the chromaticity coordinate, were measured by a Chameleon-QY system (Zolix Instruments Co., Ltd), as shown in Fig.3 (c).

4. Optical characteristics of the pixel-array phosphor layer

The RGB phosphor pixel array of patterns 7 to 15 were fabricated on the transparent PMMA film with thickness of 0.2 mm through screen printing technique, as shown in Fig.4 (a). There are 400 pixels for each pixel-array phosphor layer (20 mm×20 mm) of the sample, where the orange, light green and white region (visually light green and white colours might be difficult to distinguish in pattern13) represent red, green and blue phosphor pixels, respectively. These positions correspond to that in Fig.1 (red, green and blue square), although there are still slight differences on cubic shape (pattern9 and 14) and blank area (pattern7 and 15), which is difficult to avoid in manual operation. The optical simulation and experimental results of the patterns 7 to 15 RGB pixel-array phosphor layer are demonstrated in Fig. 4 (b) to 4 (e).

To be specific, the optical simulation and experimental light colours of RGB pixel-array phosphor pattern 7 to 9 are provided in Fig. 4 (b). The designed colours of pattern 7 to 9 were approx. international orange, razzmatazz and light coral based on the principle of mixing light, respectively. Furthermore, the optical simulation and experimental light colours of RGB pixel-array phosphor pattern 10 to 12 and 13 to15 are illustrated in Fig. 4 (c) and Fig. 4 (d), respectively. The designed colours of pattern 10 to 12 were approx. bright green, light green and malachite, respectively. Furthermore, the designed colours of pattern 13 to 15 were approx. light slate blue, electric indigo and navy blue, respectively [23].

The nine different light colours of the RGB pixel-array phosphor patterns 7 to 15 layers were in conformity with optical simulation and experimental results. However, the slight colour differences among design (Fig.1), simulation (Fig.2) and experiment results (Fig.4) can be caused by several factors. 1) The colours in Fig.1 are in an ideal situation where no light inter-absorption behaviour occur, and mixed lights were generated by RGB255 system. 2) The colours in Fig.2 are in semi-ideal conditions where the regular cubic pixel structure remained, but light re-absorption was also taken into consideration. 3) Real structure in experiment (Fig.4) shows irregular pixel shape and some blank areas on the substrate. 4) There is no environmental light assisted in simulation, therefore light colours in simulation and experiment will be somehow distinctive visually. Basically the chromaticity coordinates results in Fig.4 (e) showed a slight difference between optical simulation and measurement results can be observed for the RGB pixel-array phosphor patterns 7 to 15 layers. Considering the simulation accuracy, this difference can be neglected. The designed 15 phosphors pixel array layer can realize 15 output light colours, and more light colours can be obtained by additive colour methodology.

5. Conclusion

LEDs packaged with RGB phosphors are widely adopted in the current lighting industry, not only in white light illumination but also in decorated illumination. It has been difficult to modify output light colours once the phosphor layers have been solidified. In this work, by designing pixel-array phosphor layers with different RGB phosphors, in combination with remote phosphor-converted lighting method, LEDs can realize multi-colour light emission. The samples fabricated in this paper can achieve 15 output colours, and based on additive colour methodology more light colours can be obtained in future studies. This significant improvement extends the application route for the pixel-array phosphor layers in high-performance lighting industry.

Acknowledgements

This work was financially supported by thePriority Academic Program Development of Jiangsu Higher Education Institution (PAPD), Nanjing, China, and the Fundamental Research Funds for the Central Universities (No. kfjj20180616). Xuhai Liu gratefully acknowledges support from The Startup Foundation for Introducing Talent of Qingdao University (DC1900013518).

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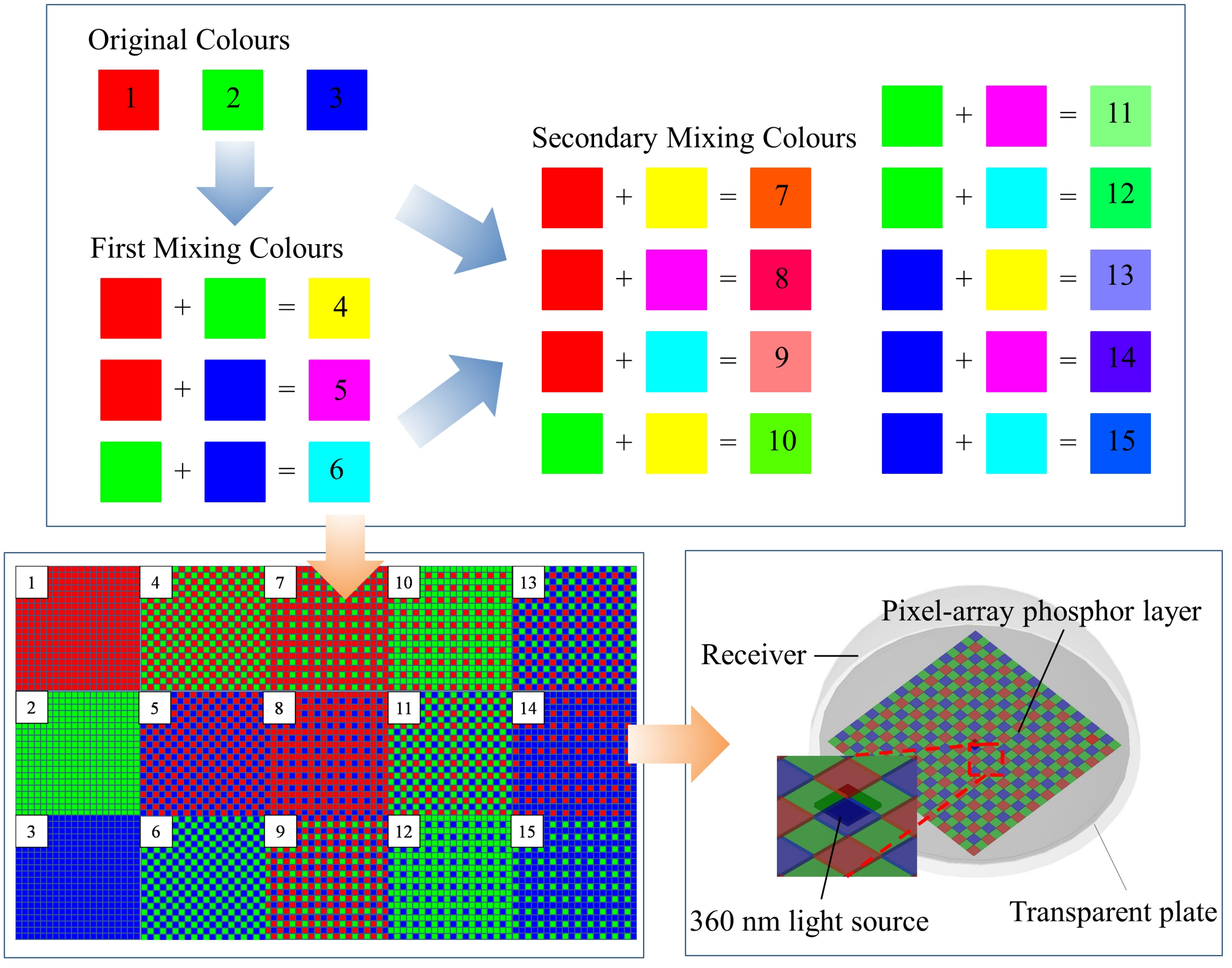
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First Mixing Colours
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Secondary Mixing Colours
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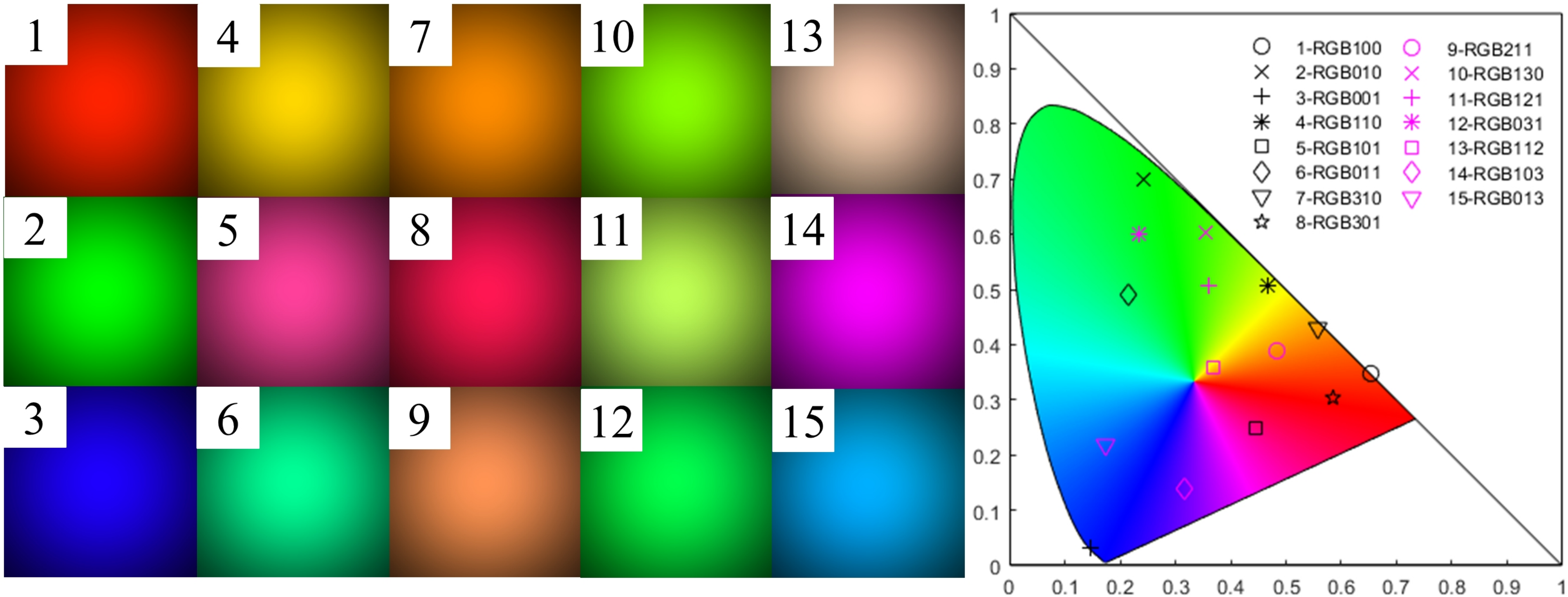
Pixel-array phosphor layer
Receiver
360 nm light source
Transparent plate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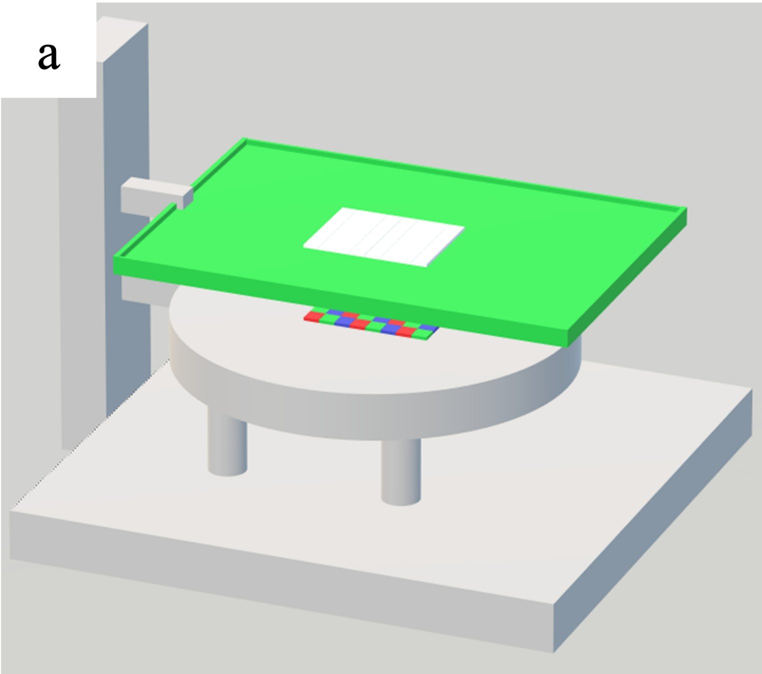
(a) (b)



b

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			1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1
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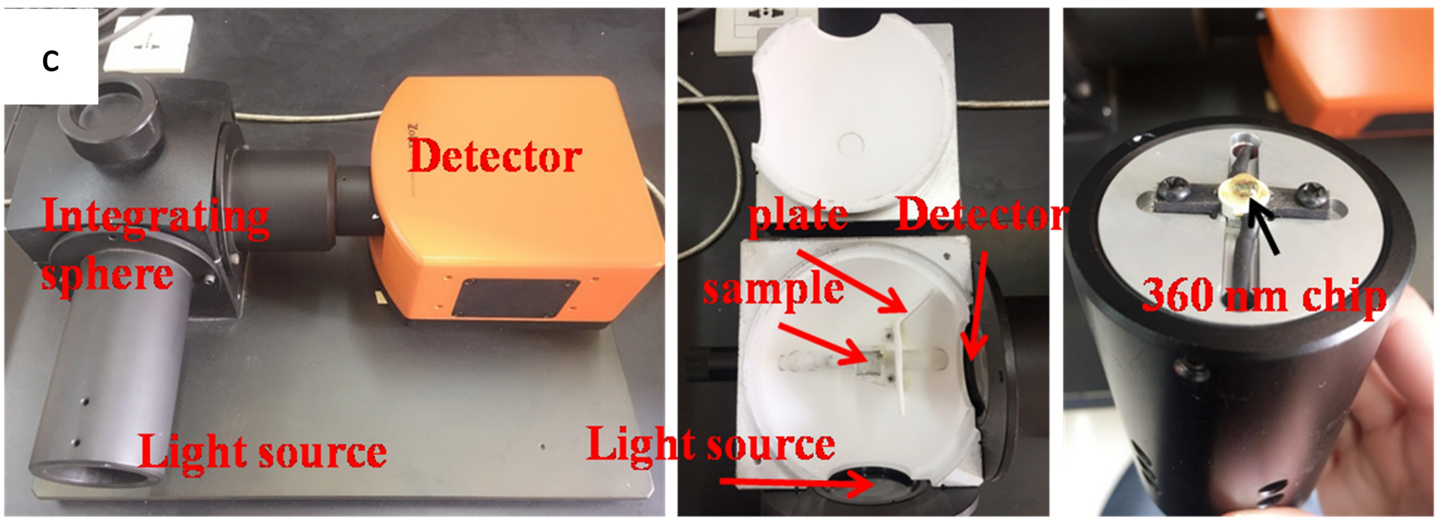
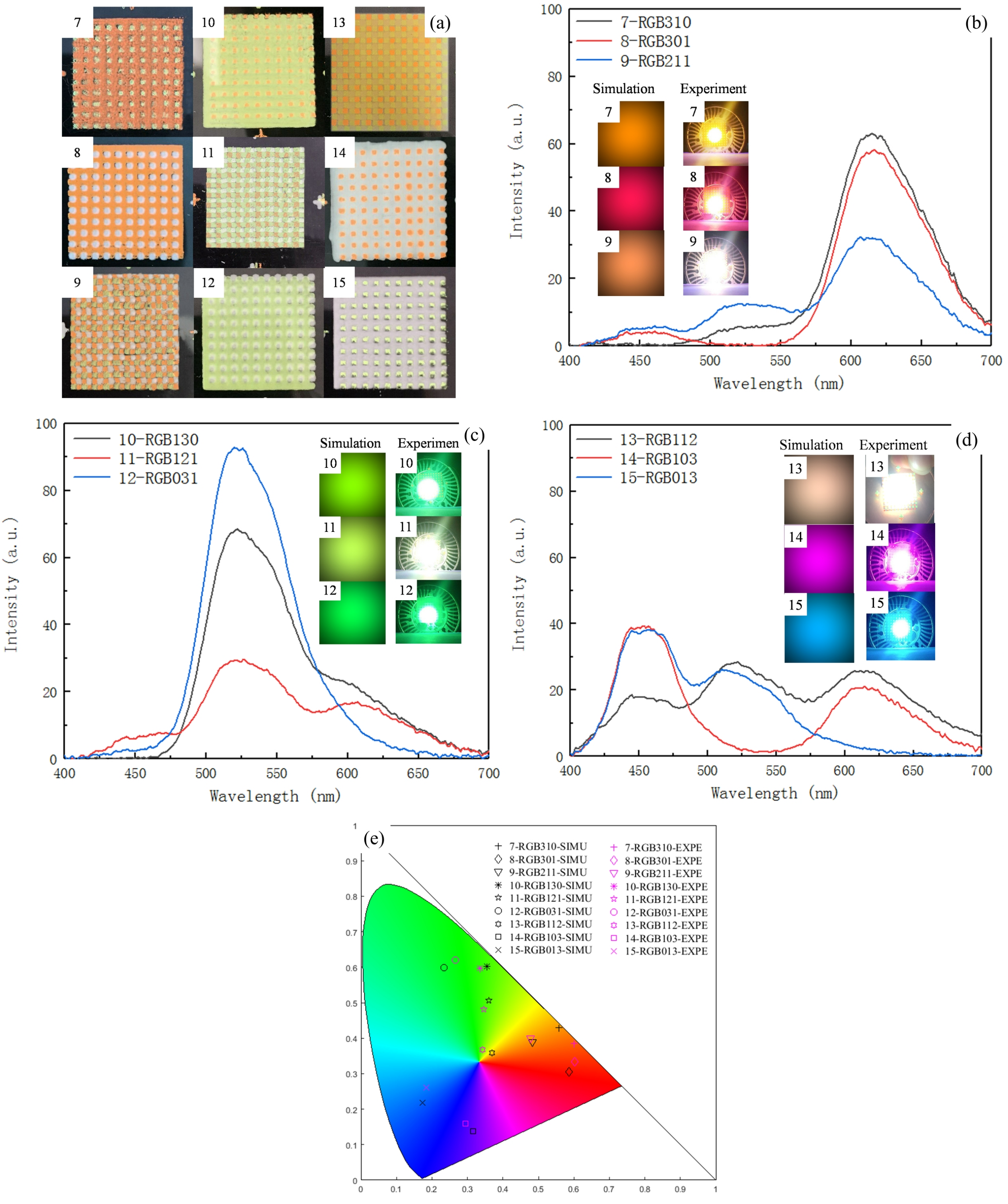
1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3
2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1
1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3
2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1
1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3
2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1
1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3
2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1	2	1
1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3	1	3

1 - Location 1   2 - Location 2   3 - Location 3





(a)
7
10
13
8
11
14
9
12
15
(b)
7-RGB310
8-RGB301
9-RGB211
Simulation
Experiment
Intensity (a. u.)
Wavelength (nm)
(c)
10-RGB130
11-RGB121
12-RGB031
Simulation
Experimen
Intensity (a. u.)
Wavelength (nm)
(d)
13-RGB112
14-RGB103
15-RGB013
Simulation
Experiment
Intensity (a. u.)
Wavelength (nm)
(e)
7-RGB310-SIMU
8-RGB301-SIMU
9-RGB211-SIMU
10-RGB130-SIMU
11-RGB121-SIMU
12-RGB031-SIMU
13-RGB112-SIMU
14-RGB103-SIMU
15-RGB013-SIMU
7-RGB310-EXPE
8-RGB301-EXPE
9-RGB211-EXPE
10-RGB130-EXPE
11-RGB121-EXPE
12-RGB031-EXPE
13-RGB112-EXPE
14-RGB103-EXPE
15-RGB013-EXPE

Conflict of Interest

Authors: Haitao Zhu, Renli Fu, Yahui Shi, Qinjiang He, He Wang, Xuhai Liu

We promise that there are no conflicts of interest to declare.

Author Statement

Authors: Haitao Zhu, Renli Fu, Yahui Shi, Qinjiang He, He Wang, Xuhai Liu

This manuscript has not been published elsewhere and is not under consideration by another journal. We have approved the manuscript and agree with submission to Journal of Luminescence.