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Abstract. We present a study of light-emitting diodes (LEDs) using organic fluorescent dyes to replace the general phosphor. The blue die with a specific organic fluorescent dye gives the LED a single color appearance. Through a color-mixing cavity, multiple LEDs are used to produce a quasisolar spectrum at a certain band and white light with a color rendering index as high as 97 at around 2800 K. [DOI: 10.1117/1.OE.54.7.070501]

Keywords: light-emitting diode; color; solar simulator; dye.

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White light-emitting diodes (LED) have become one of the major light sources in modern lighting because of its advantages, such as fast response, long life, wide-color range, and lack of mercury.[1–6] Generally, a white LED is made by a blue die with a yellow phosphor and the LED attains approximately 70 on the color rendering index (CRI),[7] which is a measure of a light source's ability to show an object's colors "realistically" or "naturally" compared to a familiar reference source, either incandescent light or daylight. The color performance of the typical phosphor-converted white LED is acceptable in outdoor lighting. In indoor lighting, however, a white LED must provide a higher CRI, therefore, two or more kinds of phosphors are used to supplement the spectrum.[6,8–12] Even the approach of multiple phosphors for a wider spectrum still has some issues that need to be solved. The first issue is that it will cause more light absorption by the multiple phosphors, so the packaging efficiency[5] is at a low level. The second issue is that it is difficult to control the vivid color of each phosphor because of multiple absorptions between the phosphors. The third issue is that the spectrum is not as flat as that of solar light and causes poor color performance in a certain spectrum, which may cause serious problems in specific applications. The third issue results in the demand for a white LED to provide a useful solar spectrum in a specific band.[13–19] In this letter, we present a study to introduce a new way for organic fluorescent dyes to produce a quasisolar spectrum with a blue die. The design concept as well as the experimental measurement is demonstrated.

Organic fluorescent dyes are transparent media with down conversion fluorescence. In contrast to commercial phosphors, the organic dyes in this study are heavy-metal-free and environmentally friendly. Generally, the spectrum bandwidth of fluorescent light is larger than that of the phosphor. It is useful for the packaging of the LED to be transparent so that the backward scattering of the blue light is reduced, thus resulting in a higher packaging efficiency.[5] A larger bandwidth enables a wide range of coverage for the composite spectrum and can even produce a specific spectrum.

In order to have a quasisolar spectrum in the visible band, we select six organic fluorescent dyes with a peak wavelength from 476 to 631 nm, which results in a high efficiency in comparison with other dyes. The dyes selected for this study are listed in Table 1. The detailed procedures for preparing the LED phosphor layer are described as follows. A mixture of 5 mg of organic dye plus 20.0 g of epoxy resin was mixed in a glass bottle and then stirred at 25°C for 1 h to form a clear polymer solution. Subsequently, the polymer solution was dip-coated on the LED chip, and was then thermally cured at 150°C under a normal atmosphere for 1 h. The fluorescent thin film was prepared by the dip-coating process at room temperature. The film thickness was 0.2 ± 0.01 mm. The absorption and emission spectra were measured with spectrophotometer HITACH U-3900H and fluorescence spectrometer HITACH F-7000, respectively. The illustration of the six organic fluorescent dyes is shown in Table 1. The peak wavelengths (bandwidth) of the six dyes are 476 (75 nm), 481 (50 nm), 505 (100 nm), 536 (102 nm), 615 (81 nm), and 631 nm (78 nm), respectively, which cover the full range of visible light. To avoid a complicated optical effect, each organic fluorescent dye is packaged with a blue die in a single package. The peak wavelength of the blue die is selected as 450 nm. The fluorescence spectrum of each organic fluorescent dye is shown in Fig. 1. The packaged LEDs are shown in Fig. 2, where the color appearance is controlled by the particular organic fluorescent dye being used.

To produce white light, we use a light pipe with a diffuser to form a color-mixing cavity for multiple light sources in different colors, as shown in Fig. 3. The cavity with a diffuser is shown to be capable of providing good color mixing.[20–25] A spectrometer is used to measure the emitted spectrum from the color-mixing cavity. To produce a quasisolar spectrum, three or more LEDs with organic fluorescent dye must be used in the color-mixing cavity. The corresponding simulations show that different recipes can be used to produce quasisolar spectrum at specific correlated color temperatures (CCT). Figure 4 shows a quasisolar spectrum at a CCT of 5200 K, where the spectra mismatch is always lower than 2% from 430 to 650 nm.

The other benefit for the proposed LED is that this approach can provide a way to produce a white light with a high CRI. Figure 5 shows the simulation results for four white lights with the color coordinates at the black body radiation. The CCTs are at 2800, 3500, 4500, and 6500 K,

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Table 1 The properties of the six organic fluorescent dyes.

No.	Name	Extinction coefficient (ε_A cm^{-1} M^{-1})	Quantum yield (Φ_A)	Emission peak wavelength (nm)	FWHM (nm)	Color coordinate (x, y)
1	3, 4, 9, 10-Perylenetetracarboxylic acid disodium salt	56,000	0.8	476	75	(0.1528, 0.3206)
2	Tetrachloro-substituted 3, 4, 9, 10-perylenetetracarboxylic acid disodium salt	55,000	0.55	481	50	(0.1344, 0.4185)
3	Tetrachloro-substituted 3, 4, 9, 10-perylenetetracarboxylic dianhydride	49,500	0.65	505	100	(0.3053, 0.5621)
4	Curcumin	54,000	0.3	536	102	(0.3582, 0.5695)
5	4-(Dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4 *H*-pyran	44,900	0.6	615	81	(0.6105, 0.3875)
6	Sulforhodamine 101 (S101)	139,000	0.9	631	78	(0.6873, 0.3087)

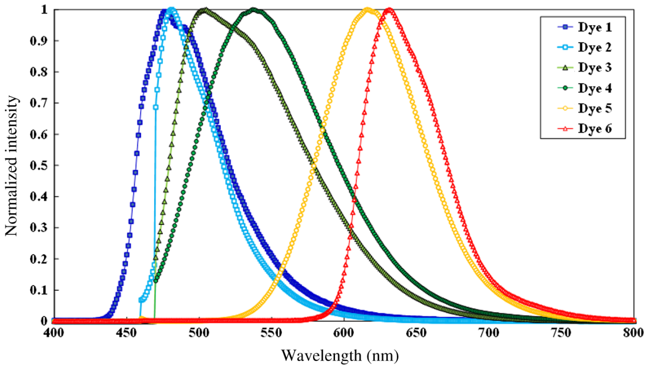


Fig. 1 The emission spectra of the six organic fluorescent dyes.

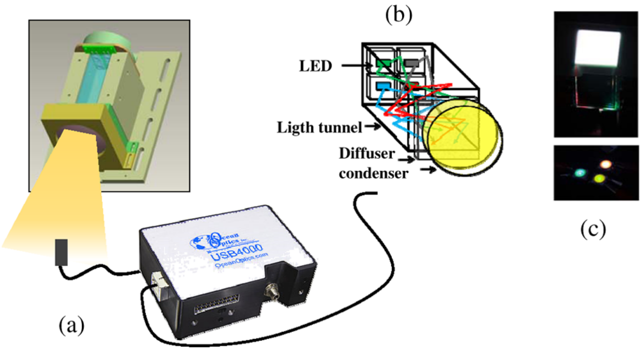


Fig. 3 (a) The geometry of the color-mixing cavity for producing white light. (b) The schematic diagram of the color mixing. (c) The photos of the single color LEDs and the emitted white light.

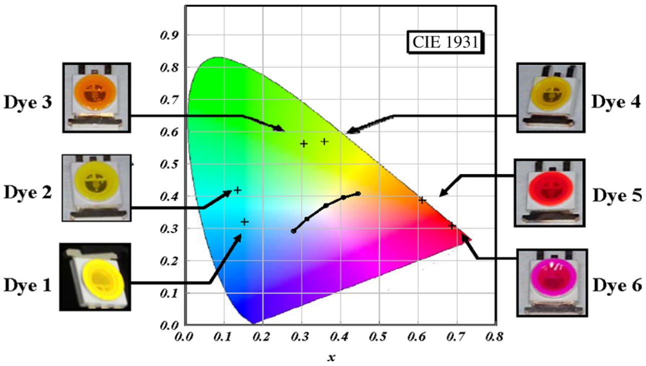
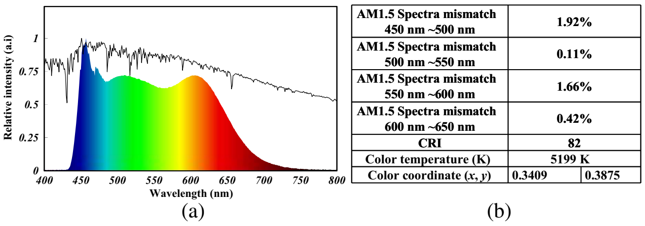


Fig. 2 The photo of the packaged light emitting diode (LED) with a single organic fluorescent dye and the corresponding color coordinates.



AM1.5 Spectra mismatch 450 nm ~500 nm	1.92%	
AM1.5 Spectra mismatch 500 nm ~550 nm	0.11%	
AM1.5 Spectra mismatch 550 nm ~600 nm	1.66%	
AM1.5 Spectra mismatch 600 nm ~650 nm	0.42%	
CRI	82	
Color temperature (K)	5199 K	
Color coordinate (*x*, *y*)	0.3409	0.3875

Fig. 4 (a) The spectrum of the quasisolar white light at the correlated color temperature (CCT) of 5200 K. (b) The table describing the spectra mismatch and color performance.

and the corresponding CRIs are as large as 97, 96, 94, and 90, respectively.

In summary, in this letter, we propose and demonstrate the capability of organic fluorescent dyes as new wavelength conversion materials. The LED is made with a blue die with a specific organic fluorescent dye to produce a single color appearance. With the use of a color-mixing cavity, the white light is demonstrated to produce a quasisolar spectrum at a specific band from 430 to 650 nm. In addition, white light with a high CRI can also be produced with different recipes, where the CRI is as high as 97 when the CCT is around 2800 K.

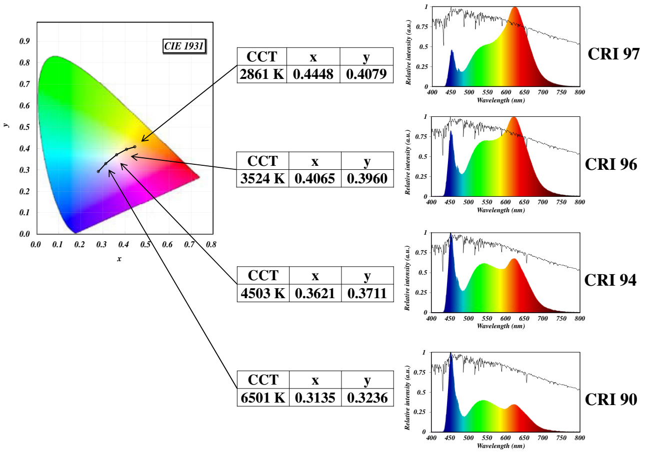


Fig. 5 The composite white lights at different CCTs and the corresponding color rendering indices.

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