High Color Rendering White LED Based on Dye-Bridged Siloxane Hybrid Encapsulation Mixed with Silicate Phosphor

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Abstract

White LED was fabricated using the thermally stable red dye-bridged siloxane hybrid (DBH) encapsulation with green silicate phosphor on the blue LED chip. The DBH encapsulation resin was synthesized by sol-gel condensation of the dye-bridged alkoxysilanes. The white LED was achieved by optimization of dye concentration and phosphor amounts in the encapsulation. The DBH/silicate phosphor based white LED shows high color gamut, color rendering index up to 90 and luminous efficacy of 33.3 lm W^{-1}. It is ensured to have long term thermal stability to be applied to solid-state lighting.

1. Introduction

Solid-state lighting has potential to reduce energy consumption and eco-friend illumination and display. White light-emitting diodes (LEDs) are main technology for solid-state lighting because of their efficiency and wide application area. Recently, tri-chromatic white LED (red/green phosphors and blue LED) is an issue because it has high color rendering index (CRI) and sufficient color temperature to be used in room lighting and display. But it is hard to select red phosphor among inorganic phosphor and quantum dot due to their high cost and stability problem.

We have reported dye-bridged hybrid materials (DBH)[1,2] that dyes are chemically bridged to oligosiloxane and it leads more stable characteristics. Dye has moderate cost and good optical properties to be used as wavelength converter, however, the stability problem may limit further researches. We have confirmed that photoluminescence of DBH was not decreased under 120 °C for hundreds hours. Also, photoluminescence intensity of DBH has less sensitivity to temperature to preserve initial characteristic on elevated temperature because internal molecular rotation is restricted by the chemical structure. Using red and green DBH, various thermally stable white LEDs were achieved.

In this study, red DBH has been synthesized as a wavelength converter of white LEDs for solid-state lighting. We have fabricated DBH based white LED mixing with green silicate phosphor. The DBH/silicate phosphor based white LED shows high color rendering index, color gamut, luminous efficacy and thermal stability to be used for solid-state lighting.

2. Experiment

For synthesis of red dye-bridged alkoxysilane, 4-diethylamino-2-(6-hydroxy-hexyloxy)-benzaldehyde, (2,6-dimethyl-4H-pyran-4-ylidene)malononitrile and 3-(triethoxysilyl)propyl isocyanate are used as precursor. Dye-bridged oligosiloxane was synthesized using the red dye-bridged alkoxysilane, 2-(3,4-epoxycyclohexyl)ethyl-trimethoxysilane and diphenylsilanediol via sol-gel reaction. We follow reference to synthesize of dye-bridged alkoxysilane and oligosiloxane.[3] The concentration of red dye-bridged alkoxysilane was controlled to have 0.1 mmol L^{-1} to 5 mmol L^{-1} in the oligosiloxane to match white emission. The silicate phosphor is mixed with dye-bridged oligosiloxane and the mixture is dispensed on blue LED and it was thermally cured at 150 °C for 2 h using hexahydro-4-methylphthalic anhydride and tetrabutyl-phosphonium methanesulfonate as hardener and initiator, respectively. The fabrication scheme is represented in Fig 1.

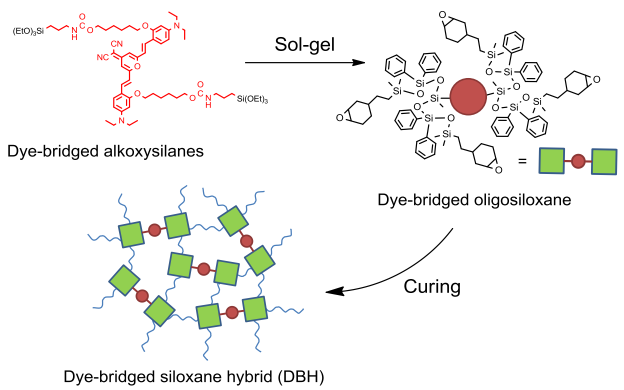

Fig 1. Fabrication scheme of dye-bridged siloxane hybrid encapsulation by sol-gel reaction and thermal curing.[1]

3. Results and discussion

DBH/silicate phosphor white LED has advantage on controlling color temperature and CRI by varying the dye concentration of DBH and the white LEDs can be fabricated in purpose. Color temperature is easily turned from cool white (7600K) to warm white (2670 K) using DBH/silicate phosphor to have merit on lighting application. When 10 wt% of silicate phosphor is dispersed in DBH and the red dye concentration is 0.05 mmol L^{-1}, it shows ideal emission that the DBH/silicate phosphor based white LED has color temperature of 5400 K and white point is at (0.3341, 0.3237) on Commission Internationale de l'Eclairage (CIE) 1931 color space (Fig 2). Comparing to commercial white LED based on inorganic phosphor, it has much higher color rendering that CRI is up to 90. The commercial white LED has di-chromatic source and CRI is 64. Fig 3 shows comparison of electroluminescence (EL) spectra of DBH/silicate phosphor based white LED and commercial white LED. The emission in the region of red and green of DBH/silicate phosphor based white LED is stronger than inorganic phosphor based white LED. By reinforcing the red and green color, it can achieve high CRI.

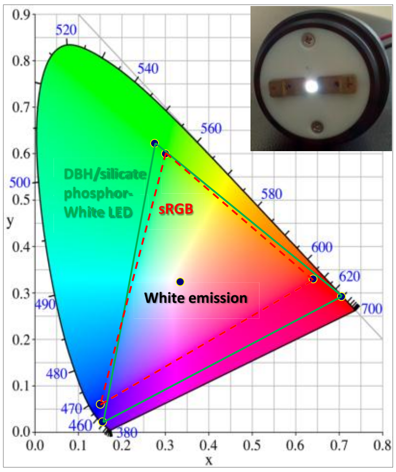

Fig 2. CIE color coordinates of the white LEDs based on DBH/silicate phosphor (Inset: Photograph of the white LED).

The DBH/silicate phosphor based white LED has large color gamut because each RGB components has vivid color. RGB color coordinates of DBH, silicate phosphor and blue LED are (0.705, 0.292), (0.275, 0.622) and (0.155, 0.022), respectively. The area of RGB color space covers 133% of a sRGB color space which is the most commonly used RGB color space for monitors and HDTVs. Thus, DBH/silicate phosphor based white LED can express more wide color space and it is applicable to display application.

DBH/silicate phosphor based white LED has sufficient luminous efficacy that the measured luminous efficacy of DBH/silicate phosphor based white LED is 33.3 lm W^{-1} at 20 mA.

The DBH/silicate phosphor based white LED was thermally aged at 120 °C in an air atmosphere for 800 hrs to confirm thermal stability. Fig 4 shows EL spectra of the fabricated DBH/silicate phosphor based white LED before and after thermal aging. The spectra of DBH/silicate phosphor based white LED have been maintained during 800 hrs. Dye molecule is chemically bonded to the robust siloxane structure and the internal molecular rotation is restricted and it would prevent thermal decomposition. And silicate phosphor is protected by dense siloxane network which has low gas permeability from moisture and it would lead more stable characteristics. These results indicate that DBH/silicate phosphor based white LED can properly operate in elevated temperature.

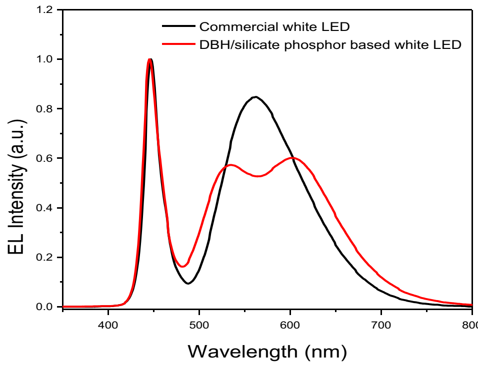

Fig 3. Comparison of EL spectra of commercial white LED and DBH/silicate phosphor based white LED.

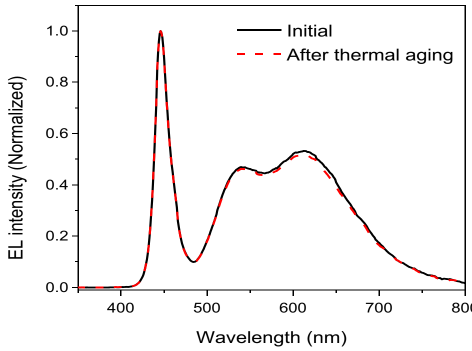

Fig 4. EL spectra of DBH/silicate phosphor based white LED before and after thermal aging.

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