Luminescent Properties of High Color Rendering Powder Electroluminescent Devices

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Abstract

A powder electroluminescent (EL) device can be fabricated by a simple printing process. The present applications of these devices are restricted to providing low-intensity backlighting. In order to realize a lighting system, it is necessary to develop EL devices that have white luminescence. It was believed that it is difficult to realize white luminescence in powder-type inorganic EL devices. However, we have produced high-luminance organic-dye-dispersed powder EL devices. Moreover, we have produced high color rendering white-light-emitting organic-dye-dispersed powder EL devices that consist of the three primary colors. The operating conditions for the organic-dye-dispersed powder EL devices are the same as those for a conventional powder EL device. The film thicknesses of both the dielectric and phosphor layers were approximately 30 µm. We have manufactured a novel white-light-emitting powder EL device with a new hybrid structure by doping the phosphor layer with organic fluorescent dyes.

Introduction

A powder EL device can be developed by a printing process [1] and used to economically realize field luminescence by a simple process. Presently, its applications are limited to providing low-intensity backlighting, for example, as a key backlight component in cellular phones, backlight for a liquid crystal display (LCD) in wristwatches, and as a backlight in point-of-purchase (POP) advertisements. In order to realize a lighting system, it is necessary to develop EL panels that exhibit white luminescence. The examination of the color rendering properties in an inorganic EL film containing a distributed rhodamine-type dye in a polymer [2] has been carried out. Therefore, it is believed that it is difficult to realize white luminescence in a powder EL device.

Organic light-emitting devices (OLEDs) are considered to be one of the most promising next-generation flat-panel displays and/or lighting systems because of their advantages such as self-emission, simple structure, wide viewing angle, and fast response speed. In the research field of OLEDs, the dye-dispersion (a guest-host system) method is widely used for obtaining high color purity and high luminance [3]. With regard to the increase in the electroluminescence efficiency of OLEDs, it is well known that the luminosity can be increased by doping Alq3 with Coumarin (an organic fluorescent dye). Thus far, we have produced high-luminance [4] and red luminance [5] organic-dye dispersed powder EL devices. Furthermore, we have produced white-light-emitting organic-dye-dispersed powder EL devices that consist of the three primary colors, implying that it is possible to realize a lighting system.

Experiment

The organic-dye-dispersed powder EL device developed in this study has a layered structure that is fundamentally the same as that of a conventional inorganic powder EL device. Only the phosphor layer is different in the developed device. The device structure is laminated on a transparent, electrically conductive film substrate with the following layers in the given order: a phosphor layer co-doped by organic fluorescent dyes, a dielectric layer, and a back electrode. Three types of organic dyes were used: the green dye 3-(2-benzothiazolyl)-N,N-diethylumbelliferylamine (Coumarin 6) and the red dyes 4-(dicyanomethylene)-2-i-propyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTB) and 4-(dicyanomethylene)-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM). The absorption and emission spectra of these organic dyes are shown in Figure 1, Figure 2, and Figure 3, respectively. The devices were produced by using a screen printer that ensured reproducible results. The phosphor layer used is a ZnS system (ZnS_{sys}) that comprises a granular phosphor. The phosphors were dispersed in a high-dielectric polymer along with the organic fluorescent dyes and then converted into a paste. The dielectric paste containing $BaTiO_3$ as the main component was used as the dielectric layer, while a silver paste with Ag as the main component was used as the back electrode. The dielectric and silver pastes were coated onto an ITO substrate.

An EL measurement system (SX-1111: Iwatsu Test Instruments) was used to evaluate the electrical properties of the developed EL device. The spectrometer used in the study was calibrated by using an LS-1-CAL lamp calibration halogen lamp (Ocean Optics), and the integration value for each spectrum was proportional to the energy value.

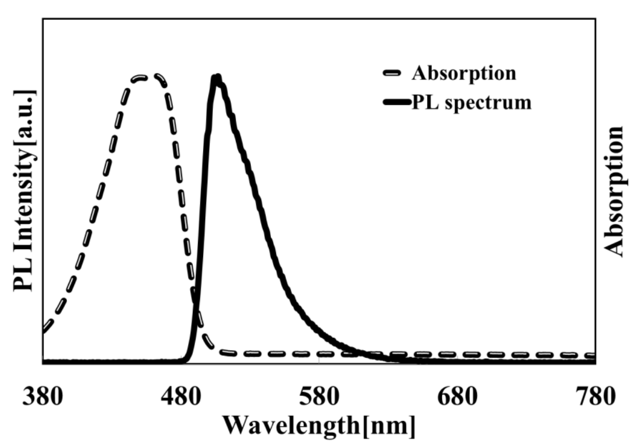

***Figure 1** Absorption and photoluminescence (PL) spectra of Coumarin 6*

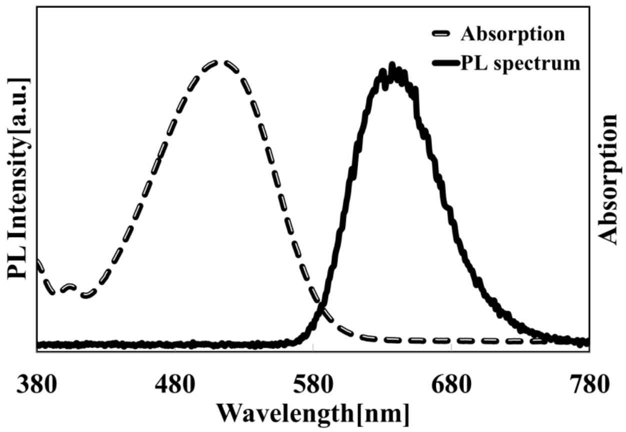

Figure 2 Absorption and photoluminescence spectra of DCJTB

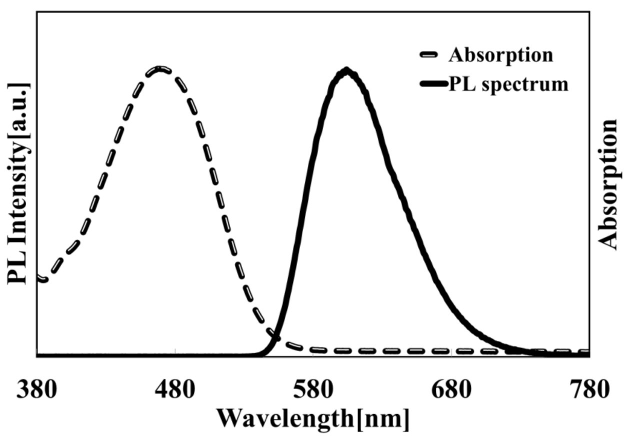

Figure 3 Absorption and photoluminescence spectra of DCM

The operating conditions for the organic-dye-dispersed powder EL device were considered to be the same as those of the conventional powder EL device. The luminosity of the powder EL device was different for different operating conditions because of the dependence of the thickness of the device film on the impressed electromotive force and frequency. Device characteristics were measured for a sine wave input with a frequency range from 1 to 15 kHz.

Results and Discussion

The frequency dependence of the emission spectrum for the white-light-emitting organic-dye-dispersed powder EL device is shown in Figure 4. Figure 5 shows the absorption and EL emission spectra of each material. The emission wavelength of the original inorganic ZnS_{sys} phosphor exhibits a peak at 455 nm. When the organic dyes are mixed with the inorganic phosphors at mixed weight ratio of Coumarin 6 to DCJTB of 3:1, the electroluminescence emission spectrum exhibits peaks at approximately 445, 510, and 625 nm. This result originates in the material in which the organic dye is excited by the fluorescent energy of the inorganic phosphor. First, the inorganic phosphor emits light. Then, part of the luminescent energy of the inorganic phosphor excites Coumarin 6, and it emits light. Furthermore, part of the luminescent energy of Coumarin 6 excites DCJTB, and it emits light. As a result, white luminescence consisting of the three primary colors is realized.

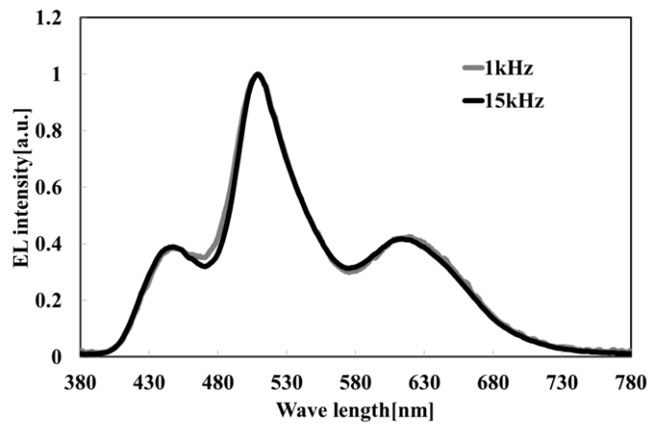

Figure 4 Emission spectrum of a white-light-emitting organic-dye-dispersed powder EL device

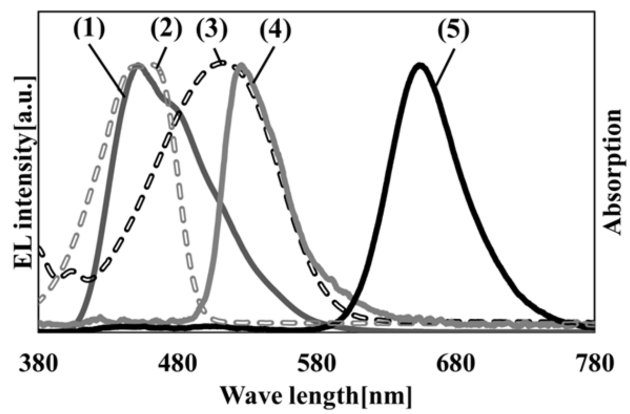

Figure 5 Absorption and EL emission spectra
(1) EL emission spectrum of the ZnS system phosphor
(2) Absorption spectrum of Coumarin6
(3) Absorption spectrum of DCJTB
(4) EL emission spectrum of Coumarin 6 accelerated by the emission of the ZnS_{sys} phosphor
(5) EL emission spectrum of DCJTB

The parameters for the color rendering properties of the white-light-emitting device are listed in Table 1. We have manufactured the novel white-light-emitting organic-dye-dispersed powder EL device with a new hybrid structure by doping the organic fluorescent dyes in the phosphor layer. We observed that the organic-dye-dispersed powder EL device exhibits white luminescence for the color coordinates x = 0.31 and y = 0.33, without a color filter [6]. This remarkable increase in the purity of white luminescence is achieved by co-doping the phosphor layer with Coumarin 6 and DCJTB. This novel white-light-emitting organic-dye-dispersed powder EL device with peaks at 440, 508, and 625 nm in the electroluminescence emission spectrum is developed using a printing technique.

Table 1 Color rendering index and color temperature for the organic-dye-dispersed powder EL devices

		Two organic dyes	Three organic dyes
General color rendering index	Ra	91	97
Special color rendering index	R9	87	95
	R10	85	98
	R11	85	86
	R12	82	96
	R13	90	98
	R14	96	98
CIE chromaticity coordinates		(0.30, 0.33)	(0.32, 0.34)
Correlated color temperature (K)		7018	6220

We consider the color conversion of the organic-dye-dispersed powder EL device. The device with blue luminescence is a conventional organic-dye-dispersed powder EL device that consists of the ZnS_{sys} phosphor and Coumarin 6 [4]. A conventional powder EL device with red luminescence has not been reported thus far. With regard to an increase in the electroluminescence efficiency of the OLEDs, it is well known that the luminosity can be increased by doping Alq3 with Coumarin [3].

Although Coumarin 6 was present, a peak at approximately 520 nm was not observed. Owing to the doping of the phosphor layer with Coumarin 6 in the first stage, the electroluminescence spectrum marginally shifted towards a higher wavelength, and the peak of the electroluminescence emission spectrum shifted from 450 nm to 520 nm. Furthermore, the peak of the electroluminescence emission spectrum shifted from 520 nm to 650 nm owing to the doping of the phosphor layer with DCJTB in the second stage. It was believed that all the excitation energy was transferred from the inorganic phosphor to the organic fluorescent dyes. Therefore, Coumarin 6 functioned as an emission-assisting dopant, without emitting light. This phenomenon is different from color conversion by means of an optical filter.

Figure 6 shows that the organic-dye-dispersed powder EL device using two organic dyes has the lowest luminescence intensity around 480 nm and 580 nm. In order to increase the luminescence intensity near 580 nm, a device additionally doped with DCM that has an emission spectrum peak near 605 nm in addition to the two organic dyes was examined. In comparison, the device using three organic dyes has an increased luminescence intensity near 580 nm in the red-light region due to the effects of red organic dye DCM. This device also exhibited a shift to 605 nm from 618 nm because there was no emission spectrum peak wavelength for the device that used two organic dyes. Further, there was no difference in the blue-light and green-light regions, which is clear from the PL peak value for DCM of 605 nm.

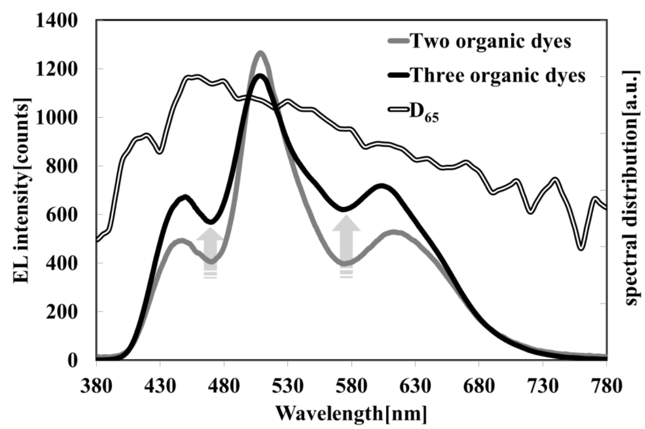

Figure 6 *Emission spectra for the organic-dye-dispersed powder EL devices*

The organic-dye-dispersed powder EL device has high color purity as compared to the conventional powder EL device. This novel white-light device based on an organic-dye-dispersed powder EL device was developed using a printing technique, without a color filter.

Conclusion

Novel white-light-emission with a high color rendering index of Ra = 97 and an EL spectrum that was close to the light intensity distribution of Standard Illuminant D_{65} was observed for the device fabricated using two organic dyes: Coumarin 6 and DCJTB (color rendering index Ra = 91). The resulting CIE xy chromaticity coordinates (0.32, 0.34) correlated with a color temperature of 6220 K for the device fabricated using three organic dyes: Coumarin 6, DCJTB, and DCM.

Thus, the color can be converted into high color rendering easily by doping the phosphor layer with an organic dye. It can be expected that a wide range of applications are available for these devices having a wide color reproduction range along with white-light emission.

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Author Biography

Toshifumi Satoh received his B.S. in engineering from the Meiji University, Japan (1984) and his Ph.D. in mechanical engineering from the Meiji University, Japan (1995).

He worked at Panasonic before moving to Tokyo Polytechnic University, where he is engaged in the research of thin films and flat-panel displays. He is now a Professor in the Department of Media and Image Technology. His fields of interests include printable devices and lighting systems. Dr. Satoh is the president of the Japanese Society of Printing Science and Technology.