Development of Multi-Boron Embedded Delayed Fluorescence Materials with Narrowband Emissions

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https://hdl.handle.net/2324/6787562

出版情報:九州大学,2022,博士(工学),課程博士 バージョン: 権利関係:やむを得ない事由により本文ファイル非公開(3)

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- 論 文 名 : Development of Multi-Boron Embedded Delayed Fluorescence Materials with Narrowband Emissions (狭帯発光を示す含ホウ素遅延蛍光材料の創製)
- 区 分 :甲

論文内容の要旨

Boron-doped multi-resonance thermally activated delayed fluorescent (MR-TADF) emitters fascinated the researchers with their unprecedented luminescent property and excellent performance in application of organic light-emitting diodes (OLEDs). Although the MR-TADF emitters have potentially overcome the major obstacles of state-of-the-art D–A-type TADF emitters, they are suffering from two main drawbacks which may affect their commercialization in OLEDs. Firstly, MR-TADF usually exhibited slow reverse intersystem crossing rate (k_{RISC}), resulting in severe efficiency roll-off and poor operational stability. Moreover, it is still difficult to realize ultrawide color gamut emitters covering deep blue to deep red. In this thesis, I have provided the several different design strategies to solute these problems.

In Chapter 2, full-color and narrowband MR-TADF emitters covering the entire visible region from blue to red have been firstly demonstrated. Wide-range emission color (or bandgap) tuning can be achieved via systematic electronic modulation by combining boron atom and carbazole units in different numbers and substitution positions. In dilute solutions, these compounds exhibited full-color photoluminescence (PL) from deep blue to deep red ($\lambda_{PL} = 466-615$ nm) with full-width at half maxima as small as <40 nm and absolute PL quantum yields as high as 85%–98%. The transient PL decay measurements revealed the TADF characteristics. Owing to their ideal emission properties, we achieved, for the first time, full-color and narrowband organic light-emitting diodes based on this family of emitters. Notably, all devices showed impressively high maximum external electroluminescence quantum efficiencies (η_{ext}) of 22%–32%.

In Chapter 3, two new green MR-TADF emitters p-CzB and m-CzB were designed and synthesized employing a wise dimerization approach. The interconnection position between the two parent cores served as a conjugation value for regulating the physicochemical properties of the materials. The design retained MR-TADF characteristic for materials similar to their parent core with concurrently achieving pure green emission, $\lambda_{PL} \sim 505-515$ nm and narrow emission spectra. Indeed, the m-CzB exhibited high k_{RISC} and short delayed lifetime (τ_d) compared to its analogue p-CzB. As a result, the m-CzB based OLED device outperformed over the p-CzB by demonstrating high η_{ext} of 23.5% and pure green CIE(x,y) coordinates of (0.20, 0.70). It is worth to mention that the CIE coordinates of the m-CzB precisely matched with the green coordinates set by the NTSC. Importantly, the m-CzB. In Chapter 4, by using the substitution of O and S atoms into B/N framework, five long-wavelength emissive MR-TADF emitters were designed and synthesized. Compared to asymmetrical isomers (**BN-Y** and **BO-G**), the photophysical analysis proved that B- π -B and N- π -N conjugation of symmetrical **BN-R** and **BO-O** can effectively reduce energy gap and generates bathochromic shift in two symmetrical molecules, **BN-R** and **BO-O**. This design strategy realized a wide color tuning ranging from green to red with extreme narrow FWHM (~19–22 nm). In addition, long-wavelength emissive OLEDs with η_{ext} as high as 12.2–21.6 % and superb color purity could be fabricated by using this family of MR-TADF emitters.

In Chapter 5, the dissertation is summarized, and perspectives are discussed.

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