Device Analysis of Thermally-Activated Delayed Fluorescence-Based Organic Light-Emitting Diodes Aimed for High Device Stability

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https://doi.org/10.15017/4060120

出版情報：九州大学, 2019, 博士（工学）, 課程博士
バージョン：
権利関係：

（様式２）

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論 文 名　：　Device Analysis of Thermally-Activated Delayed Fluorescence-Based Organic Light-Emitting Diodes Aimed for High Device Stability
（熱活性化遅延蛍光分子を用いた有機発光素子の長寿命化に向けたデバイス解析）

区　　分　：　甲

論　文　内　容　の　要　旨

In Chapter 1, the background and motivation of this thesis are introduced. Organic light-emitting diodes (OLEDs) based on thermally-activated delayed fluorescence (TADF) emitters exhibiting nearly 100% of internal quantum efficiency (IQE) are expected for display and lighting technologies. However, the device lifetime of TADF-OLEDs is still shorter than those of conventional fluorescent and phosphorescent OLEDs. To spread TADF-OLEDs widely for commercial applications, the device stability of TADF-OLEDs must be improved. In this study, the research focused on the understanding of origins of OLED degradation and molecular orientation in films to develop highly efficient and stable TADF-OLEDs. Better understanding of the mechanisms can suggest the strategies to achieve ultimate OLED performance.

In Chapter 2, the device characteristics of the OLEDs based on penta(9H-carbazol-9-yl)benzonitrile (5CzBN) and 2,4,6-tris(9H-carbaozle-9-yl)-3,5-bis(3,6-diphenylcarbazole-9-yl)benzonitrile (3Cz2DPhCzBN) having similar molecular structures but significantly different delayed fluorescence lifetimes (τ_ds) were carefully considered. The device based on 3Cz2DPhCzBN with a shorter τ_d than that of 5CzBN showed a significantly long device lifetime under constant current operation. Simple observations of the change of electroluminescence (EL) spectra in the degradation processes suggested the shift of the carrier recombination site toward the hole-blocking layer (HBL)-side and hole-injection into the HBL under continuous operation. The 3Cz2DPhCzBN-based OLED showed the smaller spectral changes than those of the 5CzBN-based OLED. Furthermore, carrier-transport stabilities of the emission layers (EMLs) were investigated. I revealed that triplet exciton-polaron interaction (TPI) is one of dominant channels of the degradation of TADF-OLEDs. Because of its short τ_d, 3Cz2DPhCzBN resulted in low triplet-exciton density and a suppressed rate of TPI in the device. Therefore, the improvement of the RISC characteristics of TADF molecules and the control of carrier transport properties of EML aimed for the suppression of TPI are significantly needed for improvement of a device lifetime.

In Chapter 3, exciton dynamics in TADF-OLEDs under operation were investigated by magnetic field effects (MFEs) in the devices. Magnetic responses of OLED characteristics could be expected to use as a method to prove the manner of exciton annihilation processes such as TPI as suggested in Chapter 2, because separation of degenerate triplet states by a magnetic field should affect the device performance and reflect the exciton dynamics in devices. Thus, I compared magnetic-field-modulated EL (MEL) profiles of TADF-OLEDs based on various emitters and revealed that the profiles contain two origins of MFEs such as polaron-pair (PP) and TPI models. Further, the MEL amplitudes based on TPI model depended on emitters' τ_d that clearly suggests that emitters with long τ_ds suffer from more significant annihilation induced by TPI than the emitters with short τ_ds. Furthermore, for an analysis of degraded OLEDs, I identified the exciplex-formation on a film interface as one of the origins of a change in MEL profiles because the device aging affects carrier transport properties in an EML. Therefore, as same as the conclusion in Chapter 2, τ_ds of TADF molecules and a control of carrier transport properties are important for improvement of device stability.

In Chapter 4, molecular orientations in organic guest: host films based on disk-shaped TADF molecules were investigated. Because the orientation is one of limiting factors of external quantum efficiency of OLEDs, the control of emitters' transition dipole moment (TDM) orientation to enhance light-outcoupling efficiency (η_{OC}) is necessary for improvement of OLEDs' emission efficiency and a decrease of driving current that results in the elongation of the device lifetime. I revealed that materials' glass transition temperature (T_g) and the polarization of a host molecule are ones of key factors to control the emitter's molecular orientation. For the nonpolar TADF molecules, a high-T_g host molecule can easily fix the molecular motion of emitter molecules, and achieve the highly horizontal emitter's TDM orientation. For polar emitter molecules, the polarization of a host molecule disturbs intermolecular dipole-dipole interactions between emitter molecules by the formation of the interaction between host and emitter molecules. Further, I demonstrated perfectly horizontal TDM orientation and η_{OC} of around 30% of an OLED based on a TADF molecule: host film. These results provide a possibility to achieve an ultimate OLED performance with the combination of 100% of IQE, extremely high η_{OC} and high device stability of the OLEDs based on TADF emitters.

In Chapter 5, the summary of this thesis and prospects are introduced.