Exciton harvesting and multiplication in near-infrared organic light-emitting diodes

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（励起子増感過程を用いた高効率近赤外有機 EL 素子の創成）

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論　文　内　容　の　要　旨

The purpose of this thesis is the improvement of the overall external EL quantum efficiency (η_{EQE}) of near-infrared organic light-emitting diodes (NIR-OLEDs) and the establishment of comprehensive material and device design rules for efficient NIR-OLEDs. This thesis is organized as follows.

In Chapter 1, the background and motivation of the thesis are described. In recent years, NIR-OLEDs have attracted significant interest especially due to their potential application such as bio-imaging and bio-sensing for the daily healthcare. The η_{EQE} of rare-metal-free NIR-OLEDs, however, have been still limited due to the lack of exciton harvesting and multiplication process in NIR-OLEDs. Furthermore, since the energy gap law dictates the acceleration of a non-radiative decay process in NIR region, careful molecular design strategies including the suppression of reorganization energy are required for developing highly emissive NIR-emitting molecules.

In Chapter 2, the utilization of a triplet harvesting process, *i.e.*, thermally activated delayed fluorescence (TADF) processes was demonstrated for the enhancement of exciton production efficiency (η_Y) in NIR-OLEDs. Using a TADF material which emit in the visible region, as an energy donor to NIR-dyes, the NIR emission originated from a reverse intersystem crossing (RISC) process in the TADF host matrix was achieved. In the OLED characterization, η_{EQE} of the TADF host-based NIR-OLEDs is about 6 times higher than that of the conventional fluorescent host-based NIR-OLEDs due to the harvesting of electrically generated triplet excitons through an efficient RISC process on the TADF host. Further, the intrinsic bipolar charge-transporting ability of TADF materials consisting of the electron-donating and electron-accepting moieties is experimentally revealed.

In Chapter 3, a singlet fission process was focused as the exciton multiplication process, and utilized for NIR luminescence. From the magnetic field dependence of the visible and NIR luminescence intensities, the realization of η_Y above the theoretical upper limit of 100% by exploiting of a singlet fission process in NIR-OLEDs was demonstrated.

In Chapter 4, a novel NIR-TADF molecule with a rigid electron-accepting unit was developed for achieving high photoluminescence quantum yield (φ_{PL}) at 700~1000 nm, useful for biological sensing and imaging applications. The newly developed NIR-TADF molecule showed an

emission peak at 729 nm in a 10wt%-doped film and its φ_{PL} exceeded 40%. The NIR-TADF molecule also functioned as an efficient energy donor for the NIR fluorescent molecule emitting around 900 nm. The NIR-OLEDs using the co-deposited film as an emitting layer showed the η_{EQE} values of 13.4% with EL peak at 734 nm and 1.1% with EL peak at 901 nm, which are the highest η_{EQE} reported for NIR-TADF-OLEDs in each wavelength. Furthermore, by integrating the developed NIR-OLEDs with a conventional organic photodiode (OPD), "all-organic" PPG sensing both in the 700 nm and 900 nm region are realized.

In Chapter 5, the conclusions of this work and the future perspectives for highly efficient NIR-OLEDs are described.

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