Development of deep-red and near-infrared organic luminescent materials and their applications in organic light-emitting diodes and continuous-wave organic semiconductor lasers

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論　文　内　容　の　要　旨

This thesis mainly focuses on the molecular design, synthesis, and properties of highly efficient deep-red and near-infrared (NIR) organic luminescent materials toward organic light-emitting diodes (OLEDs) and continuous-wave (CW) organic solid-state lasers (OSSLs) applications.

OLEDs display are replacing the traditional liquid-crystal display with organic compounds and have additional advantages such as simple structure, self-emission, and lighting precision. Such properties give OLEDs the potential to be widely used to wearable devices for health-care, audio systems, and vehicle equipment. Organic luminescent materials also offer the prospect of low-temperature solution processing to enable the roll-to-roll mass printing of large-area, integrated modules, and reducing costs. Compared with OLEDs, the light from organic lasers has high spatial coherence and can generate a directional light beam with divergence, high brightness, and good spectral control.

The state-of-the-art and challenges for OLEDs and OSSLs are described in Chapter 1. The research in recent years on organic luminescent materials has extended the spectrum from visible to deep-red and NIR because of both the academic and the industrial sector for their potential in display, medical, sensor, and optical communication, which make them become a new target in the field of OLEDs and OSSLs research and development. According to this trend, this thesis undertakes the study of various factors affecting the characteristics of OLEDs and OSSLs to advance the development of high efficiency deep-red and NIR organic luminescent materials and their applications on OLEDs and CW OSSLs.

Deep-red amplified spontaneous emission (ASE) from an squaraine (SQ) derivative (*cis*-DCSQ1) by using a host-guest system to suppress aggregation and induce efficient energy transfer is demonstrated in Chapter 2. At a *cis*-DCSQ1 doping concentration of 0.5 wt%, the lowest ASE threshold is 10 μJ/cm^2 at 687 nm. However, the rapid degradation is occurred under laser pumping and UV irradiation. A good thermal and photochemical stability should be considered for further advanced molecular design.

In Chapter 3, a novel dimeric borondifluoride curcuminoid dye is specifically designed and synthesized because of the good electron withdrawing properties of acetylacetone borondifluoride appended in meso position and excitonic coupling effects which can take place in such a molecular system. Steady-state and time-resolved PL measurements demonstrate that this dye exhibits highly efficient NIR thermally activated delayed fluorescence (TADF) emission with a quantum yield which can be as high as 45% in the doped thin

film for a maximum emission wavelength of 760 nm. NIR TADF OLEDs with a maximum external quantum efficiency of 5.1% are fabricated, which exceeds the performances typically obtained in devices based on conventional NIR fluorescent emitters. In addition, ASE with a maximum emission wavelength, which can be tuned between 801 and 860 nm, and a threshold as low as 7.5 μJ/cm^2 is observed. These results provide additional evidence that borondifluoride curcuminoid derivatives are promising candidates for NIR OSSLs.

In Chapter 4, the exciton-exciton process of borondifluoride curcuminoid derivative films aimed for lasing is studied. The mixed-order distributed feedback could realize low lasing threshold of about 1.28 μJ/cm^2 and the narrow full width at half maximum of about 1.4 nm. The no triplet excited-state absorption at ASE and lasing wavelengths minimize the triplet absorption losses in the resonator. Superior surface emitting lasing from the devices under quasi-CW operation is obtained with the emission intensity and lasing threshold being dependent of the pulse width. For laser devices studied in Chapter 4, the maximum duration time is 100 s and the lowest lasing threshold is about 1.28 μJ/cm^2. However, others effect such as charge carrier mobility and Joule heating etc., are also the important factors and will need further investigation for the realization of current driven NIR organic semiconductor laser diodes (OSLDs).

Finally, the summary of this thesis and future strategy are introduced in Chapter 5. To design further red-shifted NIR materials, energy-gap tuning should be considered. By increasing the donor ability, the conjugation can be extended, leading to the shallowing of the HOMO level, and decreasing the energy-gap to further NIR emission. Further, the introduction of a new NIR functional group should also be considered to develop advanced NIR lasing materials. Triplet accumulation also needs to be inhibited to enable CW operation of organic lasers. By considering above conceptual ideas, the author expects more and more organic luminescent materials will be developed for high efficiency deep-red and NIR OLEDs and low lasing threshold to realize the OSLDs in the future.