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Thesis Outline

High-Performance Organic Field-Effect Transistors: Additive Strategy and Memory Application

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In chapter I, the research background of organic field-effect transistors (OFETs) was introduced, including the overview of OFETs, additive strategy in OFETs and OFET memory.

In chapter II, a new DPP-based conjugated polymer, PDPPy-BDD, was designed and synthesized considering the ambipolar charge transport property of the OFETs. The OFETs based on the neat PDPPy-BDD showed well-defined V-shaped ambipolar transport characteristics with the hole and electron mobilities of 3.5×10^{-3} and 3.07×10^{-2} $cm^2\ V^{-1}\ s^{-1}$, respectively. It was found that the organic salt tetramethylammonium iodide (TMAI) can enhance both the hole and electron mobilities (5.5×10^{-3} and 3.47×10^{-2} $cm^2\ V^{-1}\ s^{-1}$, respectively) of the OFETs at the PDPPy-BDD:TMAI ratio of 48:1. Systematic characterizations revealed that the addition of a small amount of TMAI has a significant effect on the microstructure of the PDPPy-BDD thin film, and this was the main reason for the enhanced mobilities.

In chapter III, two dual-acceptor polymers based on DPP and bithiophene imide, FuI and SeI, were studied. The OFETs based on the pristine FuI polymer showed ambipolar transport characteristics with the hole and electron mobilities of 3.3×10^{-3} and 9.2×10^{-3} $cm^2\ V^{-1}\ s^{-1}$, respectively. While the pristine SeI-based OFETs exhibited higher hole and electron mobilities of 0.025 $cm^2\ V^{-1}\ s^{-1}$ and 0.122 $cm^2\ V^{-1}\ s^{-1}$, respectively, the charge carrier polarity flowing along the conducting channel of the OFETs could be directionally tuned by structure design. Specifically, with the TBAI additive, the charge carrier polarity of FuI-based OFETs could be changed from n-type dominant (μ_e/μ_h = 2.79) to more balanced features (μ_e/μ_h = 0.71-1.47), while the almost unipolar n-type property was obtained for the SeI polymer by adding the TBAI additive with the μ_e/μ_h changing from 4.9 to 264. Moreover, the electron mobility of SeI-based OFETs reached as high as 0.154 $cm^2\ V^{-1}\ s^{-1}$ through the tetrabutylammonium iodide (TBAI) additive strategy. It was demonstrated that the incorporation of TBAI does not change the molecular packing pattern, but

significantly affects the energy levels and contact resistance in OFETs, indicating that both channel doping and contact doping are demonstrated in this study.

In chapter IV, by introducing the quinoid pyrazine unit into DPP-based polymers, three new polymers, namely, FuPz, SePz, and PyPz, were successfully synthesized. Three additives (TBAI, TeNF, and $Zn(C_6F_5)_2$) were doped to the channels of the OFETs. It was for the first time demonstrated that a single ambipolar polymer (PyPz) with a high LUMO hybridization is realized by efficient doping with any of the selected p-type and ionic dopants. In contrast, both p-dominant FuPz and SePz negatively responded to any dopants. The doping effect was more pronounced for electron transport with an increment ($\Delta\mu_e$) of nearly 400%, approaching 1.0 cm^2 V^{-1} s^{-1} in the case of PyPz with 3 mol% TeNF. Density functional theory (DFT) calculations indicated that highly hybridized LUMOs favor effective orbital couplings with any dopants, leading to efficient doping. Control polymers with lowered electron affinities showed too negative electrostatic potential values to form LUMO hybridization for effective orbital coupling with dopants, resulting in inefficient doping. The results indicate that highly hybridized LUMOs for effective orbital couplings could be a normalized figure of merit in the doping study of ambipolar organic semiconductors (OSCs) with various dopants, disregarding the diverse mechanisms of various dopants and conflicts of p- and n-doping in a single OSC.

In chapter V, the tunable high-performance OFET memories were successfully developed by using solution-processed crosslinked electret dots/semiconductor bilayer structures. The memory performance of polymer electret dots incorporating p-type (PBTTT-C14) or n-type (N2200) semiconductors showed large memory windows and high charge density, with non-volatile characteristics of 10^4 s retention time and a high memory ON/OFF current ratio over three orders of magnitude. In addition, reliable endurance performance in excess of 100 cycles without obvious decay could be realized. Finally, the related memory mechanism was mainly investigated by Kelvin probe force microscopy analysis.

In chapter VI, the general conclusion and outlook were given based on the above studies.