Functional Device Engineering in Organic Field-Effect Transistors for Sensing and Neuromorphic Applications

Abstract

Organic field-effect transistors (OFETs) emerged as promising platforms for next-generation electronics due to the intrinsic advantages of organic semiconductors, such as biocompatibility, low-cost fabrication, mechanical flexibility, and chemical tunability. However, the practical implementation of these devices beyond switching applications is often limited by weak signal transduction, interfacial charge trapping and the absence of intrinsic memory and adaptive learning functionality. This dissertation addresses these challenges by systematically investigating target-specific device engineering strategies, with particular emphasis on dielectric and interface engineering to enhance the functional performance of OFETs for sensing and neuromorphic applications.

The thesis starts with a fundamental discussion of charge transport in OFETs, highlighting the dominant roles of dielectric-semiconductor and electrode-semiconductor interfaces in controlling device performance. Motivated by this foundation, the experimental chapters demonstrate application-specific dielectric and interface modifications to convert OFETs into a multifunctional platform. In the first experimental study, ultraviolet-ozone (UVO) treatment was utilised to introduce polar functional groups at the interface. This controlled interfacial modification enhances electron trapping, leading to significant improvements in photosensitivity, responsivity, and specific detectivity in DNTT-based organic phototransistors. This thesis further explores chemical sensing studies by designing a hybrid inorganic-organic channel architecture, through decoration of MoS_2-nanosheets in pentacene OFET for ammonia (NH_3) gas detection. Although the incorporation of MoS_2 nanosheets modifies the band alignment and reduces field-effect mobility, the hybrid devices exhibit markedly enhanced

sensing performance, including improved sensitivity, faster response and recovery, and reliable detection at low concentrations. The last experimental study demonstrates neuromorphic functionality through the incorporation of the metal-organic framework zeolitic imidazole framework (ZIF67) into a PMMA dielectric matrix. The synthesised composite dielectric enables resistive switching in two-terminal devices and synaptic plasticity in three-terminal devices. The OFET device successfully mimics the major biological synaptic behaviours, including spike-duration-dependent plasticity (SDDP), spike-rate-dependent plasticity (SRDP), adaptive learning and forgetting dynamics.

Altogether, this thesis establishes device engineering as a powerful method for introducing functionality in OFET beyond switching. It provides fundamental insights and scalable design approaches for organic photodetectors, gas sensors, and neuromorphic devices compatible with flexible, low-power electronics.