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Materials design and development of novel amorphous oxide semiconductors for light-emitting diodes

A thesis presented
by

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to
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Outline

OLEDs show promise for use in future displays. However, further improvement of OLEDs is required. To summarize, we can mainly categorize the issues into two groups. One comprises charge balance issues, and the other comprises intrinsic chemical instabilities, i.e., organic materials are highly sensitive to atmospheric moisture and oxygen. Thus, we reach the idea that new improved materials should be developed for carrier injection and emission layers; e.g., new inorganic materials with intrinsically low energy barriers relative to emission layers and inorganic light-emitting materials. In this thesis, new types of OLEDs and inorganic LEDs that can be fabricated on large-sized glass/plastic substrates are proposed. These OLEDs/LEDs are mainly based on AOSs because AOSs have low defect densities and can be fabricated at low temperatures including room temperature. The new materials employed include (i) electron/hole injection layers, (ii) electron/hole blocking layers, and (iii) emitting layers. This thesis aims to obtain simpler structures, longer lifetimes, and better chemical stability in large-sized self-emitting displays.

This thesis consists of six chapters. In Chapter 1, the background, motivation, and objectives of this work are reviewed, and new materials to be developed are proposed. Light-emission layers are studied in Chapters 2 and 3, and band engineering related to charge injection is studied in Chapters 4 and 5. In Chapter 2, an emission layer using an inorganic material was investigated for replacing organic emission layers in conventional OLEDs. I focused on ZnS:(Cu,Al) because it is a widely used inorganic phosphor. Thin films of ZnS:(Cu,Al) were fabricated on glass substrates, and the effects of thermal annealing were studied in relation to photoluminescence (PL) properties. I found that thin-film phosphor was successfully fabricated, but a high temperature of ~700 °C was required to obtain efficient PL.

In Chapter 3, I aimed to fabricate thin-film phosphors at low temperatures and observed those based on AOS host. Eu-doped a-IGZO thin films fabricated at room temperature exhibited intense red emission at 614 nm without post-deposition thermal annealing. In Chapter 4, the development of very wide bandgap AOSs was studied in a Ga-Zn-O system. Although conducting amorphous Ga2Ox has not yet been realized, I identified the deposition conditions required for obtaining conducting a-Ga2Ox. Furthermore, a band alignment diagram of a-Ga-Zn-O was built, which indicated that the bandgap and the valence band level can be widely tuned in this system. In Chapter 5, an a-In-Ga-Zn-(O,S) system was studied, with the initial aim of developing p-type AOS. Although conducting films were not obtained with S-containing materials, a large band bowing effect was observed, and a plausible electronic structure model was proposed.

In the Appendix, a highly efficient inverted OLED was fabricated using a newly developed transparent amorphous oxide semiconductor (TAOS) as an electron transport layer. This new TAOS has much higher mobility (~1 cm2/Vs) than conventional ETL materials (~1 × 10−4 cm2/Vs), a low work function (~3.5 eV), sufficient chemical stability, and high transparency, and can form Ohmic contacts with conventional electrodes (e.g., ITO, Al). In Chapter 6, the thesis is summarized and future prospects are discussed.