DOCTORAL THESIS (ABRIDGED)

博 士 論 文 （要 約）

Scalable High-Speed Hybrid Complementary Integrated Circuits Based on Solution-Processed Organic and Inorganic Transistors

（溶液プロセスを用いた有機無機ハイブリッド大面積高速 CMOS 集積回路）

Xiaozhu Wei

韋　瀟竹

This thesis discusses the technology to print high-performance complementary circuits on flexible substrates directly. A hybrid approach, for which organic small-molecule semiconductor 3,11-dinonylldinaphto[2,3-*d*:2',3'-*d*']benzo[1,2-*b*:4,5-*b*']dithiophene (C_9-DNBDT-NW) and amorphous metal-oxide indium-zinc-oxide (IZO) were used as p- and n-channel materials, respectively, was employed to break the limitation of a single material. We focused on developing integration processes for such new materials and discussing the device performances. The achievements of this thesis are summarized in the following sections.

Chapter 3 discusses the key factors to modulate the performances of thin film transistors (TFTs) based on solution-processed metal oxide semiconductors (MOSs). Semiconductor composition, annealing temperature, and channel layer thickness were studied respectively for high mobility, low-temperature fabrication, and good uniformity to pave a way for scalable high-speed flexible circuits. By removing Ga from the conventional indium-gallium-zinc-oxide (IGZO) system, optimizing the annealing temperature and active layer thickness, the carrier transport path, carrier concentration, and electron traps can be controlled effectively, enabling the IZO-based TFT to have enhanced electrical properties: carrier mobility as high as 12 $cm^2\ V^{-1}\ s^{-1}$, on-off current ratio (I_{on}/I_{off}) ~10^7, and off current (I_{off}) ~10^{-11} A. Significantly, almost no performance degradation was observed after exposure to the air for two years. The methods implemented in chapter 3 are strongly recommended to be utilized as guidelines to modulate TFT performance on a larger scale.

Chapter 4 introduces a damage-free integration process for solution-processed MOSs. Using a mild resist and developer to suppress chemical damage on the IZO and Al S/D electrodes induced by conventional photolithography played a pivotal role during this fabrication process. TFT arrays with channel length down to 1.7 μm still showed negligible hysteresis, carrier mobility of above 1 $cm^2\ V^{-1}\ s^{-1}$, and I_{on}/I_{off} ~10^{10}. The cutoff frequency is

estimated to be 23 MHz at V_G of 10 V. Noticeably, the voltage-normalized f_T of 2.3 MHz V^{-1} is the highest among printed n-channel TFTs reported to date. This result strongly proved the potential of as-fabricated IZO-based TFT to high-speed integrated circuits (ICs). Furthermore, the TFTs performed well even while bent to a radius of 3 mm. This damage-free patterning process could open up opportunities for implementing TFTs based on solution-processed MOSs in flexible ICs. Besides, passivation technology to stabilize MOS-based TFT in subsequent fabrication processes and practical operation were also discussed in this chapter. A poly(methyl methacrylate) (PMMA)/parylene/AlO_x hybrid passivation approach was developed. With the help of a thin PMMA buffer layer, process-induced damage during parylene deposition was perfectly precluded. Owing to the robust protection of the PMMA/parylene buffer layer, performance deterioration during AlO_x deposition was effectively suppressed. The high isolation effect of the hybrid passivation layer improved the bias stress resistance of IZO-based TFTs significantly: the I_D drop rate reduced from 67% in the non-passivated TFT to 2%; ΔV_{th} decreased from 2.8 V to 0.2 V. This passivation technology provides a simple and effective strategy to make the IZO-based TFT more plausible to practical applications.

Chapter 5 demonstrates an integration technology for hybrid complementary circuits. The high-performance materials—p-type C_9-DNBDT-NW single crystal and n-type amorphous IZO—provide a possibility for high-speed operation, flexibility, and large-area fabrication. The damage-free patterning processes for organic- and IZO-based TFTs and the highly effective passivation process for IZO-based TFT made the possibility become a reality. Hybrid complementary circuits, including inverters and ring oscillators, were fabricated successfully on polyimide substrates. The hybrid complementary inverters worked well at the ambient conditions, exhibited a large noise margin, negligible hysteresis, and power gain of 38 V/V at a supply voltage of 7 V. Notably, as-fabricated inverters also showed good long-term

stability, working well after exposure to the air for five months, and owned good flexibility. Finally, a 5-stage ring oscillator owning a propagation delay of 1.3 μs per stage has been demonstrated, proving the potential for more complex ICs. To our knowledge, the operation speed achieved in this study is the fastest one ever reported for flexible complementary inverters based on solution-processed OSCs or MOSs. The results indicate the possibility to print high-speed complementary circuits on flexible substrates directly.