論 文 の 内 容 の 要 旨

論文題目　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）

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Printing electronics has been of considerable interest in recent years since it offers a cost-efficient way to realize applications and brings new features for electronic devices such as flexibility. However, printing complementary circuits is still on the way due to the lack of suitable material systems: high-performance solution-processable n-type and p-type semiconductors of the same kind of materials. This thesis focused on a hybrid system, which combines the strengths of high-performance organic semiconductor (OSC) and amorphous metal-oxide semiconductor (MOS)[1-4] to print high-performance integrated circuits on flexible substrates.

The first part discusses the foundations of thin film transistors (TFTs) based on solution-processed amorphous MOSs. Ternary compound indium-zinc-oxide (IZO) was chosen as channel material because of its high electron performance compared with indium-gallium-zinc-oxide (IGZO). Particular focus was given to annealing temperature and channel layer thickness to control electron traps and carrier concentration. TFTs with enhanced performance, including mobility as high as 12 $cm^2 V^{-1} s^{-1}$, I_{on}/I_{off} up to 10^7, and high long-term stability, were achieved, as shown in **Figure 1**.

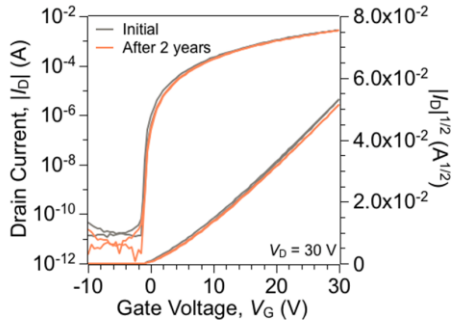

Figure 1 Transfer characteristics of an IZO-based TFT before and after exposure to the air for 2 years.

The second part discusses the integration methods for IZO-based TFTs towards flexibility, high-speed operation, and large-area fabrication. A damage-free lithography process in conjunction with ultimate optimizations of entire device processes for solution-processed MOSs was developed. IZO-based TFT arrays were successfully fabricated on flexible polyimide substrates by using a mild photosensitive material and developer. A cutoff frequency of 23 MHz in the air was achieved (**Figure 2**), which is almost twice faster than the frequency used in a near-field communication band. Furthermore, the as-fabricated IZO-based TFTs functioned well under bending stress. In addition, passivation technologies to stabilize MOS-based TFT against potential damages in subsequent fabrication processes and bias stress in practical operation have been explored, and A poly(methyl methacrylate)/parylene/AlO_x hybrid passivation approach was developed. Notably, AlO_x was successfully formed without degrading the MOS TFTs owing to the polymer buffer layers. The hybrid passivation approach ensured the satisfactory stabilization of the TFTs under bias stress due to the high isolation effect, which could prevent the penetration of environmental molecules (**Figure 3**).

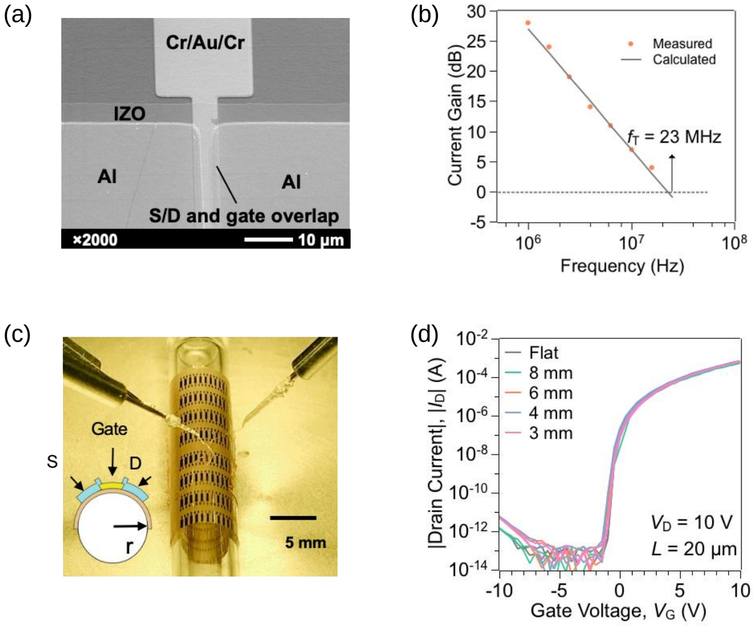

Figure 2 Photograph and characteristics of IZO-based short channel device. (a) SEM of a device with L = 1.7 mm. (b) Cutoff frequency measurement. (c) Photograph of measurement under *a* bending radium of 3 mm. (d) Transfer curves under different bending radii.

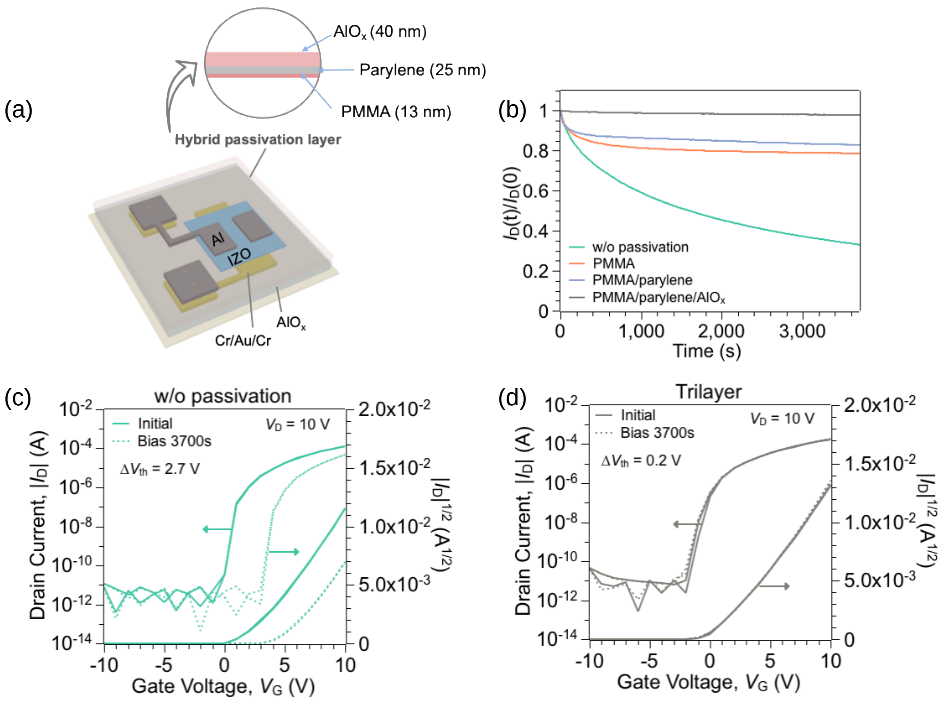

Figure 3 (a) Schematic illustration of a passivated TFT. (b) I_D changes during bias stress test for TFTs with different passivation structures and without passivation. Transfer characteristics (V_D = 10 V) before and after bias stress for 3700 s of TFTs (c) without passivation and (d) with hybrid passivation.

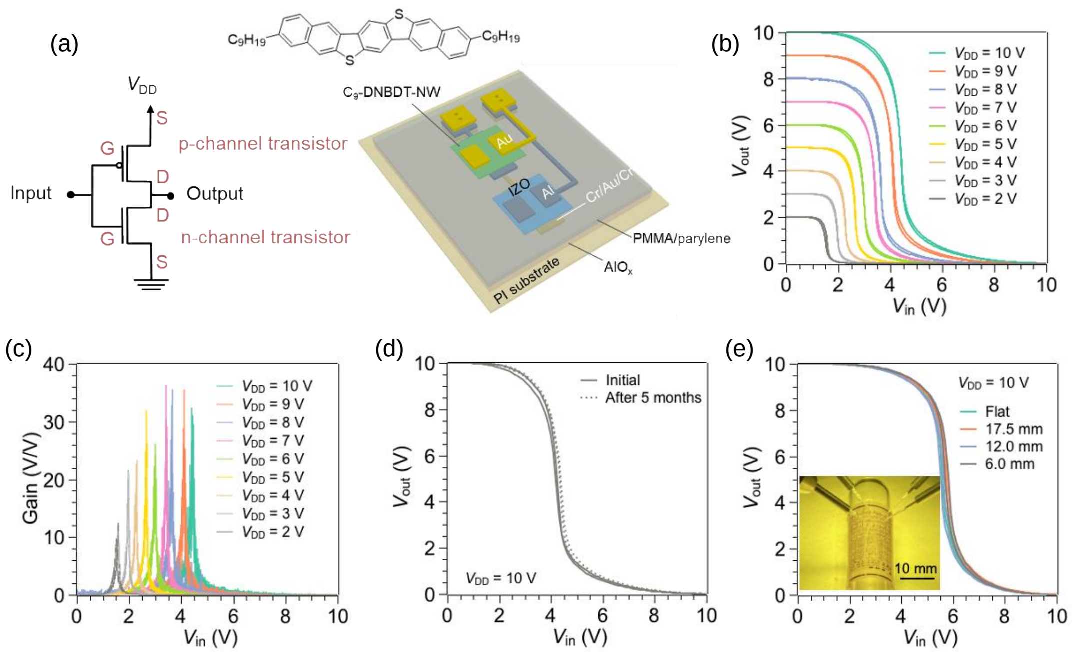

Figure 4 Electrical performance and flexibility of as-fabricated hybrid complementary inverters. (a) Schematic structure of a complementary inverter. (b) Voltage transfer curves and (c) voltage gains at V_{DD} from 2 V to 10 V. (d) Voltage transfer curves before and after storing in the ambient conditions without passivation for 5 months. (e) Transfer voltage curves of a hybrid complementary inverter while flat and bend to tensile radii of 17.5, 12.0, and 6 mm, respectively.

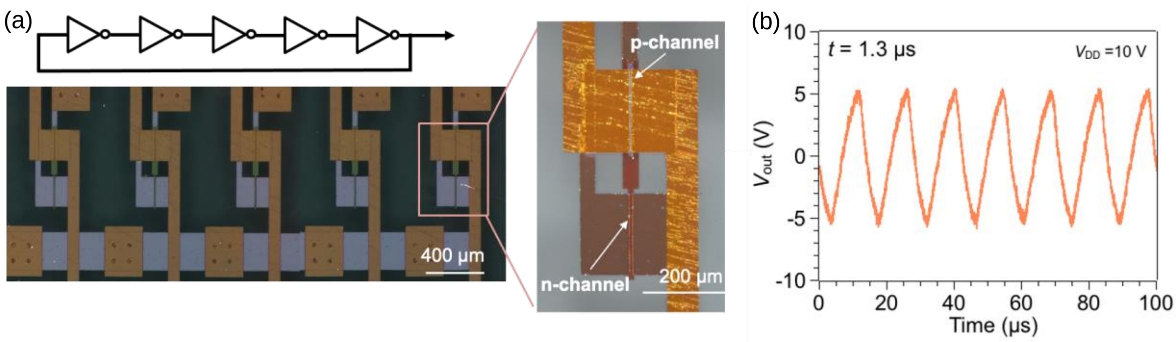

Figure 5 Electrical performance of a hybrid five-stage ring oscillator. (a) Schematic illustration and optical micrographs of a five-stage ring oscillator. (W/L = 200 µm/4 µm, ΔL_p = 3 µm for p-channel; W/L = 200 µm/8 µm ΔL_n = 1.5 µm for n-channel). The enlargement displays an optical micrograph of a hybrid inverter forming the ring oscillator. (b) Output signal of the ring oscillator at V_{DD} = 10 V.

The final part explores the integration technology for hybrid complementary circuits towards scalable fabrication and high-speed operation. Employing high-performance semiconductor materials: 3,11-dinonylldinaphto[2,3-*d*:2',3'-*d*']benzo[1,2-*b*:4,5-*b*']dithiophene (C_9-DNBDT-NW) single crystal as p-channel material and amorphous IZO as n-channel material, designing damage-free patterning processes for OSC- and IZO-based TFTs, and improving process durability of MOS-based TFTs subsequent integration of OSC-based TFT allow the successful fabrication of hybrid complementary circuits. The as-fabricated hybrid complementary inverter worked well at the ambient conditions and still functioned after storing at the ambient conditions without an extra passivation layer after 5 months. Large noise margin and power gain of 38 V/V were realized with a supply voltage of as low as 7 V. Good flexibility was demonstrated with a bending test (**Figure 4**). Furthermore, a five-stage ring oscillator owning a propagation delay of 1.3 μs per stage has been achieved (**Figure 5**), which is the fastest operation ever reported for flexible complementary inverter based on solution-processed MOSs or OSCs to our knowledge.

Summary

Scalable high-speed hybrid complementary integrated circuits based on solution-processed organic p-type semiconductor (C_9-DNBDT-NW) and n-type metal oxide semiconductor (IZO) have been demonstrated. Employing solution-processed IZO as an active layer and optimizing sol-gel process conditions enabled IZO-based TFTs to own high mobilities and good long-term stability. Furthermore, short-channel IZO-based TFTs (L = 1.7 μm) were successfully prepared by a mild photosensitive material and developer. A cut-off frequency of 23 MHz was achieved for as-fabricated TFT (L = 1.7 μm) in the air, which is the fastest among reported solution-processed MOSs. In addition, the IZO TFTs worked well under bending stress, suggesting a promise for flexible device applications. A PMMA/parylene/AlO_x hybrid passivation layer stabilized the TFTs against bias stress and damage from subsequent processes. Finally, a facile hybrid CMOS fabrication method was demonstrated. An optimal method led to the preferrable characteristics of hybrid CMOS due to the nearly balanced high performances of organic- and IZO-based TFTs. A five-stage ring oscillator with a propagation delay of 1.3 μs proved the potential of this technology for scalable integrated circuits. The proposed hybrid CMOS technology enables print high-speed complementary circuits directly on flexible substrates, and we envision it will contribute to achieving various advanced applications for the future IoT society.

Reference

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