Low-Voltage Organic Transistors for Flexible Electronics

Ute Zschieschang [a], Reinhold Rödel [a], Ulrike Kraft [a], Kazuo Takimiya [b], Tarek Zaki [c], Florian Letzkus [d], Jörg Butschke [d], Harald Richter [d], Joachim N. Burghartz [d], Wei Xiong [e], Boris Murmann [e], Hagen Klauk [a]

[a] Max Planck Institute for Solid State Research, Heisenbergstr. 1, 70569 Stuttgart, Germany
[b] Emergent Molecular Function Research Team, RIKEN Advanced Science Institute, Wako, Saitama, Japan
[c] Institute for Nano- and Microelectronic Systems (INES), University of Stuttgart, Germany
[d] Institut für Mikroelektronik/IMS CHIPS, Stuttgart, Germany
[e] Center for Integrated Systems, Stanford University, Stanford, USA

***Abstract* - A process for the fabrication of bottom-gate, top-contact (inverted staggered) organic thin-film transistors (TFTs) with channel lengths as short as 1 µm on flexible plastic substrates has been developed. The TFTs employ vacuum-deposited small-molecule semiconductors and a low-temperature-processed gate dielectric that is sufficiently thin to allow the TFTs to operate with voltages of about 3 V. The p-channel TFTs have an effective field-effect mobility of about 1 cm^2/Vs, an on/off ratio of 10^7, and a signal propagation delay (measured in 11-stage ring oscillators) of 300 ns per stage. For the n-channel TFTs, an effective field-effect mobility of about 0.06 cm^2/Vs, an on/off ratio of 10^6, and a signal propagation delay of 17 µs per stage have been obtained.**

I. Introduction

Organic thin-film transistors (TFTs) can typically be fabricated at temperatures below about 100 °C and thus not only on glass substrates, but also on a variety of unconventional substrates, such as plastics and paper. This makes organic TFTs potentially useful for the realization of flexible, large-area electronics applications, such as rollable or foldable information displays [1], conformable sensor arrays [2], and plastic circuits [3]. In some of the more advanced applications envisioned for organic TFTs, such as the integrated row and column drivers of flexible active-matrix organic light-emitting diode (AMOLED) displays [4], the TFTs will have to be able to control electrical signals of a few volts at frequencies of several megahertz. For portable applications powered by small batteries, an additional requirement is a low power consumption, which implies a complementary circuit technology and thus the availability of both p-channel and n-channel TFTs with sufficient static and dynamic performance on plastic substrates.

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II. Materials and Design Considerations

The first requirement for achieving high switching frequencies in organic TFTs is efficient charge transport in the organic semiconductor layer. This requirement can be met by choosing small-molecule semiconductors that provide good molecular ordering and usefully large field-effect mobilities even when processed at temperatures below 100 °C. In the case of organic p-channel TFTs, for example, the alkylated thienoacene C_{10}-DNTT that was recently developed in the group of Kazuo Takimiya at Hiroshima University has shown very promising field-effect mobilities in the range of 10 cm^2/Vs [5,6]. For organic n-channel TFTs, a number of naphthalene and perylene tetracarboxylic diimides equipped with strongly electron-withdrawing core substituents and fluoroalkyl chains at both imide positions have recently been developed, such as NTCDI-Cl_2-$(CH_2C_3F_7)_2$ (which was recently developed in the group of Frank Würthner at the University of Würzburg [7]) and PTCDI-(CN_2)-$(CH_2C_3F_7)_2$ (recently developed in the group of Antonio Facchetti and Tobin Marks at Northwestern University [8]), both of which have shown electron mobilities exceeding 1 cm^2/Vs along with excellent air stability.

The second requirement for achieving high switching frequencies in organic TFTs is a small channel length [9-12]. To meet this requirement, a TFT process in which high-resolution silicon stencil masks are employed for the patterning of the source and drain contacts and all other components of the transistors, including the gate electrodes and the organic semiconductor layer, has recently been developed [11,12]. With this process, bottom-gate, top-contact organic TFTs with a channel length of 1 µm can be fabricated on plastic substrates without exposing the organic semiconductors to potentially harmful organic solvents and photoresists.

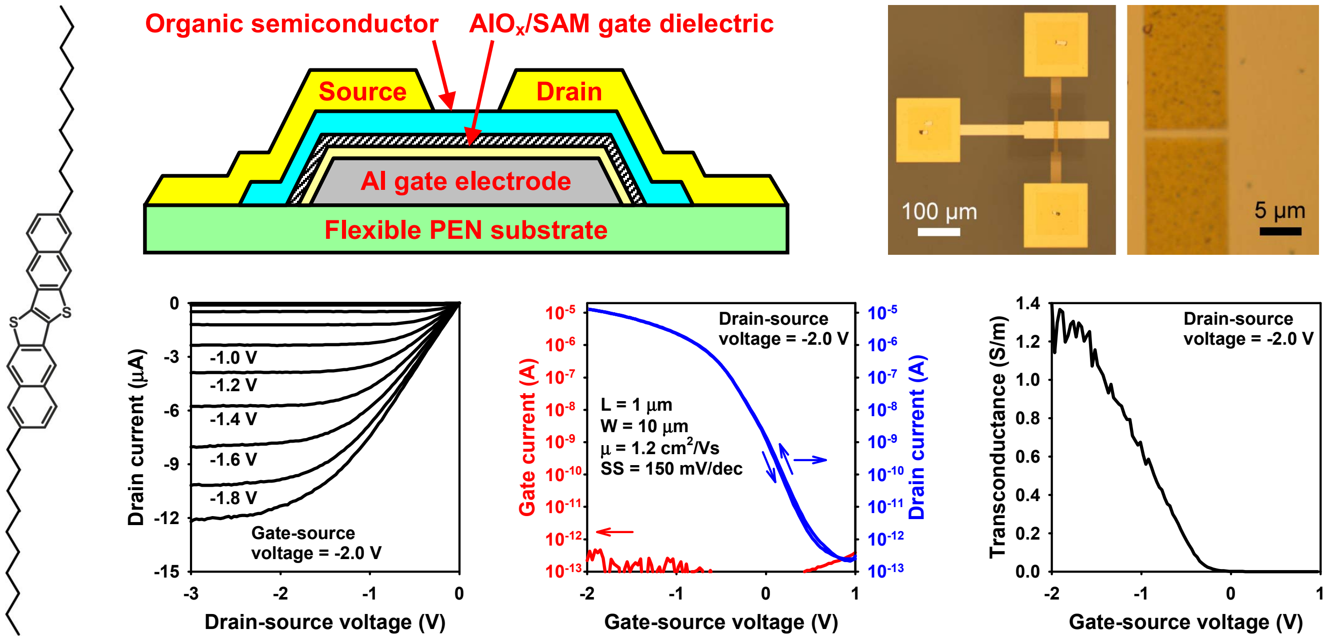


Fig. 1. Schematic cross-section, photographs, and measured current-voltage characteristics of a C_{10}-DNTT p-channel TFT with a channel length of 1 µm fabricated on a flexible polyethylene (PEN) substrate. The TFT has an effective field-effect mobility of 1.2 cm^2/Vs, an on/off current ratio of 10^7, a subthreshold swing of 150 mV/decade, and a width-normalized transconductance of 1.2 S/m. Also shown is the chemical structure of the organic semiconductor (C_{10}-DNTT) employed for these TFTs [12].

III. Transistor Fabrication Process

A set of four stencil masks is required to fabricate either p-channel or n-channel TFTs: one mask each to pattern the gate electrodes, the gate vias, the organic semiconductor layer, and the source/drain contacts. To fabricate both p-channel and n-channel TFTs on the same substrate for the realization of complementary circuits, a total of five masks are required, since in this case two different semiconductors need to be individually deposited and patterned.

In the first process step, a thin layer of aluminum is deposited directly onto the substrate by thermal evaporation in vacuum through the first stencil mask. In order to define the locations for the gate vias, a thin layer of gold is then deposited by thermal evaporation in vacuum through the second stencil mask onto specific locations on the aluminum outside of the active TFT areas. In the third step, a hybrid gate dielectric composed of a 3.6-nm-thick layer of aluminum oxide (obtained by briefly exposing the surface of the aluminum gate electrodes to an oxygen plasma) and a 1.7-nm-thick self-assembled monolayer (SAM) of *n*-tetradecylphosphonic acid (obtained by briefly immersing the substrate into a 2-propanol solution of the phosphonic acid) is then produced [11,12]. This gate dielectric forms only on the surface of the aluminum gate electrodes, but not in the gold-covered via locations. In the fourth and fifth steps, thin layers of the organic semiconductors (one each for the p-channel and n-channel TFTs) are deposited onto the AlO$_x$/SAM gate dielectric by sublimation in vacuum through the third and fourth stencil masks. Finally, a thin layer of gold is deposited by thermal evaporation in vacuum through the fifth stencil mask to define the source and drain contacts of the TFTs as well as the interconnects for the integrated circuits. The highest temperature during the fabrication process is 100 ºC.

IV. Performance of p-channel TFTs

Figure 1 shows two photographs and the measured current-voltage characteristics of a C_{10}-DNTT p-channel TFT with a channel length of 1 µm and a channel width of 10 µm fabricated on a flexible, 125-µm-thick polyethylene naphthalate (Teonex® Q65 PEN; kindly provided by William A. MacDonald, Du-Pont Teijin Films, Wilton, UK) substrate. The small thickness (5.3 nm) and large capacitance per unit area (800 nF/cm^2) of the AlO$_x$/SAM gate dielectric allow the TFTs to operate with low voltages of about 3 V. The TFTs have an effective hole mobility in the saturation regime of 1.2 cm^2/Vs, an on/off current ratio of 10^7, a subthreshold swing of 150 mV/decade, and a width-normalized transconductance of 1.2 S/m. While the width-normalized transconductance is smaller by three to four orders of magnitude than that of state-of-the-art silicon MOSFETs at the 22-nm technology node [13], it is believed to be the largest transconductance reported thus far for organic TFTs on plastic substrates.

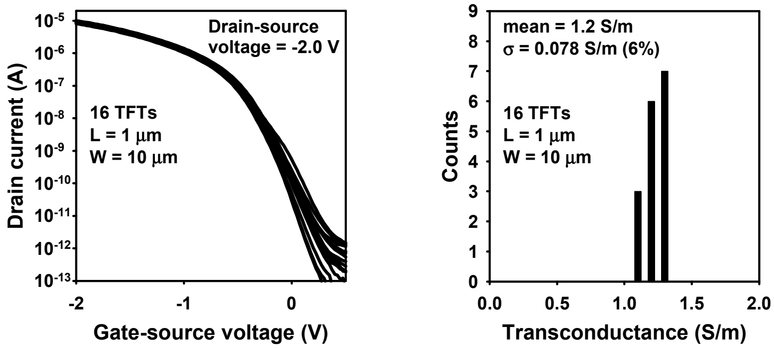


Fig. 2. Distribution of the measured transfer characteristics and distribution of the width-normalized transconductance in an array of sixteen C_{10}-DNTT TFTs with a channel length of 1 µm fabricated on a flexible PEN substrate [12].

Figure 2 shows the measured transfer characteristics and the distribution of the extracted width-normalized transconductance in an array of sixteen C_{10}-DNTT p-channel TFTs with a nominal channel length of 1 µm and a nominal channel width of 10 µm fabricated on a flexible PEN substrate. All 16 TFTs have an on/off ratio of 10^7. Across the array of 16 TFTs, the width-normalized transconductance varies between a minimum of 1.16 S/m and a maximum of 1.40 S/m, with a standard deviation of 6%. According to the standard FET equations, the width-normalized transconductance is inversely proportional to the channel length, so assuming that the other transistor parameters, including the effective field-effect mobility and the gate-dielectric capacitance, are constant across the substrate, a standard deviation in the transconductance of 6% may simply reflect a standard deviation in the channel length of 6%, which in the case of an intended channel length of 1 µm corresponds to a standard deviation of 60 nm. This is obviously substantially larger than the standard deviation of the gate length of state-of-the-art silicon MOSFETs [14], but in contrast to silicon MOSFETs, the organic TFTs were fabricated without lithographic pattern reduction, which may explain the larger standard deviation in the feature size of the TFTs.

The third requirement for achieving high switching frequencies (in addition to a large field-effect mobility and a small channel length, as discussed above) is a small gate capacitance. One component of the gate capacitance is the parasitic capacitance that is formed by the geometric overlaps between the gate electrode and the source/drain contacts, so reducing not only the channel length, as discussed above, but also the gate overlap length can be useful in view of high-frequency TFT operation [11,12].

Figure 3 shows the circuit schematic and the photograph of an 11-stage ring oscillator with output buffer comprised of unipolar inverters with saturated load based on C_{10}-DNTT p-channel TFTs fabricated on a flexible PEN substrate. In the most aggressive design, the organic TFTs have a channel length (L) of 1 µm, a gate overlap (L_C) of 5 µm, and channel widths (W) of 24 µm (for the drive TFTs) or 72 µm (for the load TFTs). In a more relaxed design, the channel length is 4 µm and the gate overlap is 20 µm. Also shown in Figure 3 are the signal propagation delays per stage measured in these ring oscillators and plotted as a function of the supply voltage. For the more aggressive dimensions (L = 1 µm, L_C = 5 µm), the measured stage delay is 1.9 µs at a supply voltage of 1 V, 730 ns at 2 V, 420 ns at 3 V, and 300 ns at 4 V. These are believed to be the first organic TFTs fabricated on flexible plastic substrates demonstrating cutoff frequencies above 1 MHz at supply voltages below 10 V.

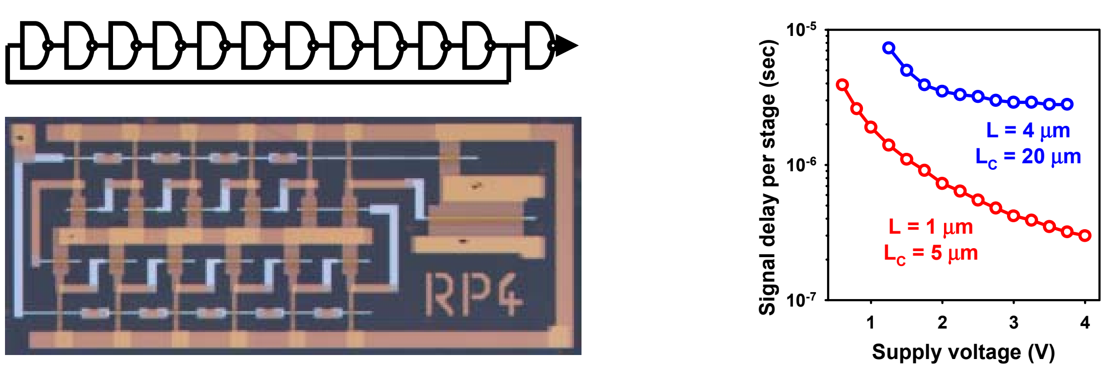
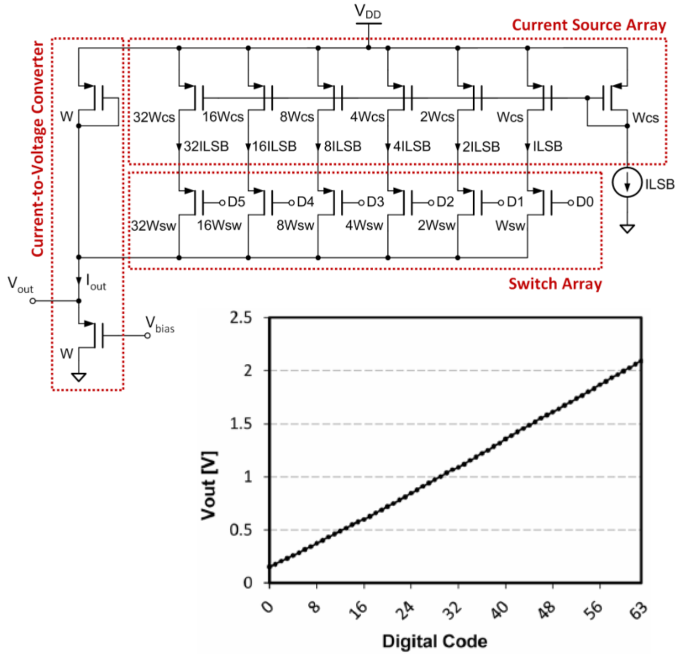


Fig. 3. Circuit schematic, photograph and measured signal propagation delay per stage as a function of the supply voltage of 11-stage unipolar ring oscillators fabricated with C_{10}-DNTT TFTs with two different channel lengths (1 µm and 4 µm) on a flexible PEN substrate. For a channel length of 1 µm, the measured stage delay is 1.9 µs at a supply voltage of 1 V, 730 ns at 2 V, 420 ns at 3 V, and 300 ns at 4 V [12].



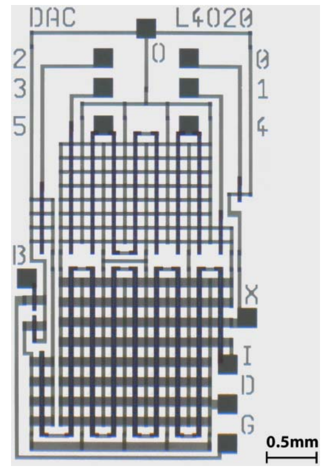


Fig. 4. Circuit schematic, photograph, and measured transfer function of a 6-bit digital-to-analog converter based on low-voltage organic p-channel TFTs with a channel length of 4 µm fabricated on a glass substrate. The DAC has a supply voltage of 3.3 V and a maximum sampling rate of 100 kS/s [15].

V. D/A Converter based on Organic p-channel TFTs

Figure 4 shows the schematic, a photograph, and the measured transfer function of a 6-bit binary-weighted current-steering digital-to-analog converter (DAC) based on low-voltage organic p-channel TFTs with a channel length of 4 µm fabricated on a glass substrate [15]. The DAC has a supply voltage of 3.3 V, a circuit area of 2.6 × 4.6 mm^2, and a maximum sampling rate of 100 kS/s.

VI. Performance of n-channel TFTs

By choosing an organic semiconductor with a large electron affinity, i.e., with a large energy difference between the lowest unoccupied molecular orbital (LUMO) and the vacuum level, it is possible to realize organic n-channel TFTs. An example for such a semiconductor is the recently developed core-chlorinated and fluoroalkyl-substituted naphthalene tetracarboxylic diimide NTCDI-Cl_2-$(CH_2C_3F_7)_2$ [7]. Figure 5 shows the chemical structure of this semiconductor, along with the measured current-voltage characteristics of an NTCDI-Cl_2-$(CH_2C_3F_7)_2$ n-channel TFT with a channel length of 1 µm and a channel width of 50 µm. The TFTs have an effective electron mobility in the saturation regime of 0.06 cm^2/Vs, an on/off ratio of 10^6, a subthreshold swing of 180 mV/decade, and a width-normalized transconductance of 0.06 S/m. While these parameters are notably inferior to those of the C_{10}-DNTT p-channel TFTs shown in Figure 2, they represent the best performance currently achievable in air-stable, low-voltage organic n-channel TFTs with such a small channel length.

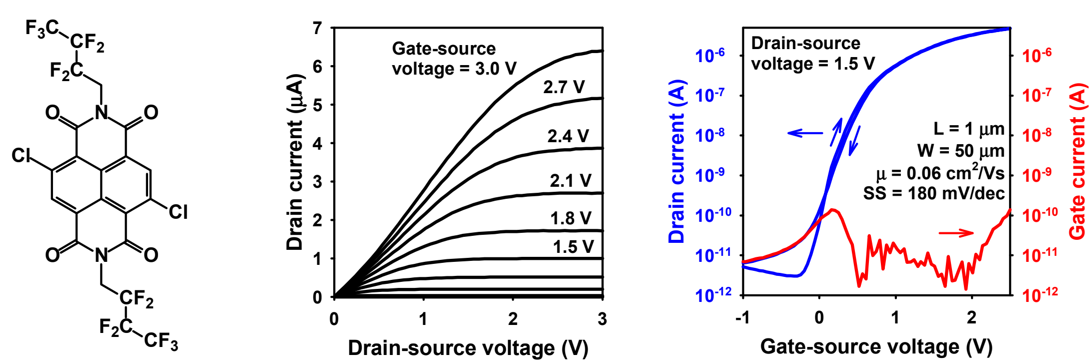


Fig. 5. electrical characteristics of an NTCDI-Cl_2-$(CH_2C_3F_7)_2$ n-channel TFT with a channel length of 1 µm. The TFT has an effective field-effect mobility of 0.06 cm^2/Vs, an on/off current ratio of 10^6, a subthreshold swing of 180 mV/decade, and a width-normalized transconductance of 0.06 S/m. Also shown is the chemical structure of the organic semiconductor NTCDI-Cl_2-$(CH_2C_3F_7)_2$ employed for these n-channel TFTs [16].

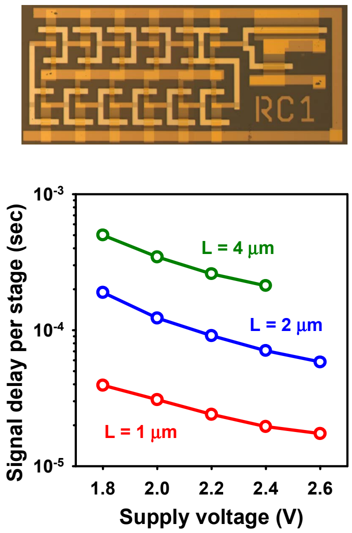


Fig. 6. Photograph and measured signal propagation delay per stage as a function of the supply voltage of 11-stage complementary ring oscillators fabricated with and NTCDI-Cl_2-$(CH_2C_3F_7)_2$ n-channel TFTs and DNTT p-channel TFTs on a flexible PEN substrate [16].

Although organic n-channel TFTs with larger electron mobilities have been reported, these TFTs either had much large channel lengths (suppressing the detrimental influence of the contact resistance on the total device resistance [16,17]) or they could only be operated in an inert ambient, due to a lack of air stability of the semiconductor, e.g., C_{60} [18].

The fact that the mobility of the n-channel TFTs is significantly smaller than that of the p-channel TFTs implies that the dynamic performance of complementary circuits based on organic n-channel and p-channel TFTs will be limited by the longer signal propagation delay of the n-channel devices. Figure 6 shows the photograph of an 11-stage complementary ring oscillator based on NTCDI-Cl_2-$(CH_2C_3F_7)_2$ n-channel TFTs and DNTT p-channel TFTs fabricated on a flexible PEN substrate, along with the signal delays measured for ring oscillators with minimum feature sizes of 1 µm, 2 µm and 4 µm. For the most aggressive design (L = 1 µm), the measured stage delay is 17 µs at a supply voltage of 2.6 V, which is indeed slower by more than an order of magnitude compared with the signal delay of the unipolar all-p-channel ring oscillators shown in Figure 3. However, regardless of the lower dynamic performance, these results demonstrate the feasibility of realizing low-voltage, low-power complementary circuits on flexible plastic substrates.

VII. Organic Complementary A/D Converter

As an early demonstration of low-voltage, low-power mixed-signal organic complementary circuits, a 6-bit switched-capacitor (C-2C) analog-to-digital converter (ADC) based on 19 thin-film capacitors, 27 organic p-channel TFTs and 26 organic n-channel TFTs (having a channel length of 20 µm) was fabricated on a glass substrate [19]. The circuit schematic, a photograph, and the measured transfer function are shown in Figure 7. The ADC has a supply voltage of 3 V, a circuit area of 28 × 22 mm^2, and a maximum sampling rate of 100 S/s.

VIII. Conclusions

Organic thin-film transistors can be fabricated at low process temperatures of typically about 100 ºC, which makes them useful for flexible electronics applications on unconventional substrates, such as plastics or paper. In this work, low-voltage p-channel and n-channel organic TFTs with a channel length of 1 µm as well as unipolar and complementary mixed-signal circuits with promising static and dynamic performance were demonstrated.

Acknowledgements

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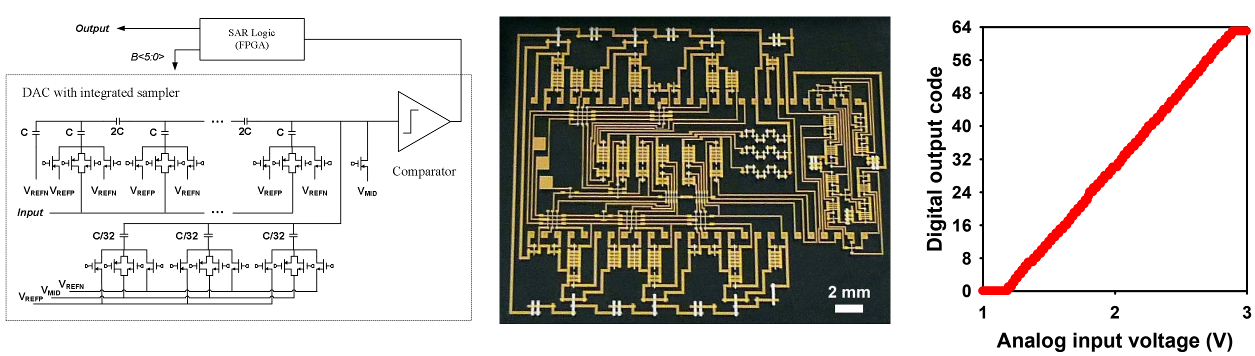

Fig. 7. Circuit schematic, photograph, and measured transfer function of a 6-bit complementary analog-to-digital converter fabricated on a glass substrate. The ADC has a supply voltage of 3 V and a maximum sampling rate of 100 S/s [19].

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