Organic thin film transistors

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Since John Bardeen, William Shockley, and Walter Brattain invented the world's first transistor in 1947, inorganic field-effect transistors (FETs) have dominated the mainstream microelectronics industry. They are the fundamental building blocks for basic analytical circuits, such as amplifiers, as well as the key elements for digital combinational logic circuits, such as adders, shifters, inverters, and arithmetic logic units, and are used to build sequential logic circuits, such as flip-flops. Moreover, transistors are essential to the modern memory devices, integrated circuits, and microprocessors used in personal computers and laptops.

Organic thin film field-effect transistors (OTFTs) are particularly interesting as their fabrication processes are much less complex compared with conventional Si technology, which involves high-temperature and high-vacuum deposition processes and sophisticated photolithographic patterning methods. In general, low-temperature deposition and solution processing can replace the more complicated processes involved in conventional Si technology. In addition, the mechanical flexibility of organic materials makes them naturally compatible with plastic substrates for lightweight and foldable products. Since the report of the first organic field-effect transistor in 1986[1], there has been great progress in both the materials' performance and development of new fabrication techniques. OTFTs have already been demonstrated in applications such as electronic paper[2-4], sensors[5,6], and memory devices including radio frequency identification cards (RFIDs)[7,8]. Although OTFTs are not meant to replace conventional inorganic TFTs – because of the upper limit of their switching speed – they have great potential for a wide variety of applications, especially for new products that rely on their unique characteristics, such as electronic newspapers, inexpensive smart tags for inventory control, and large-area flexible displays.

In this article, we will describe the basic materials requirements and fabrication methods for building these

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devices, and outline the related technical issues and challenges. Promising applications of OTFTs will also be discussed.

Operation and materials

An OTFT is analogous to its inorganic counterpart in basic design and function. It is a three-terminal device, in which a voltage applied to a gate electrode controls current flow between a source and drain electrode under an imposed bias. A basic schematic is shown in Fig. 1, where V_g and V_{ds} are the applied gate and source-drain voltages, respectively. The control of source-drain current in FETs via a third terminal has resulted in their widespread use as switches. Their utility in this capacity is gauged by several key measures of their performance. The mobility, μ, describes how easily charge carriers can move within the active layer under the influence of an electric field and is, therefore, directly related to the switching speed of the device. This parameter can be extracted from current-voltage measurements, and would ideally be as large as possible. Typical values range from 0.1-1 cm^2/V.s for amorphous-Si (a-Si) devices, with the best organic materials achieving mobilities of 1-10 cm^2/V.s[9,10]. The on/off ratio, defined as the ratio of the current in the 'on' and 'off' states, is indicative of the switching performance of OTFTs. A low off current is desired to eliminate leakage while in the inactive state. Ratios as high as 10^6 – suitable for most applications – can be reached by current-generation OTFTs[11,12].

In a traditional inorganic device, the active semiconductor layer is generally comprised of lightly doped Si, or combinations of Group III-V elements, such as GaAs. In these materials, the applied gate voltage causes an accumulation of minority charge carriers at the dielectric interface, e.g. electrons in a p-type material, termed an 'inversion layer'. In this very shallow channel, carriers injected from the source and drain electrodes may pass, resulting in current flow. In an organic transistor, the active layer is comprised of a thin film of highly conjugated small molecules or polymers (Fig. 2), such as p-channel[13-15] pentacene[9,16] (**1**), α-sexithiophene (α-6T)[17] (**2**), and poly-3-hexylthiophene (P3HT)[18-21] (**6**), or n-channel[22-25] benzobisimidazobenzophenanthroline (BBL)[26,27] (**10**) and perfluorinated copper-phthalocyanine (F$_{16}$-CuPc)[28] (**12**). In stark contrast to inorganic materials, organics pass current by majority carriers, and an inversion regime does not exist. This fundamental difference is related to the nature of charge transport in each of these semiconductors. In well-ordered inorganics, e.g. single-crystal Si, the delocalization of electrons over equivalent sites leads to a band-type mode of transport, with charge carriers moving through a continuum of energy levels in the solid. In less-ordered organic materials, the proposed mechanism is hopping between discrete, localized states of individual molecules. The presence of impurities or inconsistencies in structure may result in 'traps' that alter the relative energy levels, and inhibit the flow of charge carriers. The complexities of current flow in organic materials – which are still poorly understood – have added another dimension to the development of the devices that incorporate them. In addition to the challenges presented by fabrication, particular attention must be paid to the design of materials that will meet the performance demands of the OTFT in its parent applications. This aspect of development
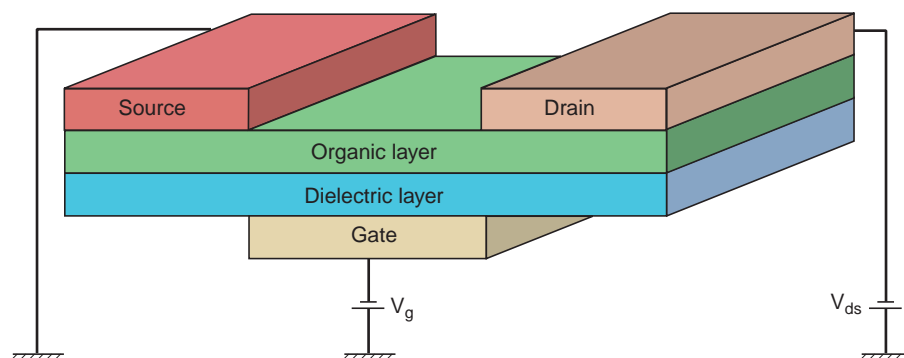

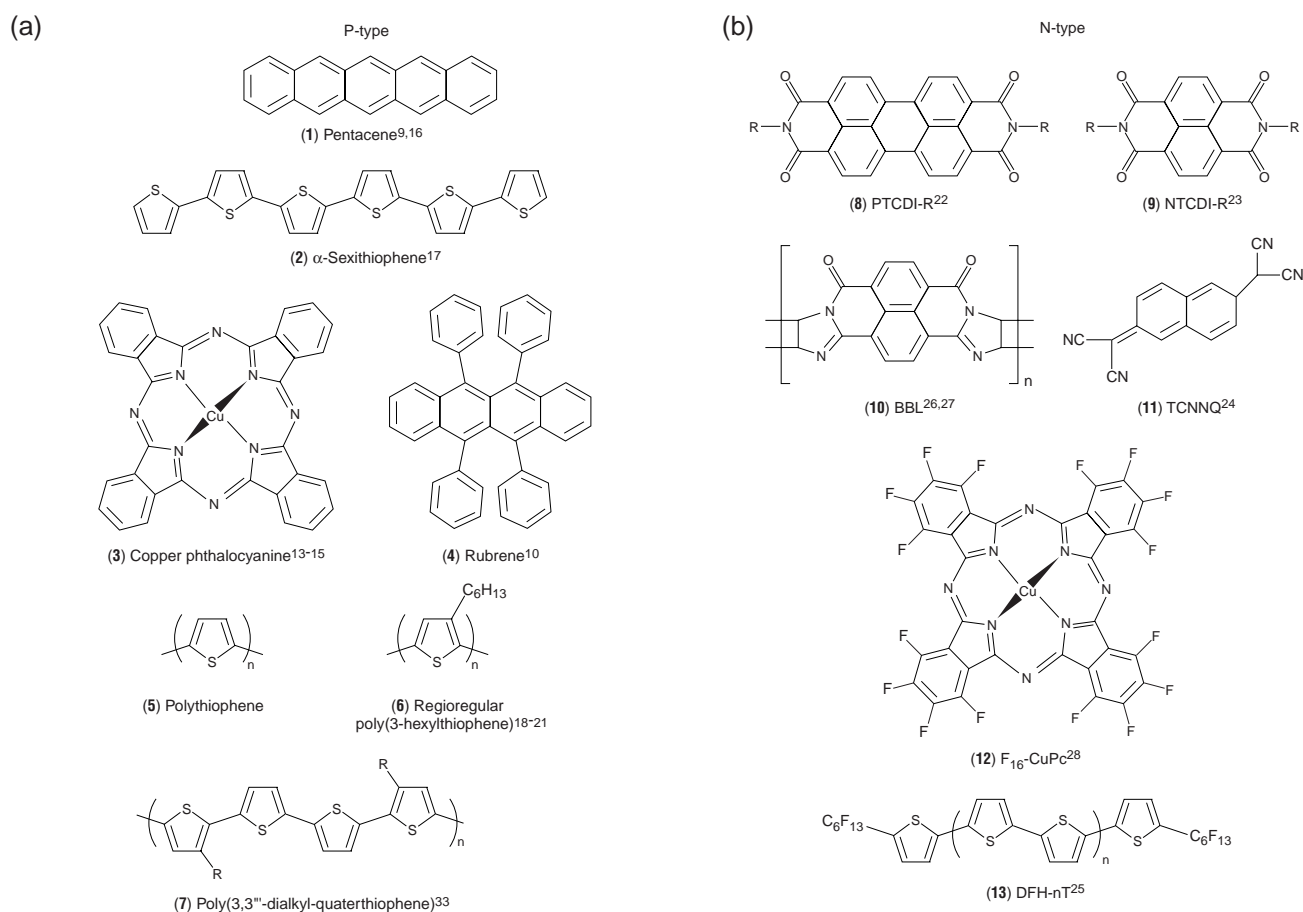Fig. 1 Basic schematic of a field-effect transistor.

(a)

P-type

(1) Pentacene[9,16]

(2) α-Sexithiophene[17]

(3) Copper phthalocyanine[13-15]

(4) Rubrene[10]

(5) Polythiophene

C_6H_{13}

(6) Regioregular poly(3-hexylthiophene)[18-21]

(7) Poly(3,3'''-dialkyl-quaterthiophene)[33]

(b)

N-type

(8) PTCDI-R[22]

(9) NTCDI-R[23]

(10) BBL[26,27]

(11) TCNNQ[24]

(12) F$_{16}$-CuPc[28]

C_6F_{13} ... C_6F_{13}

(13) DFH-nT[25]

Fig. 2 Prominent (a) p- and (b) n-type organic semiconductor materials.

has received much attention, and will continue to play an important role as the technologies progress.

The limitations of current organic technologies are clearly posed by the performance and processability of the active layer component. In addition to meeting benchmarks for performance criteria, such as mobility and on/off ratio, active layer materials should ideally be easy to process, mitigating potential fabrication challenges, and have long-term stability for device longevity. This has proven a difficult balance. The organics possessing the best electronic characteristics to date, small molecules such as pentacene[16,29] (**1**) and α-6T[17] (**2**), are insoluble and therefore difficult to process. Efforts to solubilize these materials have included the incorporation of side chains, such as the addition of alkyl groups to polythiophene polymers (**6**,**7**). The size[30], type[31], and regioregularity[20,21] of these groups have been explored extensively, with the goal of electronic property optimization. From these studies, additional insight has also been gained

regarding the relationships between morphological characteristics and charge transport. The nature of substituents, chain length, and processing conditions all affect the packing structure in the films, which is reflected in the electronic properties. For example, differences in regioregularity in P3HT (**6**) samples have been shown to cause distinctly different orientations relative to the substrate[32]. The two films, shown schematically in Fig. 3, have mobilities that differ by two orders of magnitude. In this manner, careful substituent selection has been used to tune organic semiconductors for the best all-around characteristics[33]. Molecular weight (M_w), as well as the solvent used for film deposition, has also been shown to have a significant effect on ordering and properties[15,34]. Atomic force microscopy of regioregular P3HT polymer films (Fig. 4) illustrates the dramatic differences in morphology between films of 3.2 kD and 31.1 kD M_w, whose mobilities again differ by more than two orders of magnitude[34]. Clearly, many
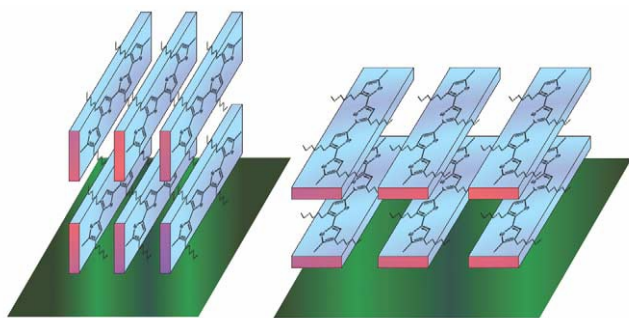
Fig. 3 Packing orientation of P3HT films relative to substrate with (a) 81% and (b) 95% regioregularity, as determined by X-ray diffraction[32]. The films differ in mobility (parallel to substrate) by a factor >100, illustrating the effect of morphology on performance.
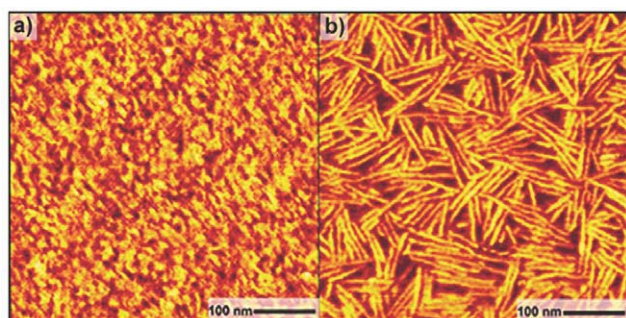

Fig. 4 Atomic force microscopy images of regioregular P3HT films with M_w of (a) 3.2 kD and (b) 31.1 kD[34]. The large molecular weight polymer forms a more ordered, crystalline film, yet has a mobility that is two orders of magnitude lower than the 3.2 kD film. (Reprinted with permission from[34]. © 2003 Wiley-VCH.)

factors are at play when active materials do not exhibit the order of single crystals, and the various effects have yet to be deconvoluted. Recent reports of mobilities up to 35 cm^2/V.s for high-purity pentacene single crystals[29], however, give hope that further investigation will continue to pay off for organic semiconductors.

Device design and fabrication

As previously mentioned, the OTFT inherits its design architecture from its inorganic counterpart, namely, the metal-oxide-semiconductor field-effect transistor (MOSFET). It is composed of three main components: source, drain, and gate electrodes; a dielectric layer; and the active semiconductor layer. Within the basic MOSFET design, there are two types of device configuration: top contact and bottom contact. The former involves building source and drain electrodes onto a preformed semiconductor layer, whereas the latter is constructed by depositing the organic over the contacts. The structures are illustrated

schematically in Fig. 5. Top contact devices have been reported to have superior performance for certain organic semiconductors compared with their bottom contact counterparts. It has been suggested that this is a result of reduced contact resistance between the electrode and the organic layer because of an increase in the area for charge injection[35]. Each of these devices has particular advantages and disadvantages in the fabrication process, which will be discussed below.

Deposition of thin film organic semiconductor layers is primarily performed through vapor or solution phase processes. Vacuum deposition is used for small molecules and oligomers. It is somewhat costly because of expensive equipment and low deposition throughput, but produces films with high field-effect mobility and on/off ratios. Examples of organic semiconductor films that have been deposited in this manner are oligothiophene and oligofluorene derivatives[12,36-38], metallophthalocyanines[15,39], and acenes[40-44] (pentacene and tetracene). OTFT device performance can be improved by controlling the deposition rate and temperature, which affect the morphology of the semiconductor. Modification of the interface between the substrate and the organic layer and post-deposition treatments, such as annealing, also improve molecular ordering.
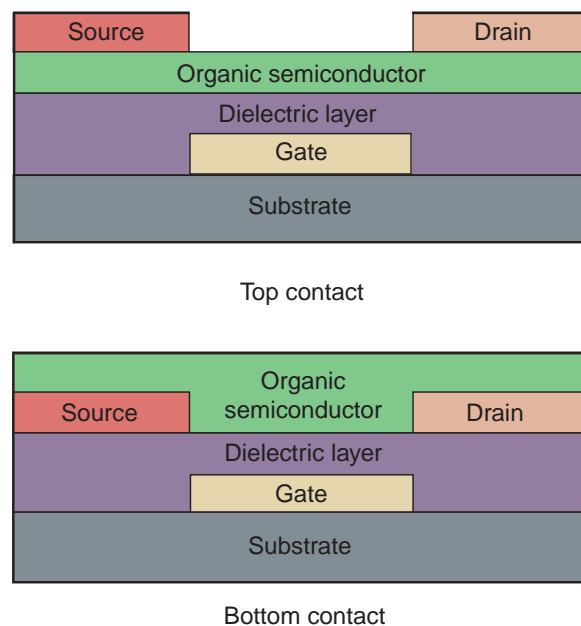

Top contact

Bottom contact

Fig. 5 Top and bottom contact OTFT architectures.

For solution-soluble organic semiconductors, two forms of deposition are available: deposition of a soluble precursor from a solution and subsequent conversion to the final film[45-49], and direct deposition from solution. The motivation to use soluble precursors is that most conjugated oligomers and polymers are insoluble in common solvents unless side chain substitutions are incorporated into the molecular structures. The addition of side chains can interfere with molecular packing or increase the π-π stacking distance between molecules, decreasing mobility, but when used properly can be incorporated to promote better molecular packing, such as in the case of regioregular P3HT. Determining the processing temperature can be challenging, however, as the conversion temperature from precursor to semiconductor may be too high for compatibility with low-cost plastic substrates. Furthermore, the precursor conversion requires an additional processing step.

Spin-coating and solution casting are two popular ways of direct solution deposition, which is often used for polymers such as regioregular P3HT[21,32-33] or various soluble oligomers[50-52]. Post-processing treatments, such as thermal annealing, improve molecular ordering and grain sizes of the thin film and frequently result in better device performance. It is often difficult, however, to purify the polymers and achieve good molecular ordering over large-area substrates. Another major concern for solution processing methods is the effect of the solvent on underlying organic features, requiring chemically compatible materials. For this reason, dry processing methods are being developed[53]. Generally, it is important to consider the organic solution concentration and solubility, solvent evaporation rate, and substrate surface. Because of their effect on the quality of the resulting semiconductor film, these processing parameters should be carefully controlled. High mobilities have been reported with several oligomers by optimizing the deposition conditions[50-52].

Dielectric films are fabricated in a similar manner to the semiconductor layer. Examples of vacuum-deposited dielectrics include silicon dioxide[41] and parylene[54]. An example of a solution-processed dielectric layer is poly-4-vinylphenol (PVP)[55], which is deposited by spin-coating and then cross-linked at 200°C.

Patterning is a crucial part of the fabrication of OTFTs. The organic semiconductor must be confined to the channel region to eliminate parasitic leakage and reduce cross-talk in order to achieve better device performance[56]. The drain, source, and gate electrodes need to be patterned with the required feature size depending on the application. Typically, the smaller the distance between the drain and source electrode (channel length), the higher the current output and the faster the transistor switching speed. The following sections will discuss some typical patterning methods used for OTFT fabrication. The most desirable methods involve direct printing of the active materials, in which the patterning and deposition are carried out in one single step. Such methods provide the possibility of processing over a large area, increasing production throughput and therefore reducing the cost per device[30].

Optical lithography is a well-developed conventional technique for the patterning of mesoscopic features and components for microelectronic and photonic devices[57]. In this process, geometric shapes from a mask are transferred to a substrate (e.g. Si wafer), enabling patterning of the active materials and electrodes. Both metal and conducting polymer electrodes can be fabricated using standard photolithography followed by lift-off[12,58]. Conducting polymer electrodes have also been patterned by light exposure to change their conductivity without having to remove the polymer in the channel region[3,49]. Although optical lithography can achieve 100 nm resolution, it is a relatively expensive process. This method is also less suitable for the patterning of organic semiconductors because the exposure of organic semiconductors to solvents and etchant tends to cause degradation in device performance.

Screen printing involves squeezing a specially prepared ink through a screen mask onto a substrate surface to form the desired pattern. This method is capable of printing all the active components in OTFTs[59], but has limited feature size resolution (75 μm or larger). Components of OTFTs can also be deposited using ink-jet printing, which is similar to the operation of a conventional ink-jet printer, but uses specially formulated inks[60,61]. Resolution on the order of 25 μm can be achieved without surface modifications, while hydrophobic dewetting patterns[62] can be used to obtain resolutions approaching 200 nm. This hybrid method, in which ink-jet printing and traditional photolithography are combined to achieve the resolution necessary for device applications, is being pursued by Plastic Logic[63]. An additive dry printing method[53] for depositing conducting polymers has been developed using a thermal (laser) imaging technique for the

ablative transfer of a patterned layer onto a flexible receiver layer with resolution down to 5 μm. This can be used to process successive layers without the use of a solvent that could degrade the underlying organic layers.

Soft lithography encompasses a wide variety of patterning techniques in which a master structure is fabricated in a material, such as Si, by conventional lithographic processes, and then used to make elastomeric replicas in a material such as poly(dimethylsiloxane) (PDMS). In microcontact printing, elastomeric stamps are used for molecular transfer to produce a contact-induced chemical modification of a surface[64-66]. The chemical modification can produce hydrophobic and hydrophilic patterns allowing for selective solution phase deposition of the organic semiconductor. Alternatively, an alkanethiol protecting layer can be microcontact printed on Au or Ag to prevent the metal underneath from being etched away to form electrode patterns[2]. In soft lamination, source and drain electrodes are deposited on one substrate, then laminated onto another substrate that already contains the gate, dielectric, and semiconductor, thus completing the transistor[67].

Applications

Organics have long been attractive for use in electronics because of their light weight, flexibility, and low cost compared with their Si counterparts. Recent increases in performance, however, have rapidly expanded organic FETs from niche markets, making them targets for a wider range of applications.

Organics offer potential advantages in displays, where TFTs are implemented as switches to activate individual pixels. Hand-held devices (cell phones, PDAs, etc.) with ultrathin displays can achieve higher resolution and information content, while new technologies, such as flexible displays and electronic paper, are potentially revolutionary advancements. Integrated smart pixels, with an OTFT switching an organic light-emitting diode (OLED) pixel, have been demonstrated, even though actual OTFT active-matrix OLED displays are yet to be demonstrated[19,20,68,69].

An alternative to active-matrix flexible displays is an innovative example by E-ink utilizing an OTFT backplane with a laminated electronic ink frontplane, consisting of a layer of electrophoretic microcapsules on a transparent electrode[70,71]. The OTFT backplane controls the contrast of the display by moving charged black and white pigments to the transparent layer, as shown in Fig. 6. In late 2000, E-Ink presented the world's first flexible (16 cm x 16 cm) electronic ink display using an OTFT backplane circuit created by Lucent, consisting of an array of 256 transistors fabricated using a low-cost, rubber stamp printing process[2]. The printed transistors from Lucent and a typical flexible display are presented in Figs. 7 and 8, respectively. Plastic Logic, a company actively developing ink-jet-printed plastic TFTs, subsequently has demonstrated a bistable reflective display driven by an ink-jet-printed active-matrix backplane together with Gyricon Media, the provider of SmartPaper™ reusable display material[63]. This first experimental prototype is a display featuring 3024 pixels (63 x 48) at 50 dpi on a glass substrate. More recently, Philips and E-Ink jointly demonstrated a similar electronic ink display driven by OTFTs[3] with 320 x 240 pixels, a diagonal length of 127 mm, a resolution of 85 dpi, and a bending radius of 2 cm. Philips has also announced
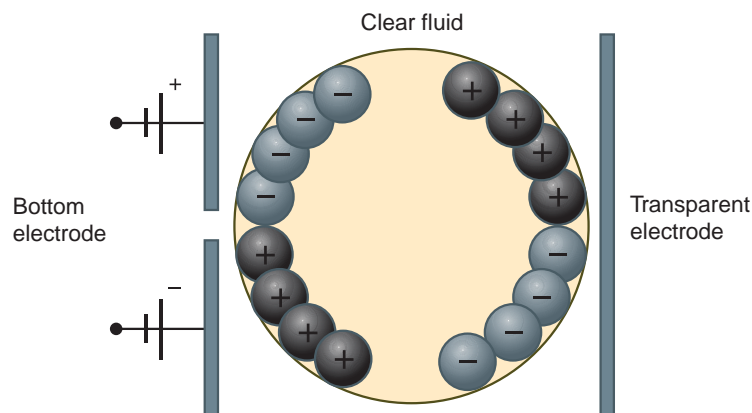

Fig. 6 Schematic of an E-ink display[70]. An OTFT backplane addresses each element, with encapsulated, charged pigments shifting to the transparent electrode surface.
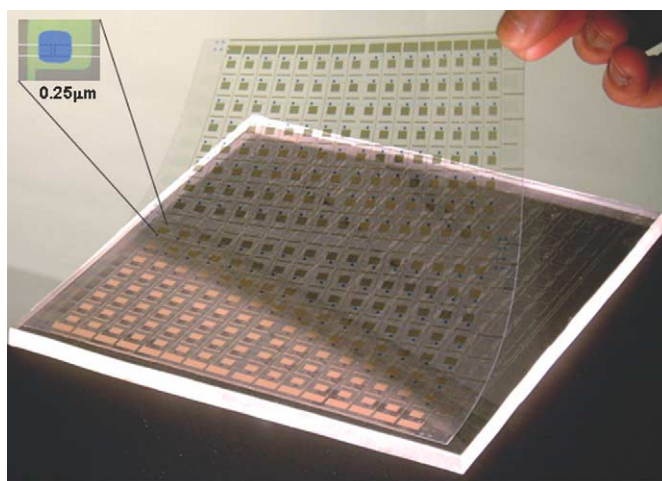
Fig. 7 A 256-transistor array produced by Lucent using a rubber stamp printing process. (Reprinted with permission from[76]. © 2001 Materials Research Society.)

that it had formed a technology incubator company, Polymer Vision, to partner with other companies interested in ultrathin, rollable displays that could double as electronic paper.

Building upon the basic OTFT component, complementary metal-oxide-semiconductor (CMOS) technology now shows promise in organic semiconductors. The ready availability of both n- and p-channel semiconductors and recent discoveries of ambipolar functionality, where a single semiconductor layer is capable of conducting both electrons and holes, is enabling the design of robust circuitry with low heat dissipation[41,72]. This ambipolar behavior has been realized for an n- and p-type polymer dispersed layer, and for a single organic material with a low band gap, and thus a low barrier for electron and hole injection[73,74].

RFID cards are made possible with this complementary circuitry for applications involving identification, verification, and tracking. Tasks similar to inventory management can be immensely simplified by exploiting a system of many transponder circuits with a single reading instrument[75]. Other low-resolution applications involving logic functions, like smart cards, or disposable sensors are also made possible[8].

Summary

In conclusion, the physical properties and relatively facile processing of organic materials allow for the demonstration
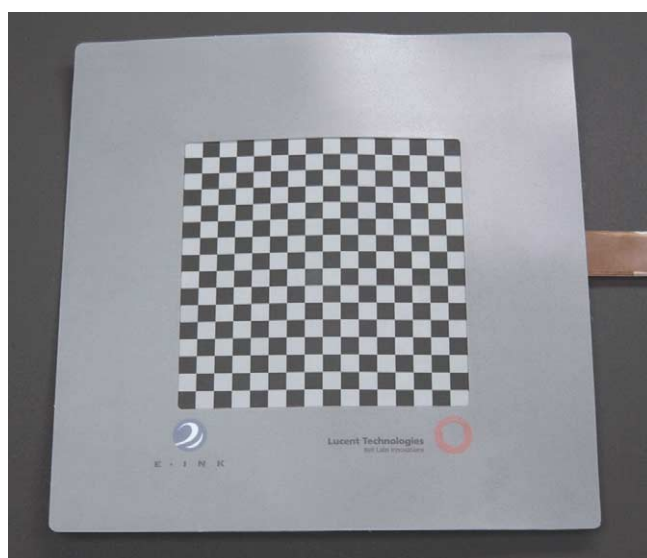

Fig. 8 The world's first flexible electronic ink display driven by organic transistors. (Reprinted with permission from[2]. © 2001 National Academy of Sciences USA.)

of flexible, low-cost, large-area devices using OTFTs. Advances in fabrication methods and the development of higher-performance semiconductor materials have improved on existing technology and expanded the scope of potentially realizable applications. Much of this progress has been a result of the deduction of structure-property relationships of the active layer component. Molecular tuning based on this understanding has yielded materials with better electronic properties and simplified processability. The discovery of ambipolar functionality of organic materials has increased OTFT versatility, enabling their incorporation in complementary logic devices. The numerous applications presented thus far have already showcased the potential of OTFTs, which will continue to grow. While challenges exist for large-scale manufacturing, their rapid development shows great promise for their future in plastic electronics. MT

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