Recent advances in ZnO transparent thin film transistors

E. Fortunato*, P. Barquinha, A. Pimentel, A. Gonçalves, A. Marques, L. Pereira, R. Martins

Materials Science Department, Faculty of Sciences and Technology of New University of Lisbon and CEMOP-UNINOVA, Campus da Caparica, 2829-516 Caparica, Portugal

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Abstract

Zinc oxide is a well-known wide band gap semiconductor material (3.4 eV at room temperature, in the crystalline form), which has many applications, such as for transparent conductors, varistors, surface acoustic waves, gas sensors, piezoelectric transducers and UV detectors. More recently, it is attracting considerable attention for its possible application to thin film transistors. In this paper, we present some of the recent results already obtained as well as the ones that are being developed in our laboratory. The main advantage presented by these new thin film transistors is the combination of high channel mobility and transparency produced at room temperature which makes these thin film transistors a very promising low cost device for the next generation of invisible and flexible electronics. Moreover, the processing technology used to fabricate this device is relatively simple and it is compatible with inexpensive plastic/flexible substrate technology.

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1. Thin film transistors: historical background

Even though the field-effect transistor did not come into widespread use until the 1960s, its invention predated both the junction and point-contact transistors by many years. As it is normal with many innovations, its practical realization was delayed until adequate materials and technologies were available for its fabrication. We can even say (and for most of us could be a surprise) that the thin film transistor (TFT) was the first solid-state amplifier ever patented. The basic principle of the field-effect transistor (what we call now JFET) was proposed by the first time by Julius Edgar Lilienfeld as early as 1925 and patented in 1930 [1] (see Fig. 1a where an adaptation of the cross section of the device is shown). The field-effect transistor he described was probably the first successful solid-state amplifier invented. The advantages of this device over the vacuum tubes, the only alternative high frequency amplifier available at that time, are also described in this patent and in later ones. Lilienfeld’s later patent from 1933 [2] described a new field-effect transistor (called now by MISFET) where he specifies the thickness of the dielectric layer, which insulates the metal control electrode from the copper sulfide channel, to about $10^{-5}$ cm (1000 Å). This is, in fact, the typical thickness used in the insulated-gate transistors, which were developed many years later.

Nevertheless, the first functional working TFT was demonstrated by Weimer in 1962 [3]. He used thin films of polycrystalline cadmium sulfide, similar to those ones developed for photodetectors. The simplified structure is shown in Fig. 1b. Other TFT semiconductor materials like CdSe, Te, InSb and Ge were investigated, but in the mid-1960s with the emergence of the metal oxide semiconductor field effect transistor (MOSFET) based on the crystalline silicon technology and the possibility to perform integrated circuits, led to a decline in TFT development activity by the end of the 1960s.

What dramatically changed the prospects for TFTs in the 1970s was the realization that with crystalline silicon, low cost was inseparable from miniaturization, whereas some applications required large arrays of low cost electronics, like for example displays. By this time, many researchers and engineers have been engaged in improving the characteristics of liquid crystal displays (LCDs), recently
discovered. The first active matrix LCD was successfully demonstrated by Brody et al. in 1973 [4], where a CdSe TFT was used as a switching element for each pixel of a 120 × 120 matrix.

In spite of the many successful demonstrations of CdSe TFT LCDs, the industry did not enter this market until the report on the feasibility of doping amorphous silicon (a-Si:H) by the glow discharge technique [5,6]. After that and in 1979, LeComber, Spear and Ghaith described a TFT using a-Si:H as the active semiconductor material [7], whose structure is indicated in Fig. 1c.

After LeComber reported the first a-Si:H TFT, many laboratories started the development of AMLCDs formed on glass substrate. Although this result attracts much attention, the major disadvantage of a-Si:H TFT is its low electron mobility that limits the ultimate speed of devices. However, an adequate device speed for the switching applications in the LCD has been achieved.

Since the mid-1980s, the silicon-based thin film transistors become the most important devices for active matrix liquid crystal displays (AMLCDs) and have successfully dominated the large area LCD product market [8].

The year 1990 marked the debut of a new class of TFT, based upon organic semiconductor active layer material [9], with electron mobilities similar to that of a-Si:H. This new class of TFTs are very promising candidates for integration onto flexible plastic substrates for a future generation of rugged, lightweight displays than can be rolled up like a map.

More recently, a new generation of oxide semiconductors are being studied and applied as the active material to TFT, in special zinc oxide (ZnO). This will be described in the next sections.

2. Zinc oxide TFTs

Transparent electronics are nowadays an emerging technology for the next generation of optoelectronic devices. Oxide semiconductors are very interesting materials because they combine simultaneously high/low conductivity with high visual transparency and have been widely used in a variety of applications (e.g. antistatic coatings, touch display panels, solar cells, flat panel displays, heaters, defrosters, optical coatings, among others) for more than a half-century. Transparent oxide semiconductor based transistors have recently been proposed using as active channel non-doped ZnO [10–18]. One of the main advantages exhibited by these transistors lies on the magnitude of the electron channel mobility leading to higher drive currents and faster device operating speeds. The mobility reported in the literature is ranging from 0.2 to 7 cm²/V s with an on/off current ratio from 10⁵ to 10⁷ and a threshold voltage (V_TH) between −1 and 15 V. To date, ZnO channel layers have been deposited using substrate heating or submitted to post-thermal annealing in order to increase, mainly the crystallinity of the ZnO layer and so the film’s mobility. The main advantage of using ZnO deals with the fact that it is possible to growth at/near room temperature high quality polycrystalline zinc oxide, which is a particular advantage for electronic drivers, where the response speed is of major importance. Besides that, since ZnO is a wide band gap material (3.4 eV), it is transparent in the visible region of the spectra and therefore, also less light sensitive.

Thin film transistors based on a-Si:H technology present some limitations like: light sensitivity and light degradation accompanied with a low mobility (<2 cm²/V s). On the

<table>
<thead>
<tr>
<th>Technique</th>
<th>Temperature (°C)</th>
<th>Channel mobility (cm²/V s)</th>
<th>On/off ratio</th>
<th>V_TH (V)</th>
<th>I_sat (μA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ion beam [11]</td>
<td>600–800</td>
<td>0.3–2.5</td>
<td>10⁶</td>
<td>10–20</td>
<td>70</td>
</tr>
<tr>
<td>Sol gel [15]</td>
<td>700</td>
<td>0.2</td>
<td>10⁷</td>
<td>not mentioned</td>
<td>14</td>
</tr>
<tr>
<td>Pulsed laser [10]</td>
<td>450</td>
<td>0.031–0.97</td>
<td>10⁶</td>
<td>−1.0 to 2.5</td>
<td>80</td>
</tr>
<tr>
<td>rf magnetron sputtering [17,18]</td>
<td>room temperature</td>
<td>20–70</td>
<td>5×10⁵</td>
<td>1.8</td>
<td>1000</td>
</tr>
</tbody>
</table>

* The devices present some instability and after several measurements they collapse.
other hand, the polysilicon TFT in spite exhibiting a high mobility (50 cm²/V s=μ≤500 cm²/V s) their opacity limits the aperture ratio for active matrix arrays, highly important when for instance, organic light emitting diodes (OLEDs) have to be addressed. Apart from that, if flexible substrates based on polymers are intended to be used, the processing temperature is also a quite limiting factor. One possible way to overcome such problems is the utilization of efficient and reliable oxide based thin film transistors. Table 1 presents the state of the art concerning fully transparent ZnO-TFTs.

3. Experimental details

3.1. ZnO optimization

Our ZnO films (doped and undoped) were deposited onto soda lime glass substrates by rf (13.56 MHz) magnetron sputtering using a ceramic oxide target ZnO from Super Conductor Materials, Inc. with a purity of 99.99% and 2 in. diameter. The sputtering was carried out under room temperature and the argon deposition pressure was 0.15 Pa. The distance between the substrate and the target was 10 cm and the rf power was changed between 50 and 175 W. The deposition rate was varied between 15 and 30 nm/min. The film thickness was measured using a surface profilometer (Dektak 3 from Sloan Tech.). The electrical resistivity (ρ) was measured as a function of temperature in the range of 300 to 500 K using thermally evaporated aluminium electrodes in a coplanar configuration. X-ray diffraction measurements were performed at room temperature in air, using Cu-Kα line (Rigaku DMAX III-C diffractometer). The surface morphologies were analyzed using a Field Emission Scanning Electron Microscope (FE-SEM, S-1400 Hitachi). The optical transmittance measurements were performed with a Shimadzu UV/VIS 3100 PC double beam spectrophotometer in the wavelength from 200 nm to 2500 nm.

3.2. ZnO-TFT fabrication

Fig. 2 shows a schematic illustration of the ZnO-TFT produced, using only oxide materials.

In order to evaluate the performances of the TFTs, different channel width and channel lengths were used, varying the W/L ratio from 1 to 65.

The devices were electrically characterized in air at room temperature, in the dark using a microprobe station connected to a semiconductor parameter analyzer (model HP 4145B). The reproducibility of the device characteristics was confirmed by measuring more than 200 TFTs, each one several times.

The gate dielectric of the ZnO-TFT is based on a engineered insulator consisting of a superlattice of Al₂O₃ and TiO₂ (ATO) [19] deposited on a glass coated with an indium tin oxide (ITO) 200 nm thick films, exhibiting a transparency of 85%, a resistivity of 2.3×10⁻⁴ Ω cm, a carrier concentration of 7.7×10²⁰ cm⁻³ and a mobility of 36 cm²/V s. The average capacitance of the ATO layer measured nearby the TFTs produced was 55 nF/cm², having a total thickness of 220 nm. From this measurement, the relative dielectric constant was determined as being around 14. The electrical resistivity of the ATO layer was 3.6×10¹⁵ Ω cm and the breakdown field is higher than 4 MV/cm.

The channel is based on undoped ZnO, 100 nm thick, whose electrical properties were properly controlled during the deposition pressure aiming to obtain a material where the conduction is dominated by electrons. In this case, the background electron concentration must be reduced in order to increase the carrier’s mobility. This attempt was achieved by optimizing the deposition conditions for undoped ZnO films produced by rf magnetron sputtering. To do so, we choose to vary the rf power instead of the oxygen partial pressure (easier for controlling the stoichiometry), because the use of oxygen during the deposition is not compatible with the lift-off technique used (the photoresist is removed by a plasma that contains oxygen ions) and also to minimize the defects at the channel–insulator interface. Moreover, the processing technology used is relatively simple and it is compatible with inexpensive plastic/flexible substrate technology.

The drain and source are based on highly conductive gallium doped zinc oxide (GZO) [20], 150 nm thick and patterned by the lift-off technique. These depositions were also carried out at room temperature.

4. Results

Fig. 3 shows the dependence of the electrical resistivity and the average optical transmittance at the visible spectra (between 400 and 700 nm), as a function of rf power density.
The highest resistivity (of $10^8 \ \Omega\ cm$) was obtained for $P=5\ W/cm^2$. For $P$ around $5\ W/cm^2$, the films became close to stoichiometry with low structural defects and consequently higher resistive. As we decrease or increase the rf power density from $5\ W/cm^2$, a deviation from stoichiometry is obtained accompanied by a decrease on the electrical resistivity, due to a lower carrier concentration or/and electron mobility.

This is also confirmed by a decrease on the optical transmittance, especially for rf power densities lower than $5\ W/cm^2$ and for the dark conductivity as a function of temperature, as it is indicated in Fig. 4. Here, the dark conductivity as a function of absolute temperature for two ZnO films produced at 2.5 and 5 W/cm$^2$, respectively, is presented. The data show the conductivity to be thermally activated, presenting the film deposited at 5 W/cm$^2$ an oxide/semiconductor-like behavior, while for the other one the film exhibits a semi-metallic behavior. From the data, it is possible to observe two types of conductivity giving rise to two different activation energies. Since we are in the presence of a polycrystalline semiconductor oxide, with grains surrounded by regions with a higher concentration of oxygen, we have the influence of such regions on the measured conductivity as a function of temperature. More studies are going on in order to understand this type of behavior.

For the films with lower resistivity ($<10^2\ \Omega\ cm$), we measured Hall mobilities of about $2\ cm^2/V\ s$ and a carrier concentration of $3 \times 10^{16}\ cm^{-3}$ (n-type). For these undoped films, an excess of interstitial Zn ions or/and oxygen vacancies can contribute with free electrons to the electrical conduction. Concerning the ZnO films used for the TFTs, due to the high value of the electrical resistivity ($\sim 10^8\ \Omega\ cm$), it was not possible to measure the Hall mobility (out of the range of the Hall effect equipment used).

The X-ray diffraction patterns of these films are shown in Fig. 5a. As $P$ increases, the diffraction peak corresponding to the (002) orientation becomes predominant up to a rf
power density of 5 W/cm², where the resistivity is maximum. Besides that, the data also show that this film mainly constitutes of dense and small crystallites (nano-crystalline with sizes up to 10 nm). For higher rf power densities, the peak disappears, meaning that the crystallinity of the ZnO degrades, owing to a decrease on film oxidation. That is, the amorphous phase start is being predominant.

Fig. 5b shows the X-ray diffraction pattern for the ZnO film produced at \( P = 5 \text{ W/cm}^2 \) for a wider \( 2\theta \) range. For all the films produced, only the ZnO (002) peak at \( 2\theta = 34.1^\circ \) is observed, revealing that the films are nanocrystalline with a hexagonal structure and a preferred orientation with the c-axis perpendicular to the substrate, which is also consistent with the results obtained by scanning electron microscopy (see Fig. 6). The large width of the peaks reveals a limited crystalline domain. For the film produced at \( P = 5 \text{ W/cm}^2 \), using the full-width at half-maximum of the peak at an angle \( 2\theta = 34.1^\circ \), we calculated by using the Scherrer formulation an average crystallite size of the order of 10 ± 1 nm. The low crystallite size is also associated with the fact that the films were processed at room temperature without any type of post-thermal annealing.

Fig. 7a shows the source-to-drain current \( I_{DS} \) as a function of the gate voltage \( V_{GS} \). It is observed that the ZnO-TFT has an n-channel, since electrons are generated by the positive \( V_{GS} \). A high \( I_{DS} \) = 230 \( \mu \text{A} \) is obtained for \( V_{GS} = 40 \text{ V} \) and \( V_{DS} = 20 \text{ V} \). In addition, we clearly observe a hard saturation current at the post pinch-off voltage, in accordance with the standard theory of field-effect transistors. The saturation mobility \( \mu_{sat} \) and the threshold voltage \( V_{TH} \) were calculated by fitting a straight line to the plot of the square root of \( I_{DS} \) vs. \( V_{GS} \), according to the expression:

\[
I_{DS} = \left( \frac{C_i \mu_{sat} W}{2L} \right) (V_{GS} - C_{TH})^2 \quad \text{for} \quad V_{DS} > V_{GS} - V_{TH}
\]

for the saturation region, where \( C_i \) is the capacitance per unit area of the gate insulator. The obtained \( \mu_{sat} \) is around \( \mu_{sat} = 20 \text{ cm}^2/\text{V s} \) and the \( V_{TH} = 21 \text{ V} \), showing that the ZnO-TFT operates in the enhancement mode. Enhancement mode is preferable to depletion mode since it is not necessary to apply a gate voltage to switch off the transistor, because the circuit design is simpler and the power dissipation is lower. The high value of \( \mu_{sat} \) agrees with the effective mobility \( \mu_{FE} = 19 \text{ cm}^2/\text{V s} \) [21] determined for \( V_{DS} \leq 2 \text{ V} \) (in order to guarantee a uniform distribution of carriers in the channel along the source to drain region). Here we also have to notice that the average field effect mobility \( \mu_{FE} \) obtained from the transconductance [21] data in the same \( V_{DS} \) region and for \( 30 \text{ V} \leq V_{GS} \leq 40 \text{ V} \) is \( \mu_{FE} = 21 \text{ cm}^2/\text{V s} \). These data confirm the high quality of the undoped ZnO films (improved crystallinity fraction of the nanocrystals and low oxygen vacancies and/or Zn interstitials working as donors) as well as the good channel–insulator interface obtained. The magnitude obtained for the \( V_{TH} \) is directly proportional to the gate insulator thickness. So, these values could be easily reduced by reducing the gate insulator thickness. The off-current is low, on the order of \( 1.3 \times 10^{-9} \text{ A} \), and the on/off ratio is \( 2 \times 10^5 \). That is, the TFT is able to sustain currents as high as \( 2.3 \times 10^{-4} \text{ A} \). The gate voltage swing \( S \) defined as the voltage required to increase the drain current by a factor of 10 was 1.24 V/decade for the ZnO-
TFT under analysis. The $S$ is given by the maximum slope in the transfer curve, Fig. 7a.

Fig. 7b shows the $I_{DS}$ current curves as a function of the drain current $V_{DS}$ for different $V_{GS}$. The saturation of $I_{DS}$ was about 230 $\mu$A under a gate bias of 40 V. Besides, the high value obtained for the saturation current, the device exhibits “hard” saturation, evidenced by the flatness of slope of each $I_{DS}$ curve, for large $V_{DS}$, without the need of post-heating treatments. This indicates that the entire thickness of the ZnO channel layer is depleted and so the device exhibits a high resistance (defined as the inverse of the slope of the $I$–$V$ curve for a given gate voltage, in the off saturation regime) larger than 20 M$\Omega$, as required for most circuit applications. The on resistance ($R_\text{ON}=1/g_d$) defined as:

$$R_\text{ON} = \frac{L}{\mu_{\text{eff}}C_iW(V_{GS} - V_{TH})}$$

(2)

is of about $R_\text{ON}=45$ K$\Omega$ ($V_{GS}=30$ V). The relation between the $R_{OFF}$ and $R_\text{ON}$ is within what is expected for fast switching device behaviors. Besides that, the device response time, defined as being proportional to $R_\text{ON}C_i$ presents values of $R_\text{ON}C_i=25$ ps/$\mu$m$^2$, highly promising for device miniaturization.

We have also investigated the effect of $W/L$ on the channel mobility as it is presented in Fig. 8. The data show that as $W/L$ increases, the value for $\mu_{\text{sat}}$ decreases due to sidewall effects associated with source/drain resistance (see the insert in Fig. 8) and to fringing electric field effects that lead to an additional current flow beyond device edges. As the length of the TFT decreases, we will have a more pronounced influence of the region beneath the drain/ source, consequently with the increase of the resistance as well as with the increase of the electric field, reducing the channel mobility. In order to overcome these effects, the miniaturization of the TFT should be accomplished by using small $W/L$ ratios, or by decreasing as much as possible the side wall effects, through a different device configuration. A similar effect was already observed in amorphous silicon TFTs [22].

Fig. 9 shows the optical transmission spectrum of the entire ZnO-TFT in the wavelength range between 200 nm and 2500 nm (including the glass substrate with 1.1 mm thickness). The average optical transmission in the visible part of the spectrum is 80%, while for 550 nm (maximum sensitivity for the human eye) is 85%, which indicates that transmission losses due to the ZnO-TFTs in comparison with the uncoated glass substrate are negligible (7%). The absorbance at the near infrared region is associated to the increase of the carrier concentration of the ITO and GZO films, which leads to a lower value associated to the plasma frequency [23].
The exposure to ambient light of these ZnO-TFTs has no effect on the current–voltage characteristics, which is an advantage in electronic drivers for displays. We have also measured the electrical characteristics of the firsts ZnO-TFTs produced, after 9 months, and no degradation effect was observed. Fig. 10 shows the transfer characteristic of a typical ZnO-TFT. The $V_{TH}$ decreases slightly (about 6%), the saturation mobility remains the same, while the dark current increases from $4 \times 10^{-9}$ A to $1.5 \times 10^{-8}$ A, causing a small decrease in the on–off ratio. In contrast to what happens to amorphous silicon TFTs, the electrical properties presented by ZnO-based TFTs are slightly improved with time.

5. Conclusions

We have presented some of the recent progress obtained on ZnO TFTs. We have also demonstrated the possibility to produce fully transparent high quality ZnO-based TFT at room temperature by rf magnetron sputtering on glass substrates. ZnO is one of a few oxides that can be grown as a polycrystalline or even as a high compact and dense nanocrystalline material at relatively low or even at room temperature on a variety of substrates, such as amorphous glasses and plastics or metal foils. This particularity enables one to realize efficient and reliable TFTs with high mobility’s, and thus opens new doors for so-called “invisible electronic circuits”, highly important for the next generation of invisible and flexible electronics, such as switching for addressing organic light emitting matrices.

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References