Reliability of OTFTs on flexible substrate: Mechanical stress effect

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Abstract

Flexibility will significantly expand the application scope of electronics, particularly large-area electronics. Over the last ten years, printed organic electronic is believed to be one of the next major technological breakthroughs in the field of microelectronic and the use of printing technology to process organic field-effect transistors (OFETs) opens promising perspectives for low cost, large area circuits integrated on flexible, plastic substrates. With amorphous polymer-based thin films transistors acceptable electrical performances are now achieved with relatively good stability at ambient air. In literature a lot of work has been devoted to study degradation of device characteristics under bias stress conditions but only few papers deal with the mechanical behaviour. In this paper, we review our first reliability results obtained on flexible organic thin film transistors under mechanical stresses. The variations of electrical characteristics under bending tests, both in compression and tension, have been studied. Using specific equipment, we have also evaluated the reliability of transistors under cyclic bending tests. The stress dependency of the transfer characteristic deviates from the one observed for inorganic material like silicon.

1. Introduction

Organic transistors based on conjugated polymers or small molecules have been intensively studied in recent decades, and their performance has continually improved [1] with understanding and progress on chemistry design and synthesis of solution-processed organic semiconductors [2] [3]. Organic thin films transistors (OTFTs) have the advantages of flexibility, large area capability, low cost, and low temperature processing, and so have various potential applications such as in displays [4], radio frequency identification tags [5], and large area sensors [6] on flexible substrates. These applications are commonly subjected to bending strains, which could modify the performance of organic devices. One of the next challenges is now to demonstrate the robustness and reliability of OTFTs to support the use in the frame of these applications. While degradation of device characteristics under bias stress conditions has been extensively studied [7], only few papers deal with the effect of mechanical stress on the electrical characteristics. In particular, small molecules-based OTFT and changes in mobility observed in bent organic TFT devices have been reported to result from changes in the energy barrier to hole hopping due to variation of the intermolecular spacing under a bending strain [8]. In this paper, we aim to investigate the effect of mechanical stresses on our printed OTFT characteristics.

2. Material and methods

In this work, the mechanical behaviour of a printed p-channel organic field effect transistor is studied.

Samples are achieved in the CEA-Liten laboratories. The structure of the transistor is a “top gate – bottom contacts” design (Fig. 1.b). The semi-conducting layer is a screen-printed triarylamine copolymer film, which is a well known family of air stable and solution-processed P-channel semiconductor [9]. The non-planar conformation of the triarylamine and the lack of order make this class of semi-conducting material suitable for printing and annealing processes. Gold source and drain contacts were patterned by laser ablation on a 125 µm plastic PolyEthylene

Naphthalate (PEN) substrate from Dupont Teijin sputtered with 30 nm of gold. Above the organic semiconductor, a 1µm thick low k perfluoropolymer dielectric layer (Cytop®) [10] and a silver conductive gate have been also screen printed. Each 5 cm x 5 cm test sheet contains 30 transistors with channel lengths of 50 µm, 80 µm and 100 µm and channel widths ranging from 500 µm to 2 mm. For fatigue tests, larger test sheets 8 cm x 20 cm, also composed 30 transistors have been designed to fit the machine.

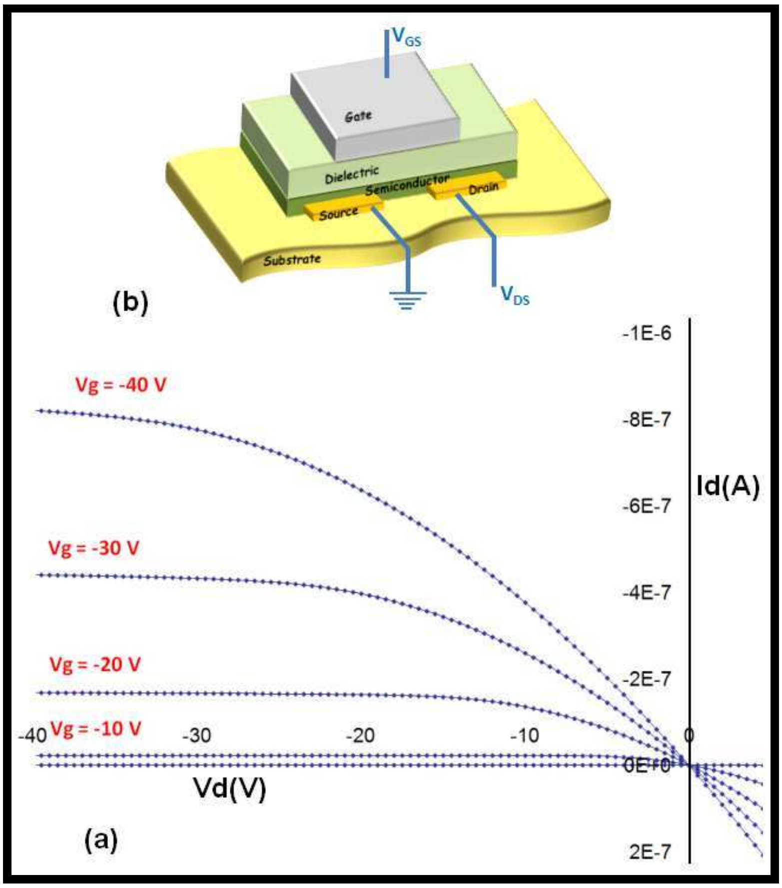

Fig.1: (a) illustration of “top gate – bottom contacts” organic TFT on PEN substrate. (b) Typical dc characteristics without stress, showing source-drain current I_D as function of source-drain voltage V_D. Gate voltage V_G is changed from 0 to −40 V in −10 V steps.

Electrical characterizations of the OTFTs have been performed in the dark, in air without encapsulation, using a Keithley 4200 semiconductor analyser combined with a semi-automatic Süss Microtech prober. Transistors parameters such as saturation mobility, threshold voltage and $I_{on/off}$ ratio were extracted following the 1620TM IEEE standard test procedure. Figure 1.a shows the typical output characteristics without stress, source-drain current I_{DS} is monitored as a function of source-drain voltage V_{DS} and gate voltage V_{GS} was changed from 0 to −40 V in −10 V steps. The measured mobility μ was 0.03 cm^2 /V.s and the *on/off ratio* was 10^5.

3. Results and discussion

Bending tests both in tension and compression have been performed while recording the electrical characteristics of the transistor (Fig. 2). Curvature radii *R* from 50 mm down to 3 mm have been tested in tension. Due to probing problems, only large radii between 50 and 30 mm have been achievable in compression mode.

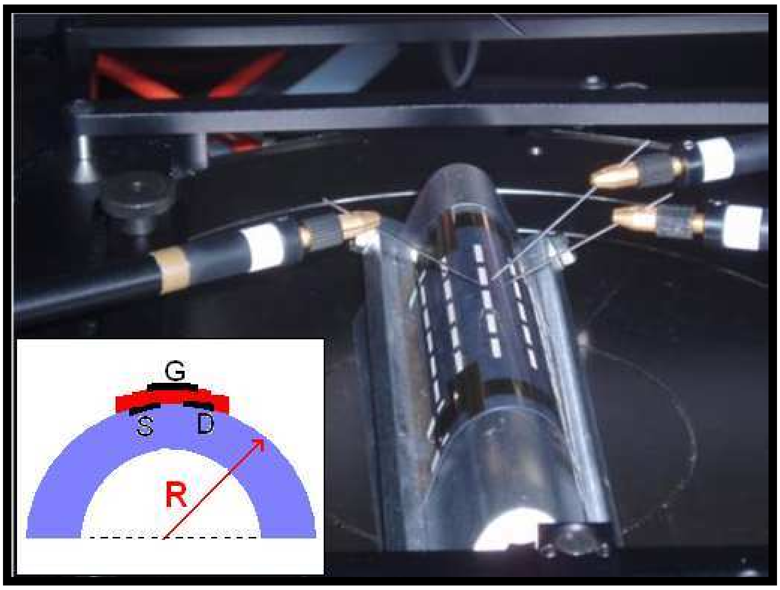

Fig.2: Photograph and Schematic illustration of electrical characterization by tips probing under tensile bending. Using half-cylinder forms with Curvature radiuses *R* from 50 mm (ε = 0.13%) down to 3 mm (ε = 2.08%).

Since the transistor layers are very thin (µm or below) compared to the PEN substrate thickness t_s, absolute value of the mechanical strain ε in the transistor can be evaluated from bending radius using the following formula (Eq.1) [11]. By convention, compressive strains are negative values while tensile strains are positive.

$$|\varepsilon| = (t_s / 2R) \qquad (1)$$

Transistors are tested with two different stress directions under identical conditions so that their source-drain current paths may be arranged precisely perpendicular and parallel to the direction of strain, referred to as perpendicular position and parallel position, respectively.

The shape of I_{DS}-V_{GS} and I_{DS}-V_{DS} characteristics under bending strain remains close to the initial one (Fig.3.a and Fig.3.b respectively). The transistor still works normally under bending stress but presents electrical performance variations. Gold source and drain electrodes give good ohmic contact for all I_{DS}-V_{DS} curves and contact resistance does not change drastically. Leakage current increases by one decade : from 1pA to 10 pA.

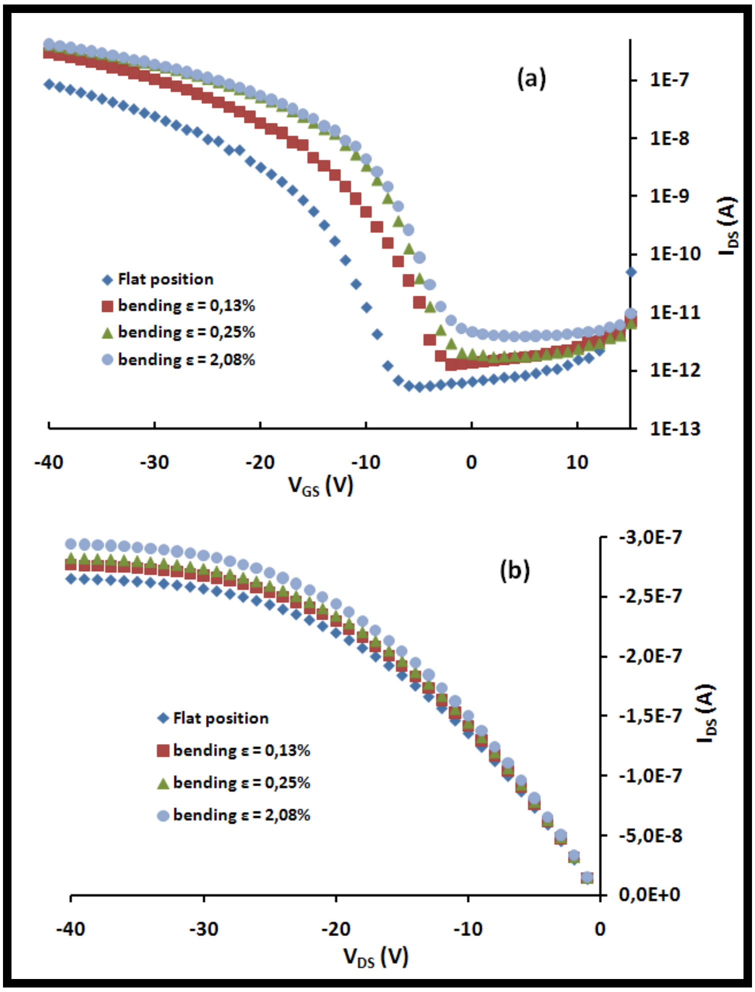

Fig.3: I_{DS}-V_{GS} characteristics at V_{DS} = -40V (a) and I_{DS}-V_{DS} characteristics at V_{GS} = -40V (b) with bending strains ε from 0 (flat position) to +2.08% (bending radius of 3 mm).

The electrical properties of transistors were also measured while changing bending radius. Normalized saturation current and mobility have been plotted versus the mechanical strain $\boldsymbol{\varepsilon}$ with the parallel position (Fig.4.a and Fig.4.b respectively). Saturation current increases clearly with increasing tensile strain. The change in saturation current is +12% at bending radius R=3 mm. On the opposite, in compression mode, although we have only investigated small strains, the saturation current tends to decrease. Such large changes cannot be explained only by the deformation of the device structures at R=3 mm, corresponding to strain of only 2.08%. Indeed, using the classical Poisson model (Eq.2) [12] where γ_P and γ_i are Poisson coefficients of semiconductor and gate insulator, respectively, changes in the structural parameters of OTFT, namely, L (channel length), W (channel width), and t_i (the thickness of gate insulator) are evaluated and their insertion in the equation Eq.3 shows that for 2.08% of strain geometrical deformation causes only about -2% of saturation current.

$$W \rightarrow W\,(1-\gamma_P\,.\,\varepsilon)\;;\;L \rightarrow L\,(1+\varepsilon)\;;\;t_i \rightarrow t_i\,(1-\gamma_i\,.\,\varepsilon) \qquad (2)$$

Finally when the stress is released, we recover the initial electrical characteristics of the transistor. The change in source-drain current is reversible and reproducible. This reversibility shows that this phenomenon is not associated with the time-dependent drift of electronic performance.

We evaluated field-effect mobility μ using the equation (Eq.3) [13], where I_{DS} is the source drain current, C_i is the insulator capacitance, and V_T is the threshold voltage, in the saturation mode and plotted it normalized by the value without strain μ_0 in the Fig.3. In this figure the mobility increases by 8% when strain is 2.08% of tension.

$$I_D=(\mu C_i/2)\,(W/L)\,(V_G-V_T)^2 \qquad (3)$$

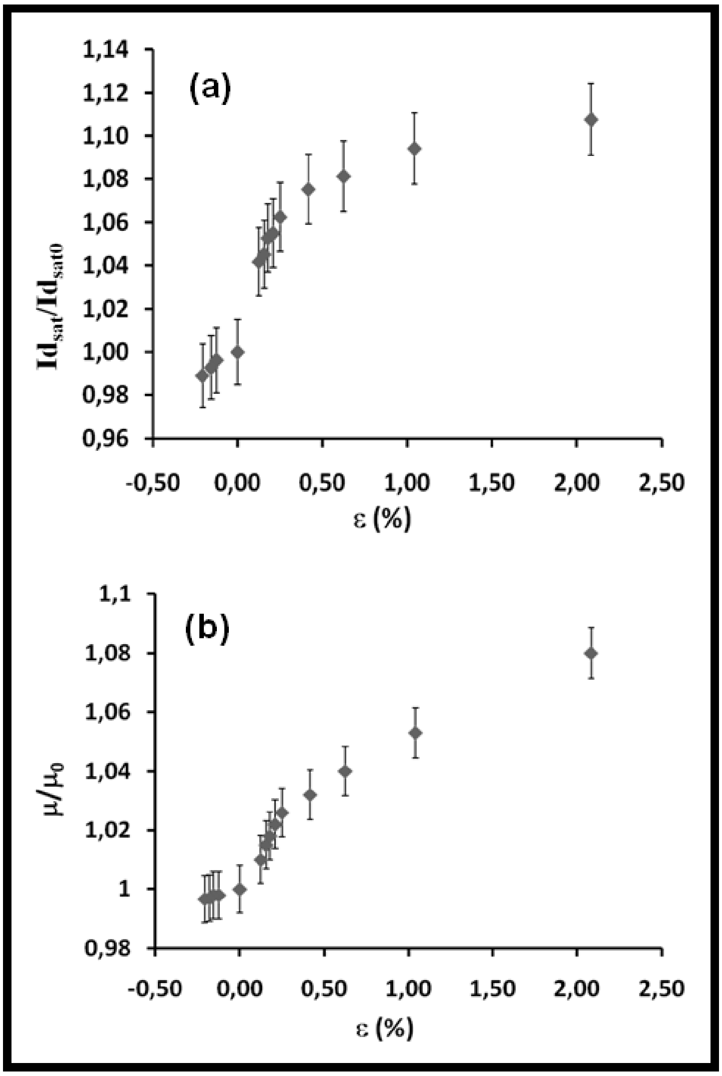

Fig.4: Relative change in (a) saturation current I_{Dsat} and (b) mobility μ plotted as function of strain tensile strain ε. Compressive strains are negative values (only ε from -0.13% to -0.21%) while tensile strains are positive (ε from +0.13% to +2.08%).

The observed strain effect on the mobility of these organic TFTs with amorphous polymer semiconductor is the opposite of the results obtained with crystalline or polycrystalline organic semiconductors like pentacene films [14] [15] when mobility decreases with increasing tensile strain and increases with increasing compressive strain. In this last case, because of the size and the organization of pentacene small molecules, mobility change by mechanical stress is quite reasonable since the transport properties of organic molecular systems follow the hopping model rather than the band model. Under tensile strain, as described in a theoretical study [16] energy barrier for hole hopping increases due to the larger spacing between ordered molecules, resulting in an decreasing in the mobility. In contrast, in the case of our study, the

polytriarylamine has an amorphous and non-planar structure with a very long polymer chains muddled randomly [17] which can explain the opposed behaviour compared to pentacene molecules.
Interestingly, we note that parallel position and perpendicular position show similar strain dependences of mobility, although the directions of strain are perpendicular and parallel to the direction of the source-drain current path for these two different positions. Such an isotropic electrical property indicates that transport is also consistent with the hopping model in amorphous thin films, in which a conducting path from the source electrode to the drain electrode is formed by coupled grain chains oriented in random directions [13].
We have also studied the total resistance variation versus the mechanical strain. Using the transmission line method (TLM), we have observed that the channel resistance decreases while the source and drain contact resistance increases under a tensile bending strain. At tensile strain of about 2% which is close to the maximum strain of gold film [18], the device is still functional and the contact resistance variation shows that charge injection still occurs at source/semiconductor interface; confirmed by the I_{DS}-V_{DS} characteristic. This result could be explained by the fact that, in the transistor configuration, gold electrodes are encapsulated by the organic semiconductor layer which minimizes the degradation of gold/semiconductor interface. The channel resistance change is consistent with the mobility variation under tensile stress. Thus, the increase of the mobility could be explained by a modification of the polymer structure. Indeed tensile stress could induce a slip mechanism of the polymer chains which improve the relative order in the polymer material [19] which leads to an increase of the conductivity [20]. Electrical observation and parameters variation seem to confirm the coherence of this explanation based on literature analysis and although we have no enough clear proof yet of this structural modifications under strain. So the next step of this study will be a detailed structural analysis under stress by AFM measurements and RAMAN spectroscopy [21] [22] in order to highlight the structural variation of the polytriarylamine during mechanical stress.

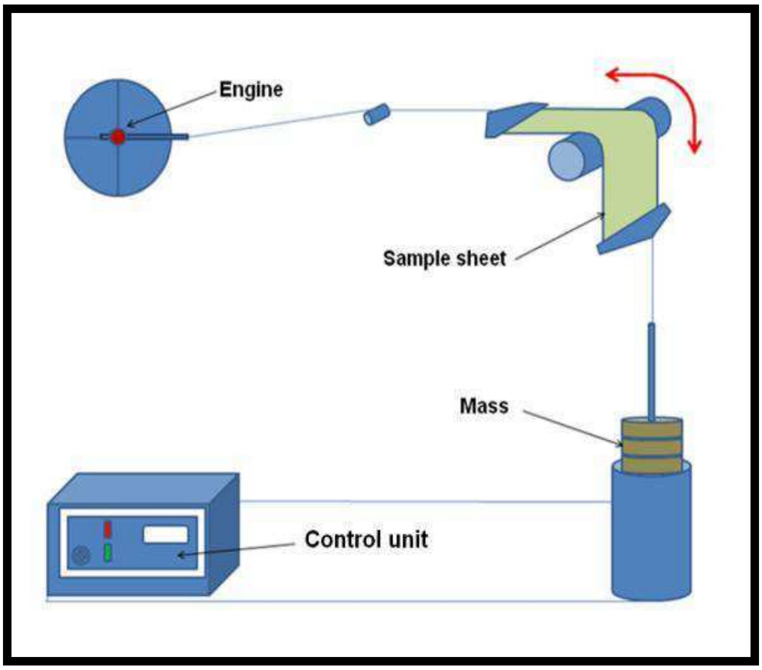

Fig.5: bending fatigue machine. Different curvature radii can be used (5 mm, 10 mm and 25 mm) with a maximum speed of 15 cycles by min.

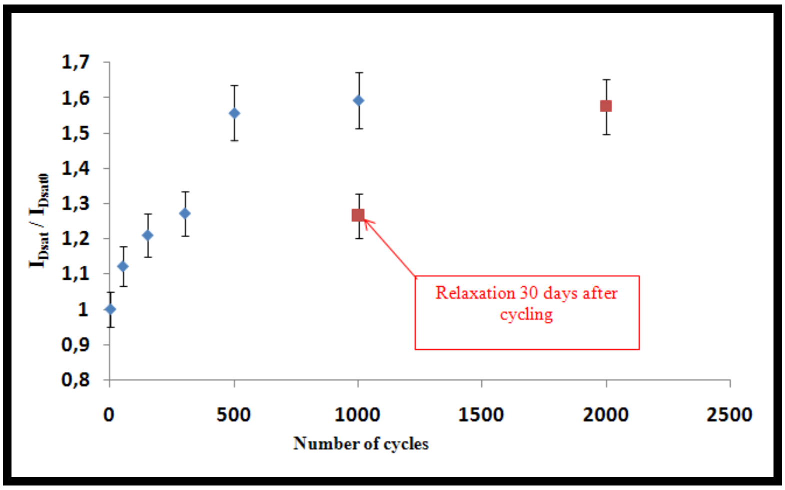

Fig.6: Normalized saturation current I_{Dsat} as a function of the number of bending cycles (R=5 mm). The data are normalized by the start of the measurement. This procedure is repeated on bending cycles of 15 times for 1 min.

Complementary to these static tests and in order to investigate the recovery performance after stressing transistors, some preliminary fatigue tests are performed using a dedicated cyclic bending machine (Fig. 5). A lot of bending cycles with bending radius of 5 mm, which corresponds to a strain of 1.25%, are applied to the OTFTs. First, the transistors sheet is bent from flat R=∞ to R=5 mm, and immediately released up to flat state at the highest frequency of our system (15 cycles per minute). Measurements are performed at 50, 100, 300, 500, and 1000 bending cycles.

The normalized saturation current as a function of the number bending cycles is shown in Fig.6. We observed an unexpected increase of the saturation current I_{Dsat} by ~10% as soon as we reached 50 cycles. I_{Dsat} continues increasing with the number of cycles. After 1000 times, the augmentation is estimated to 60%. 4 weeks after this experiment, we measured again our devices and observed a change of the electrical characteristic; indeed, saturation current has decreased from 60% to 20% (Fig. 5). That means that after stress, a slow relaxation phenomenon occurs. In spite of the lack of catastrophic degradation after a lot of cycles, we have no good stability under mechanical fatigue contrary to encapsulated organic transistors which are more stable under a lot of bending cycles [23]. But this performance remains better than amorphous-silicon field effect transistors (a-Si FETs) which fail mechanically by periodic cracks in the amorphous layers of the TFT at a strain of about 0.5% [24] [25]. More experiments are necessary with increasing the number of cycles and investigating the relaxation after fatigue to get a better understanding of this phenomenon which could be a real problem for the functionality of electronic systems.

4. Conclusion

In summary, we have studied the robustness of organic field-effect transistors (OFET's) made of amorphous triarylamine copolymer semiconductor on a PEN flexible substrate under mechanical stress. Under tensile bending tests, we have observed a reversible increase of the saturation current and mobility of about 10% for a mechanical tensile strain of 2%. This could be explained by an improvement of the relative order of the semiconductor structure. Further ways of analysis could still be investigated in order to achieve more information about the structural behaviour of the amorphous active layers. Indeed, using micro-Raman spectroscopy which is a sensitive technique of structure study, polymer organisation generated by mechanical strain can be brought out after analysing Raman spectra. Besides that, the AFM microscopy, with phase image in tapping mode, can supply the illustration of these structure variations due to mechanical strain. We have also performed preliminary cyclic bending tests and observed a 60% increase of the saturation current after 1000 cycles with a slow relaxation phenomenon. Different transistor geometries and orientations will be tested to get a better understanding of these mechanisms.

Acknowledgments

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