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# $Ta_2O_5$ as gate dielectric material for low-voltage organic thin-film transistors

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**Abstract**

In this paper we report the use of $Ta_2O_5$ as gate dielectric material for organic thin-film transistors. $Ta_2O_5$ has already attracted a lot of attention as insulating material for VLSI applications. We have deposited $Ta_2O_5$ thin-films with different thickness by means of electron-beam evaporation. Being a relatively low-temperature process, this method is particularly suitable for organic thin-film transistor fabrication on plastic substrates. Deposition and patterning are achieved in one step by the use of shadow masks. The dielectric can be evaporated on top of the semiconducting layer. In this way a large variety of structures can be realized. Poly(3-hexylthiophene) was used as semiconducting material in the transistor structure. Such transistors are operating at voltages smaller than $-3$ V. Having a high dielectric constant ($\varepsilon_r = 21$), $Ta_2O_5$ facilitates the charge carrier accumulation in the transistor channel at much lower electrical fields. The properties of the dielectric material as well as the operation of the organic transistors with a $Ta_2O_5$ gate dielectric are discussed.

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*Keywords:* $Ta_2O_5$; Poly(3-hexylthiophene); Organic transistor; Low voltage

## 1. Introduction

Organic thin-film transistors (OTFTs) have gathered great interest in the last decade since organic conjugated materials were considered as potential candidates to conventional inorganic ones in applications requiring large area, flexibility, low-temperature processing and especially low-cost. Such applications include flat-panel displays, low-end smart cards, electronic identification tags as well as sensing devices [1].

In order to be implemented as viable solutions for this kind of applications, they must offer a substantial performance advantage compared with the current technology. From the performance point of view, the most important parameters are charge carrier mobility, on/off current ratio and the operational voltage range. Up to now, the highest mobility values, superior to a:Si, have been obtained for pentacene-based OTFTs with sufficiently high on/off ratios [2]. However, the operating voltages required to produce such performances were unpractically high, around 100 V.

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The reason for that was shown to be the mobility dependence on the accumulated charge in the OTFT channel. Since this charge is proportional with both the dielectric constant and the gate voltage, it has been suggested that the use of high dielectric constant materials will allow the necessary charge to accumulate at much lower voltages [3].

In this paper, we are reporting, for the first time to our knowledge, the use of tantalum pentoxide ($Ta_2O_5$) deposited by electron-beam evaporation as dielectric material for low operating voltage OTFTs. $Ta_2O_5$ thin films have already attracted a lot of attention as insulating materials for MOS field-effect transistors and high density memory cells [4], due to their high dielectric constant ($\varepsilon_r = 20$–$35$), high refractive index and chemical and thermal stability compatible with microelectronics processing. The deposition technique of $Ta_2O_5$ thin films significantly affects the dielectric properties. A variety of techniques such as anodization, RF sputtering, thermal oxidation, chemical vapor deposition (CVD), low pressure CVD (LPCVD), photo-CVD, plasma CVD, metal-organic CVD (MOCVD) and ion assisted deposition have been used to produce $Ta_2O_5$ films [4,5]. Each technique has its merit and its drawbacks. For example, MOCVD can be used for large scale production, but an elevated growth temperature is required for cracking the metal-organic source.

The use of $Ta_2O_5$, deposited by anodic oxidation of metallic tantalum in aqueous solutions, as a high-capacitance gate dielectric in organic transistors, has been proposed by Tate et al. [6]. The anodization process always takes place at a metal electrode and this is not completely converted into an oxide. There are always a few nanometers of metal left in order to sustain the oxide on the substrate and, therefore, this method allows to realize transistors in the bottom-gate configuration only.

Here we propose the e-beam evaporation technique of a $Ta_2O_5$ target as deposition method for the thin dielectric films. Being a low-temperature process, e-beam evaporation is perfectly compatible with the use of plastic substrates. This method provides films with good electrical properties for the operation of organic thin-film transistors, excellent adhesion on plastic substrates and good thickness uniformity.

We have fabricated OTFTs, on both glass and silicon substrates. As semiconducting material we have used poly(3-hexylthiophene) regioregular (P3HT), a commercially available conducting polymer from Sigma-Aldrich. The e-beam evaporation of $Ta_2O_5$ allows to realize devices in both staggered and inverted staggered configuration, as they are known for the a:Si TFT arhitecture. In fact, the carrier mobility in the OTFT proves to be higher for the staggered configuration compared with the commonly used inverted one. P3HT thin films (100 nm) can stand very well the evaporation of the dielectric layer. Operating voltages smaller than $-3$ V have been achieved for such transistors. The fabrication and performances of these devices are described.

## 2. Experiment

$Ta_2O_5$ films have been deposited by e-beam evaporation from a $Ta_2O_5$ 99.99% target purchased from Unaxis, using the High Vacuum Laboratory System PLS 580 from Pfeiffer. Films with different thicknesses (100 and 200 nm) were grown on oxidized silicon, glass and plastic (polycarbonate and PMMA) substrates. The substrate was not heated during deposition. In all the cases, the depositions were carried out at a pressure of about $2 \times 10^{-4}$ mbar inside the chamber. This pressure is caused by the outgassing of the $Ta_2O_5$ target during the e-beam bombardment. The optimum deposition rate for the quality of $Ta_2O_5$ films was found to be 5 Å/s.

In order to analyze the dielectric properties, capacitor structures were fabricated using Al electrodes deposited onto the dielectric film through a shadow mask. The dielectric constant of the material was extracted from *C*–*V* and impedance measurements carried out with a HP4294A impedance analyzer. The results are shown in the following section.

The OTFTs have been fabricated in two different configurations, as shown in Fig. 1(a) and (b). The "inverted" staggered configuration (Fig. 1(a)), in which the gate electrode is the first de-

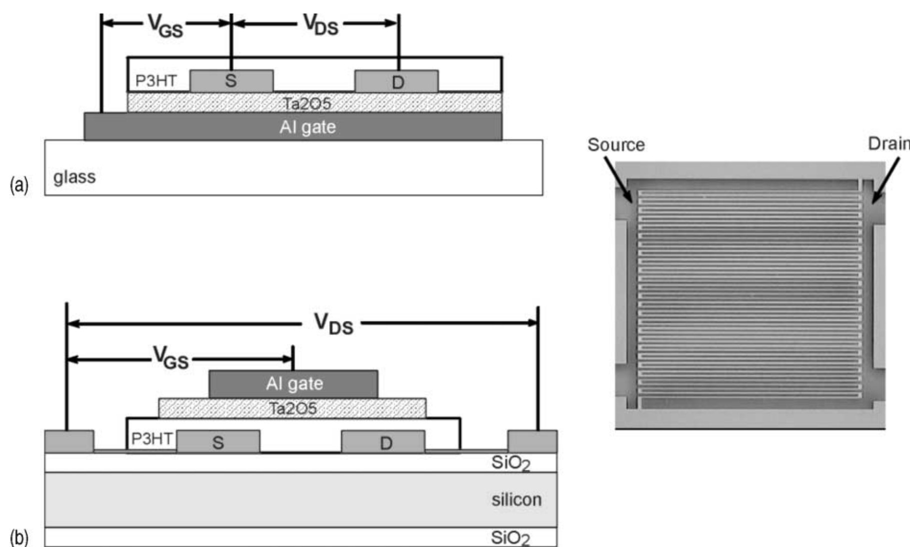



Fig. 1. (a) Inverted staggered TFT configuration; (b) staggered TFT configuration. The interdigitated source and drain electrodes are shown in the inserted photograph.

posited layer, is commonly used for organic transistors. A glass wafer was used as substrate. A non-patterned Al gate with a thickness of 300 nm was evaporated onto the substrate. The dielectric $Ta_2O_5$ film was deposited subsequently by e-beam evaporation in the conditions mentioned above. Afterwards the Au source and drain electrodes (100 nm thick) with an interdigitated configuration have been patterned by lift-off technique. In the last step, a thin layer of P3HT has been spin-coated from a chloroform solution (0.7 wt%).

The staggered configuration, illustrated in Fig. 1(b), proves the perfect compatibility of the dielectric deposition method with plastic materials. In a first step, the source and drain electrodes have been patterned onto an oxidized silicon substrate and subsequently coated with the P3HT layer. In the next step, the $Ta_2O_5$ film was e-beam evaporated through a shadow mask onto the polymer layer. Finally, an Al gate electrode with a thickness of 300 nm was also evaporated via a shadow mask on the dielectric.

The electrical characterization of the OTFTs was carried out with a HP4156A Precision Semiconductor Parameter Analyzer and the results are discussed in the following section.

## 3. Results and discussion

### 3.1. Dielectric properties of evaporated $Ta_2O_5$

As deposited $Ta_2O_5$ films are amorphous and show good electrical properties for the operation of OTFTs. We have extracted the dielectric constant from the frequency measurements performed on $Al/Ta_2O_5/Al$ structures. The area of the capacitor is 44 900 $\mu m^2$. The results obtained for capacitor structures with 100 and 200 nm $Ta_2O_5$ films are displayed in Fig. 2. From the results illustrated in Fig. 2(b), we have calculated the relative dielectric constant of the evaporated films as being $\varepsilon_r = 21$. This value is in agreement with the reported values for the dielectric constant of $Ta_2O_5$ deposited by different techniques [4,5].

The breakdown strength of the evaporated films was higher than 1 MV/cm, similar to the data reported in literature [4,5]. Corresponding to this dielectric strength, a maximum surface charge density of about 2 $\mu C/cm^2$ can be induced in the organic semiconducting layer, sufficient for organic TFT applications.

As a first indication of the material quality, we have also measured the dielectric loss angle, $\theta$, as being around $-85°$. This can be caused either by

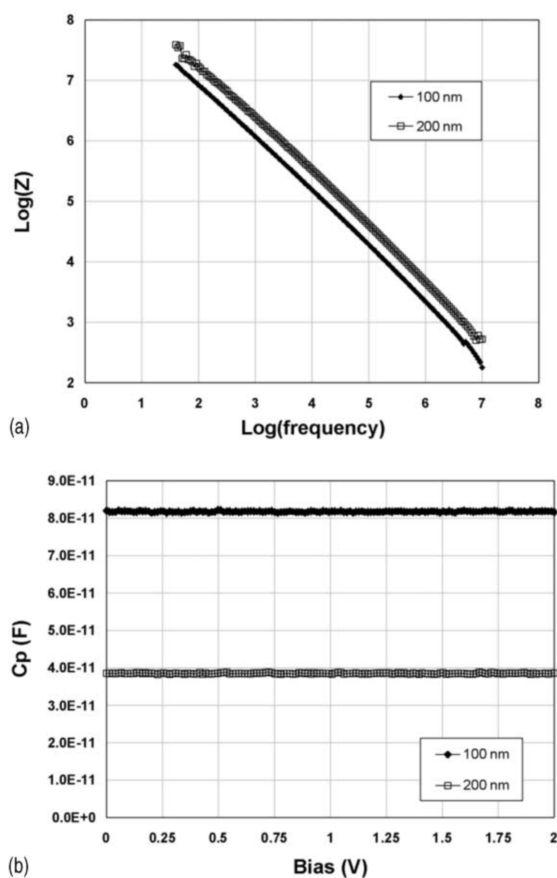



Fig. 2. Dielectric properties recorded on capacitors with 100 and 200 nm $Ta_2O_5$ dielectric: (a) logarithmic plot of impedance versus frequency; (b) *C–V* plots.

the defects present in the film as well as by the use of Al as electrode material. As discussed in the literature, the electrical characteristics of $Ta_2O_5$ films strongly depend on the top electrode material [7]. It seems that aluminum is partly oxidized by the excess oxygen generated during the deposition step and the accumulated space charge in this oxide layer is changing the electric field distribution. Nevertheless, the quality of the dielectric was reasonably good for the applications described in this paper.

### 3.2. OTFTs characterization

All the organic transistors were characterized in ambient atmosphere using a HP4156A Precision Semiconductor Parameter Analyzer. Fig. 3 illustrates the plot of the drain current, $I_{DS}$, versus drain voltage, $V_{DS}$, at different gate voltages, $V_G$, for the OTFT in the commonly used inverted staggered configuration. Fig. 4 depicts the same output characteristics recorded for the transistors fabricated in the staggered configuration. The geometry of the source and drain electrodes is identical in the two transistor configurations. The drain current is flowing between $N = 49$ parallel channels with the length $L = 10$ μm and the width $W = 1$ mm. As shown in Figs. 3 and 4, both kinds of transistors are operating in accumulation mode at voltages smaller than $-3$ V.

The leakage current density through the $Ta_2O_5$ dielectric, measured at a gate field of 0.5 MV/cm, was $10^{-8}$ A/cm$^2$ for the OTFT from Fig. 1(a) and $10^{-7}$ A/cm$^2$ for the OTFT from Fig. 1(b).

Nevertheless, for the configuration from Fig. 1(b), the drain current is higher and it has an increasing trend at high $V_{DS}$ voltages. This can be explained if one analyzes the current flowing in the device in the absence of the gate bias, $V_G = 0$ V. Obviously this is higher compared with the current shown in Fig. 3. We believe this to be due mainly to the increase of the doping level of the P3HT during the $Ta_2O_5$ deposition. During the evaporation of $Ta_2O_5$ onto the P3HT film, the oxygen content inside the chamber is increasing due to outgassing from the $Ta_2O_5$ target and it is expected that oxygen may diffuse in the P3HT layer and act as an acceptor. This is supported by the measured conductivity of the film. Prior $Ta_2O_5$ deposition, the conductivity of the film was $\sigma = 2 \times 10^{-5}$ S/cm, while after the evaporation it became $\sigma = 2 \times 10^{-4}$ S/cm. The increase in the bulk conductivity of P3HT is further reflected in the higher mobility calculated for this transistor compared to the mobility extracted from the OTFT in inverted configuration.

The field-effect mobility dependence on the gate bias, $V_G$, was extracted from the transconductance, according to [8]. The transconductance is easily calculated from $I_{DS}$–vs–$V_G$ characteristics recorded for small $V_{DS}$ (in the linear regime),

$$g_m = \left(\frac{\partial I_{DS}}{\partial V_G}\right)_{V_{DS}\to 0} = N\frac{W}{L}\mu C_{ins} V_{DS}. \quad (1)$$

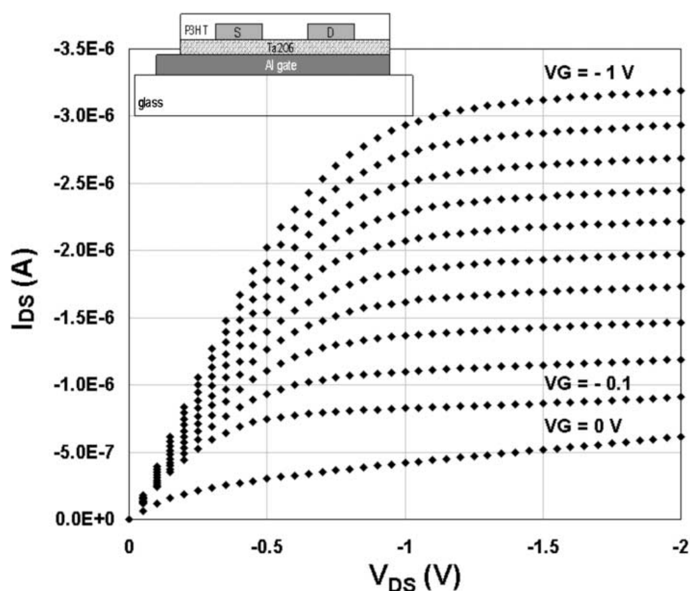



Fig. 3. Output characteristics recorded for an OTFT in inverted staggered configuration: 100 nm $Ta_2O_5$ and $N = 49$ parallel channels (channel length $L = 10$ μm and the channel width $W = 1$ mm). $V_G$ is swept from 0 to $-1$ V in steps of $-0.1$ V.

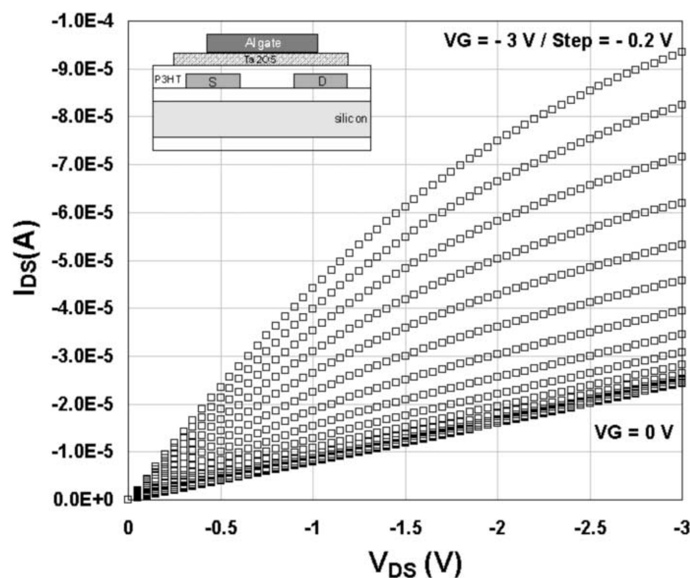



Fig. 4. Output characteristics recorded for an OTFT in the normal staggered configuration: 100 nm $Ta_2O_5$ and $N = 49$ parallel channels (channel length $L = 10$ μm and the channel width $W = 1$ mm). $V_G$ is swept from 0 to $-3$ V in steps of $-0.2$ V.

Then the mobility of the accumulated charge as a function of the gate bias may be calculated,

$$\mu(V_G) = \left(\frac{L}{NWC_{\text{ins}}V_{\text{DS}}}\right)\left(\frac{\partial I_{\text{DS}}}{\partial V_G}\right)_{V_{\text{DS}}\to 0}. \qquad (2)$$

Figs. 5 and 6 show the transfer characteristics, $I_{DS}$–vs–$V_G$ recorded for the two transistor configurations, at small $V_{DS}$, $V_{DS} = -0.1$ V, on the left axis and the mobility, calculated according to Eq. (2), on the right axis.

Comparing the two plots from Figs. 5 and 6, one can see that when the $Ta_2O_5$ dielectric was evaporated onto the P3HT semiconductor, the mobility is also one order of magnitude higher, and tends to saturate towards $V_G = -3$ V, supporting the hypothesis of oxygen doping during dielectric deposition. The density of states at the Fermi level increases with the acceptor density. Given that charge transport in polymer systems such as P3HT is indeed dominated by variable range hopping at the Fermi level, it can be expected that both the

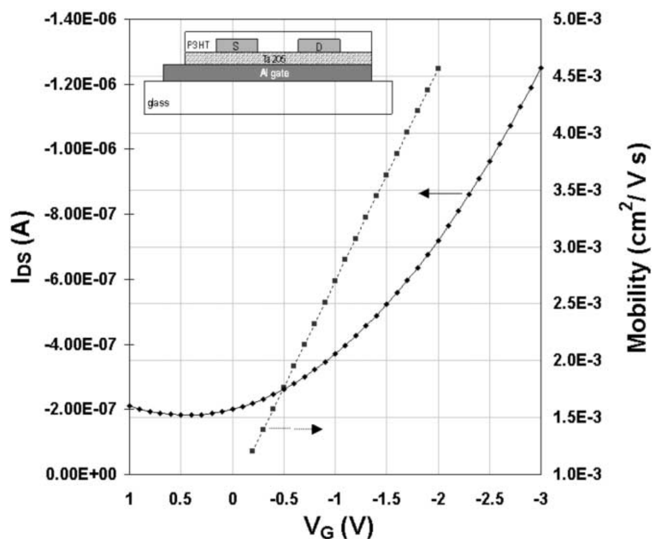



Fig. 5. Plots of $I_{DS}$–vs–$V_G$ recorded for $V_{DS} = -0.1$ V (left axis) and field-effect mobility (right axis) for the OTFT in inverted staggered configuration ($C_{ins} = 1.8 \times 10^{-7}$ F/cm$^2$).

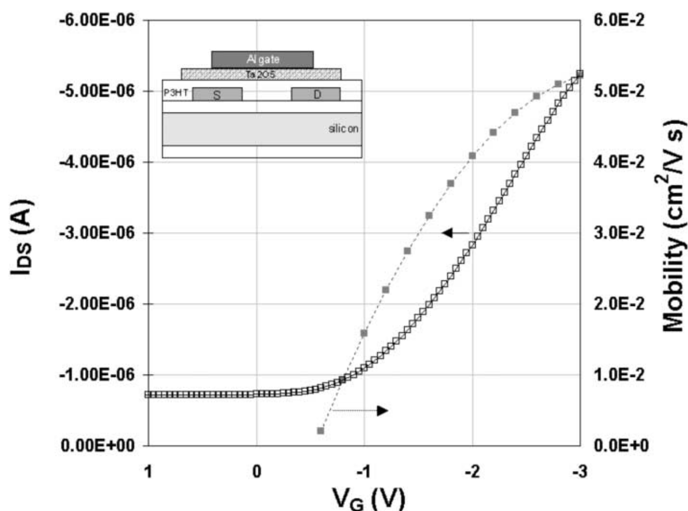


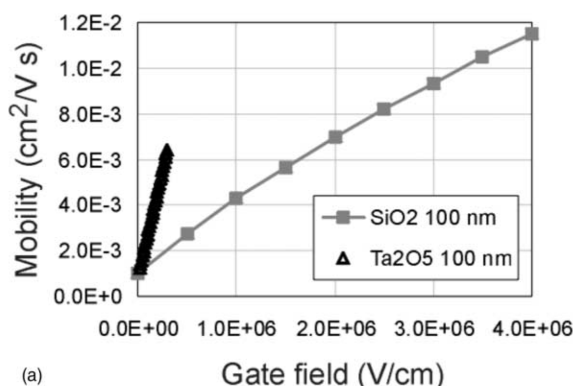

Fig. 6. Plots of $I_{DS}$–vs–$V_G$ recorded for $V_{DS} = -0.1$ V (left axis) and field-effect mobility (right axis) for the OTFT in normal staggered configuration ($C_{ins} = 1.8 \times 10^{-7}$ F/cm$^2$).

conductivity and the field-effect mobility should increase upon doping. The field-effect mobility dependence on the conductivity has already been reported for a wide range of organic systems [9].

The mobility values obtained for the P3HT semiconductor, spin-coated from chloroform solutions, in air, without additional treatments, are in agreement with the reported data for this material [10]. In order to characterize the organic semiconductor, we have also fabricated transistor structures with $SiO_2$ as dielectric material, having a bottom-gate configuration (see Fig. 1(a)). The charge carrier mobility measured on such devices was about $10^{-2}$ $cm^2/V\,s$, the same as for the $Ta_2O_5$/P3HT transistors with the same configuration. Fig. 7 displays the mobility dependence on the gate field as well as on the induced charge per unit area, $Q_S$, for the $SiO_2$/P3HT and $Ta_2O_5$/P3HT transistors, with a dielectric thickness of 100 nm and a channel length, $L = 10$ μm. As one can see, the mobility is increasing quasi-linearly with both the gate field and the accumulated charge density and its dependence on the charge density is the same for both dielectric materials. The difference consists in the fact that the same charge density is achieved in the case of $Ta_2O_5$ at much lower voltages.

We have also calculated the mobility from the slope of the plot $|I_{DS}|^{1/2}$–vs–$V_G$, in the saturation regime. For the OTFTs in the inverted staggered configuration, the calculated mobility was 0.004 $cm^2/V\,s$ and the threshold voltage $V_T$ was $-77$ mV, while for the transistors in staggered configuration the mobility was 0.02 $cm^2/V\,s$ and $V_T$ was $+260$ mV. The positive $V_T$ correlated with a higher bulk conductivity obtained for the second transistor structure is in agreement with the results obtained by Horowitz et al. [11] for dihexyl-sexithiophene (DH6T) semiconductor, doped by the oxygen from the ambient air. According to these authors, the positive $V_T$ is not a threshold voltage, but a zero voltage connected to the equilibrium free carrier density.

Since the on/off current ratio is proportional to the ratio between the mobility and the bulk conductivity of the film, it may appear that the $Ta_2O_5$ evaporation onto P3HT films leads to poor on/off current ratios in transistors with a top-gate configuration. Nevertheless, rather poor on/off current ratios are generally obtained for P3HT-based transistors if no additional treatments are used to reduce the bulk conductivity [10]. The reason for that is the high dopant concentration induced unintentionally in the polymer by its synthetic process. Additional tests have to be performed in order to verify the P3HT doping hypothesis.



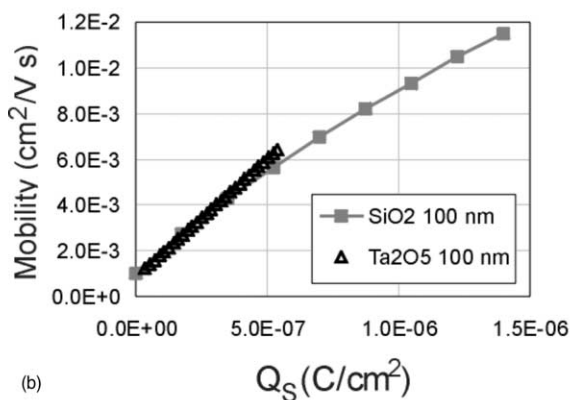




Fig. 7. Mobility dependence on (a) the gate field; (b) induced charge per unit area, $Q_S$, in P3HT/$SiO_2$ and P3HT/$Ta_2O_5$ OFETs with the same dielectric thickness (100 nm) and same electrodes geometry ($L = 10$ μm).

## 4. Conclusions

We have fabricated OTFTs with e-beam evaporated $Ta_2O_5$ as dielectric material. Having a low-thermal budget, e-beam evaporation of high-$k$ dielectrics is perfectly suitable to organic device

fabrication. It allows to realize the OTFTs in both staggered and inverted staggered configurations, depending on the specific application. The operation voltages are reduced to less than −3 V for both configurations. The carrier mobility in the P3HT film was 0.02 $cm^2/V\,s$ for the transistor in staggered configuration and 0.004 $cm^2/V\,s$ for the inverted structure. However, problems may be encountered when the $Ta_2O_5$ film is evaporated on top of organic semiconductors that are susceptible to oxygen doping. Due to the doping, the conductivity and the mobility are increasing, while the on/off ratio is decreasing. A systematic study concerning the influence of the $Ta_2O_5$ evaporation conditions on the electronic properties of the organic semiconductor is currently carried out.

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## References

[1] C.D. Dimitrakopoulos, D.J. Mascaro, IBM J. Res. Dev. 45 (1) (2001) 11.
[2] S. Nelson, Y.Y. Lin, D.J. Gundlach, T.N. Jackson, Appl. Phys. Lett. 72 (1998) 1854.
[3] C.D. Dimitrakopoulos, S. Purushothaman, J. Kymissis, A. Callegari, J.M. Shaw, Science 283 (1999) 822.
[4] E. Ezhilvalavan, T.Y. Tseng, J. Mater. Science: Mater. Electron. 10 (1999) 9.
[5] C. Chaneliere, J.L. Autran, R.A.B. Devine, B. Balland, Mater. Sci. Eng. R22 (1998) 269.
[6] J. Tate, J.A. Rogers, C.D.W. Jones, B. Vyas, D.W. Murphy, W.J. Li, Z.A. Bao, R.E. Slusher, A. Dodabalapur, H.E. Katz, Langmuir 16 (2000) 6054.
[7] H. Matsuhashi, S. Nishikawa, Jpn. J. Appl. Phys. 33 (1994) 1293.
[8] A.R. Brown, C.P. Jarrett, D.M. de Leeuw, M. Matters, Synth. Met. 88 (1997) 37.
[9] A.R. Brown, D.M. de Leeuw, E.E. Havinga, A. Pomp, Synth. Met. 68 (1994) 65.
[10] Z. Bao, A. Dodabalapur, A.J. Lovinger, Appl. Phys. Lett. 69 (1996) 4108.
[11] G. Horowitz, R. Hajlaoui, H. Bouchriha, R. Bourguiga, M. Hajlaoui, Adv. Mat. 10 (1998) 923.