Hybrid gate dielectrics: a comparative study between polyvinyl alcohol/SiO₂ nanocomposite and pure polyvinyl alcohol thin-film transistors

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Abstract. Polyvinyl alcohol (PVA) thin films as polymer gate dielectrics, with and without SiO₂ nanoparticles were fabricated using spin-coating. Surface roughness and hydrophilicity of PVA and PVA/SiO₂ thin films were studied by contact-angle measurements and atomic force microscopy. The dielectric properties were characterized *via* capacitance and leakage-current measurements on metal–insulator–metal structures. In order to further investigate the application potential of such materials as a replacement for conventional inorganic dielectrics such as SiO₂ in organic thin-film transistors, devices were fabricated based on these polymers using α , ω -dihexylquaterthiophene as an active layer. Performance of the devices was realized by electrical measurements and Kelvin probe force microscopy. All transistors showed hole and electron mobilities in the low-voltage range. PVA/SiO₂ films showed larger capacitance, less hydrophilicity, rougher surfaces and considerable leakage currents compared with those with neat PVA. Although integrating nanoparticles modified surface electronic properties and showed a shift in surface potential as observed in Kelvin probe force measurements, it appears that non-polymeric and neat polymeric dielectric materials could still be a privilege to nanocomposite polymeric dielectrics for optoelectronic applications.

Keywords. Polymer dielectrics; surface chemistry; electrical and structural properties; scanning probe microscopy (SPM); ambipolar thin-film transistor.

1. Introduction

Organic thin-film transistors (OTFTs) have been studied over the last few decades, due to their potential applications in low-cost, easily processable, flexible and large area electronic devices [1–3]. Since some organic materials such as alkyl-substituted oligothiophenes can easily be solution processed, solution-based techniques including spin-coating, ink-jet and screen printing, have been employed in order to expand the applications of OTFTs in a diverse range from flexible displays and radio-frequency identification (RFID) tags to various sensors [4,5]. Organic semiconductors are generally used in unipolar devices and typically exhibit p-type behaviour [6–9]. However, it has been shown that electrons can also be mobile in many conjugated polymers if an appropriate gate dielectric, in most cases polymers, is chosen leading to the n-type behaviour [10]. OTFTs which can operate in both p-type and n-type conduction modes (ambipolar behaviour) are ideal candidates for the simple and low-cost development of complementary like circuits [11]. Polymer dielectrics have recently been used as gate dielectrics in OTFTs due to their simple processing *via* spin-coating or -casting, and easy tuning of surface chemical properties [12].

It is shown that they can lead to better performances in, for instance, pentacene-based OTFTs compared with those fabricated on SiO₂ dielectrics [13,14] and also lead to ambipolar characteristics [15–18].

In order to lower the operating voltage of OTFTs, ceramic-based inorganic dielectrics with a high-dielectric constant (high-*k*) can be considered. Nevertheless, ceramics are usually brittle and costly to prepare. These materials suffer from poor mechanical properties which make them inconvenient to be used in flexible electronics. Therefore, it is important to develop a cheap and easy way, like a solution-based method, to fabricate gate dielectrics with both high-*k* and mechanical flexibility. OTFTs using an organic dielectric with a high-dielectric constant seem to be promising for operation at low voltages. However, neat polymer materials have relatively low-dielectric constant which limits their use in low-voltage applications. In order to overcome this problem, different nanocomposite dielectrics, containing inorganic nanoparticles with a high-*k*, have been studied [18–22]. Nevertheless, the leakage problem and rather low mobility values of such devices compared with their inorganic counterparts raise the question of their commercialization and is still under debate.

In this work, in order to study the effectiveness and reliability of hybrid polymeric materials as gate dielectrics in OTFTs, we present ambipolar organic transistors based on α,ω -dihexylquaterthiophene (DH4T) with neat and hybrid polymer dielectrics deposited on glass substrates coated with indium tin oxide (ITO). SiO_2 nanoparticles embedded in a polyvinyl alcohol (PVA) polymer matrix were used to form nanocomposite (hybrid) gate dielectrics. The first type of devices used neat PVA gate dielectric and the second type used PVA/ SiO_2 nanocomposite gate dielectric. PVA was adopted as a solution processable organic insulating polymer since it is a non-toxic, low-cost and commercially available polymer which has two important properties: it is soluble in water (and insoluble in organic solvents) and it already has a relatively high-dielectric constant ($k > 5$), provided the film thickness lies in the range of hundred nanometres to several micrometres [23,24]. PVA and its copolymers can also be chemically and thermally cross-linked which may allow the stabilization of the layer. The surface roughness and hydrophilicity of the PVA layers with and without SiO_2 nanoparticles were first studied by means of contact-angle measurement and atomic force microscopy (AFM). The dielectric properties were characterized *via* capacitance and leakage-current measurements within metal–insulator–metal (MIM) structures. Then the performances of the OTFTs were investigated by electrical measurements and Kelvin probe force microscopy (KPFM). It is shown that the control of the surface chemistry of the gate dielectric allows us to tune the behaviour of OTFTs. In spite of the reduced electronic properties, to the best of our knowledge, ambipolar behaviour for DH4T-based TFTs has been reported for the first time.

2. Materials and methods

2.1 Materials

The organic component, PVA (99% hydrolysed grade), average M_w 89,000–98,000, used in this study was obtained from Sigma-Aldrich. The inorganic component, LUDOX R SM colloidal silica, also supplied by Sigma-Aldrich, is an aqueous dispersion containing 30 wt% of silica nanoparticles (average particle size of 7 nm) at a pH of 9.1. Both PVA and colloidal silica were used as received from the suppliers without further purification. DH4T was synthesized as described elsewhere [25].

2.2 Preparation of PVA and PVA/ SiO_2 nanoparticle solutions

A 30 g l⁻¹ homogeneous solution of PVA was first prepared by dissolving PVA powder in boiling water and then continuously stirring for 1 h. For preparing PVA/ SiO_2 nanoparticle solutions (5 wt%), a requisite amount of aqueous dispersion of colloidal silica was added to the PVA solution and

continuously stirred for 24 h at room temperature to obtain a perfectly homogeneous solution.

2.3 OTFT fabrication

Transistors were fabricated on ITO substrates (thickness: 100 nm). The ITO substrates were washed for 10 min in acetone and isopropyl alcohol and then rinsed with distilled water before being dried using nitrogen. After patterning the ITO substrates in order to deposit the PVA or PVA/ SiO_2 gate dielectric layers, neat polymer or polymer/nanoparticle solutions were spin-coated on the substrates (2500 rpm, 1000 rpm s⁻¹ and 60 s). The obtained films with a thickness of about 165 nm were then cured at 150°C in air for 1 h and at 80°C in a vacuum oven for 12 h. The OTFT devices were designed as a bottom gate (ITO) and bottom-contact electrode structure. The gold source and drain electrodes were deposited through a shadow mask with a channel length (L) of 40 or 100 μm and a width (W) of 7 mm. A 2-nm-thick Cr layer was deposited prior to gold as an adhesion promoter.

Finally, DH4T was deposited by spin-coating (5000 rpm, 1000 rpm s⁻¹, 15 s) from a filtered solution in toluene (0.4 wt%). Since PVA is hygroscopic, the samples were stored inside desiccators filled with anhydrous silica gel before characterization.

2.4 Characterization methods

The transmittance of the PVA and PVA/ SiO_2 solutions was quantified with a UV–visible spectrophotometer (VARIAN) in the wavelength range from 200 to 800 nm at room temperature. The surface energy of the polymeric gate dielectrics was evaluated from the water contact angle measured using a Data-physics OCA 20 contact angle meter. The surface morphology was analysed by AFM (Agilent 5500 SPM microscope). KPFM measurements were also realized with the same instrument (for more details see [26]). The current–voltage and capacitance–voltage measurements were implemented under ambient conditions using a single instrument, a low signal probe station with a probe shield (PM8PS, 4 SMU's with 10 fA resolution, two SMUs with 0.1 fA resolution and C–V measurements from 1 kHz to 5 MHz with 0.1 fF resolution).

3. Results

The solubility of PVA in water led to clear solutions of polymer and a good dispersion of the SiO_2 nanoparticles in the solution (inset of figure 1a). Although PVA could inhibit the aggregation of the nanoparticles and keep them well dispersed, some agglomeration was observed after a long time. To avoid these agglomerates of nanoparticles, the supernatant of the solution was used to make the dielectric layers. To verify the presence of SiO_2 nanoparticles in the supernatant, a UV–visible spectroscopic study was

Figure 1. (a) UV–visible spectroscopic results of PVA and PVA/SiO₂ (5 wt%) solutions. The inset shows a clear solution of PVA in distilled water and chemical structure of PVA. AFM images of the (b) PVA and (c) PVA/SiO₂ (5 wt%) composite films deposited on ITO (vertical colour scale: (b) 1 nm, (c) 4.8 nm). The insets show the contact angles on both films.

performed. Figure 1a shows the transmittance of PVA/distilled water solutions with and without colloidal SiO₂ nanoparticles. The transmittance gently decreases upon adding nanoparticles due to the variation of the refractive index of the solution. However, all solutions show reasonably high transparency with more than 80% of the visible light transmitted.

The surface characteristics of the gate dielectric may strongly affect the electrical properties of OTFTs. To study the surface properties of PVA and PVA/SiO₂ composite films, AFM images were acquired. The images in figure 1b and c

show that the PVA/SiO₂ nanocomposite film has a rougher surface with a root-mean-square (RMS) roughness value of 3.8 nm, whereas the surface of the PVA film is smoother and has a RMS roughness value of 0.8 nm. Water contact angle measurements on PVA and PVA/SiO₂ nanocomposite films gave values of $\approx 39^\circ$ and $\approx 52^\circ$ for PVA and PVA/SiO₂, respectively. The surface energy of each dielectric (γ_p) was evaluated using the following equation:

$$\gamma_p = \frac{\gamma_w}{4} (1 + \cos \theta_0)^2 \quad (1)$$

Figure 2. (a) Leakage current density and (b) capacitance measured at 800 kHz of neat PVA and PVA/SiO₂ (5 wt.%) composite dielectric layers.

where γ_w is the surface energy of water (73.0 mJ m⁻²) and θ_0 is the measured contact angle at equilibrium [27]. Based on the contact angle results, the surface energy for PVA and PVA/SiO₂ nanocomposite films is equal to 29.4 and 12.8 mJ m⁻², respectively. Long-term functionality of OTFTs under ambient conditions where the humidity and water molecules are present is a key issue that should be taken into account. The adsorption of water molecules by the surface of a device can progressively deteriorate its performance at the end. Therefore, surface with lower adsorption affinity towards water molecules (hydrophobic property) is preferred from technical and practical points of view. The lower surface energy calculated for PVA/SiO₂ composite films compared with the neat PVA films indicates that the incorporation of silica nanoparticles into the PVA polymer matrix has modified the surface chemistry of the dielectric layer in a fashion that it showed lower tendency to adsorb water molecules and became less hydrophilic. One of the main concerns with any gate dielectric material is the gate leakage, especially for polymer gate dielectrics [28]. To investigate this effect, current density–voltage (J – V) characteristics of neat PVA and PVA/SiO₂ gate dielectrics were measured on MIM structures (ITO-PVA-Au or ITO-PVA/SiO₂-Au). Figure 2a shows the leakage current density of the neat PVA and PVA/SiO₂ gate dielectric layers as a function of applied bias. The leakage current at ± 1 V is about 10⁻⁵ A cm⁻² for neat PVA and 10⁻³ A cm⁻² for the PVA/SiO₂ nanocomposite system. The capacitance of PVA/SiO₂ nanocomposite dielectric layers increases compared with that of neat PVA layers (figure 2b). Capacitance values of PVA and PVA/SiO₂ measured at 800 kHz are equal to about 0.2 and 4.9 nF cm⁻², respectively.

Conventional OTFTs need more than 15 V to be fully operational which is incompatible with portable, battery powered applications [29]. Considering this, electrical measurements in our experiments were mainly focused on low range gate and drain voltages. Figure 3 presents the transfer

and output characteristics of DH4T-based OTFTs with PVA and PVA/SiO₂ nanocomposite dielectric layers deposited on patterned ITO-gate glass substrates and with Au as source and drain electrodes.

Figure 3a and b reveals that the OTFTs based on neat PVA and on PVA/SiO₂ present ambipolar behaviours. Indeed, when they are operated at the negative gate bias, p-conduction is observed while applying positive gate voltages lead to n-type conduction. Figure 3a and b also shows that the output drain current for the PVA/SiO₂-based device is higher than that of the PVA-based device. As shown in figure 3c and d, the on-state drain current of the PVA/SiO₂-based device is also higher than that of the PVA-based device.

Considering figure 3c and d, the output drain current of the PVA/SiO₂ device is higher than that of the neat PVA device. As shown in figure 3a and b, the on-state drain current of the PVA/SiO₂ device is also higher than that of the PVA device. The on–off ratio in our transistors can be written as $r = I_{\text{on}}/I_{\text{off}}$ according to figure 3a and b. The subscripts ‘on’ and ‘off’ denote the values of the drain current at gate voltages of -5 and -4 V for the p-type channel, and gate voltages of 0 and -4 V for the n-type channel, respectively, when the drain voltage is equal to -4 V. As an example, the value of r for the p-type and n-type channels in the PVA system for the drain voltage of -4 V can be calculated to be 30 and 200, respectively (pink curve in figure 3a). This value will be 200 and 2000 for the p-type and n-type channels in the PVA/SiO₂ device, respectively, for the same drain voltage, i.e., -4 V (pink curve in figure 3b). The higher drain current of the PVA/SiO₂ device in the output and transfer characteristics can be attributed to the higher dielectric capacitance of the nanocomposite gate dielectric layers as shown in figure 2a. The increase of the capacitance of the PVA/SiO₂ system can be related to the presence of SiO₂ nanoparticles. A higher dielectric capacitance can contribute to accumulating more charge carriers in the conducting channel. The values of the as-defined on–off ratio are presented in table 1. In both

Figure 3. Output characteristics of the transistors with (a) PVA and (b) PVA/SiO₂ nanocomposite dielectric layers for various gate voltages. Transfer characteristics of the DH4T-based TFTs with (c) PVA and (d) PVA/SiO₂ nanocomposite dielectric layers for various drain voltages. The inset in c shows schematic of the fabricated DH4T-based TFTs.

Table 1. On-off ratio, $r = I_{\text{on}}/I_{\text{off}}$ and field-effect mobility, μ , of the PVA-based and the PVA/SiO₂-based devices for p-conduction and n-conduction calculated on the basis of the data presented in figure 3c and d.

Dielectric	r		μ ($10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)	
	p-Channel	n-Channel	p-Channel	n-Channel
PVA	30	200	0.18 ± 0.02	0.22 ± 0.01
PVA/SiO ₂	200	2000	0.19 ± 0.02	0.32 ± 0.02

conduction modes, the OTFTs with the PVA/SiO₂ composite dielectric present a larger on-off ratio.

The field-effect mobilities of both types of devices were calculated from the characteristic curves using the following

expression:

$$\mu = \frac{dI_{\text{DS}}}{dV_{\text{GS}}} \frac{L}{W} \frac{d}{\varepsilon} \frac{1}{V_{\text{DS}}} \quad (2)$$

where L and W are the channel length and width respectively, d is the polymer dielectric film thickness, ε its dielectric constant, $dI_{\text{DS}}/dV_{\text{GS}}$ is the transconductance calculated from the linear region of the $I_{\text{DS}}-V_{\text{GS}}$ curves at the left side (p-channel) and right side (n-channel) of the minimum current value in figure 3c and d. The mobility values were calculated and averaged on 10 devices and are given in table 1 for a drain voltage of -2 V . They all are in the range of $10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for both PVA- and PVA/SiO₂-based transistors. The mobilities are slightly larger for the PVA/SiO₂-based devices. However, it should be noted that the current flow through the PVA/SiO₂

Figure 4. (a) Topographic and (b) potential images obtained on a DH4T-based OTFT with a PVA dielectric. (c) Topographic and (d) potential images obtained on a DH4T-based OTFTs with a PVA/SiO₂ nanocomposite dielectric. Potential images were acquired at $V_G = V_D = 0$ V. (e) Potential profiles acquired along the horizontal white lines shown in **b** and **d**; the dashed area locates the channel.

layer, due to the leakage paths shown in figure 2a, would certainly influence the performance of our devices and the calculated mobility values are a contribution of both current flow in dielectric layer and the semiconductor.

Figure 4 presents topography images and typical potential images obtained for 40 μm wide-channel transistors with DH4T spin-coated on neat PVA (figure 4a and b) and on

PVA/SiO₂ nanocomposite dielectric (figure 4c and d). The surface potential profiles in figure 4e were extracted from the surface potential images along the white line indicated in figure 4b and d at zero gate and drain voltages.

On the basis of the topography images (figure 4a and c) and the phase images (not shown here), in both cases, DH4T crystals are formed on top of a thin and continuous DH4T film

covering the whole channel. The thin crystals are observed both on the channel and on the gold electrodes. However, it seems that the number of thin crystals is higher in the device with PVA/SiO₂ compared with the device with neat PVA.

DH4T films deposited by spin-coating have island morphology. Spin-coated crystals which are formed on top of a very thin, continuous layer are more suited for performing microscopic measurements like KPFM; although the presence of the cracks and pores together with the smaller grain size can be the reason behind the modest macroscopic electrical performance of spin-coated DH4T films. The surface potential images (figure 4b and d) and profiles (figure 4e) reveal a uniform potential distribution in the channel for both PVA- and PVA/SiO₂-based systems. However, the surface potential measured on the PVA/SiO₂-based device is shifted towards positive values compared with the potential measured on the PVA-based device. For the PVA-based OTFT, the surface potential is slightly negative in the channel (black curve). On the other hand, the surface potential in the channel of the PVA/SiO₂-based OTFT is clearly positive (red curve).

4. Discussion

Regarding the leakage current (figure 2a), the larger value measured on the PVA/SiO₂ systems is probably due to the structural defects induced by the presence of the SiO₂ nanoparticles [19,30]. This hypothesis is supported by the fact that the surface roughness of the dielectrics increases when SiO₂ nanoparticles are embedded in the gate polymer film (figure 1b and c). The increase in the leakage current which appeared to be unavoidable in our devices, affected the performance of transistors in terms of mobility enhancement. It seems that integrating nanoparticles into the polymer matrix could not improve the transfer properties compared with the neat PVA system, though some parameters such as source–drain current values and the on–off ratio were increased. The higher drain current of PVA/SiO₂-based devices in the output characteristics and higher on–off ratio in the transfer characteristics may be attributed to the higher capacitance of the nanocomposite gate dielectric layers as shown in figure 2b. Apparently, the presence of the SiO₂ nanoparticles ($k \approx 5.2$ [31]) improved the dielectric characteristics of the PVA layer. A higher capacitance can contribute to a larger accumulation of charge carriers in the conducting channel.

Ambipolar behaviours are observed for both systems. This observation is in contradiction with previous reports which showed that DH4T-based OTFTs present p-type behaviours in both organic [32] and inorganic [33] dielectric materials. However, it has already been reported that organic semiconductors can be ambipolar on hydroxyl-terminated dielectrics though surface dipoles make n-channel behaviour less likely [10,16,23,34]. So far, there is less information about the correlation of ambipolar transport with the morphology and surface properties of the organic gate dielectric layer and the

Figure 5. Schematic representation of the interface between the channel and (a) PVA or (b) PVA/SiO₂ dielectric gate films leading to different charge accumulation. Free charges (holes or electrons) are in red; trapped electrons are in green-yellow and silica nanoparticles are represented by pink discs.

structure of the organic semiconducting films at the interface [35,36]. The functional groups present on the surface of the polymer gate dielectrics can be an important parameter to determine the type of accumulated charge carriers and the performances of the OTFTs. Embedding SiO₂ nanoparticles may strongly affect the surface functionality of the PVA gate film. This is indeed what is observed. The decrease of surface hydrophilicity of PVA/SiO₂ nanocomposite films compared with neat PVA films suggests that the hydroxyl groups, generally appearing on the surface of neat PVA, are mainly suppressed due to the presence of nanoparticles inside the polymer. Indeed, one may expect that hydrogen bonds are formed between the PVA hydroxyl groups and the silanol groups present on the surface of the silica nanoparticles as already mentioned in the literature [37,38].

On that basis, the improved characteristics observed for the PVA/SiO₂ thin films may in fact be due to a balance between the increase of the dielectric constant of the gate dielectric film and the chemical and/or morphological modifications of its surface. In figure 5, a schematic model is proposed to visualize the possible surface modifications due to the embedding of nanoparticles and their effects on the charge carriers.

On bare SiO₂ substrates, organic semiconductors show p-type behaviour due to strong trapping of electrons. Indeed in the absence of special surface modification, SiO₂ exhibits a large interface trap density of $\approx 10^{12}$ – 10^{13} cm⁻². This large interface trap density is mainly linked to the silanol (Si–OH) species assisted by adsorbed H₂O [39,40]. These species preferentially trap electrons resulting in the suppression of the n-channel conduction. The proposed electron trapping mechanism on SiO₂ involves the release of protons by the silanol moieties and the capture of electrons by the protons to release H₂ [41]. This phenomenon may explain why DH4T only shows p-type conduction on SiO₂. The same mechanism may occur at the surface of PVA with the hydroxyl moieties (figure 5a). This idea can be further supported by the surface potential profile obtained on the PVA system which shows that the surface is slightly negatively charged due to the accumulation of electrons at the interface (figure 4e). However, alcohols and phenols (C–OH) are less effective proton donors ($pK_a \approx 10$ – 15) than silanols ($pK_a \approx 5$). The electron trap density on PVA may thus be smaller than that on SiO₂ allowing the formation of an n-channel and leading to the observation of an ambipolar behaviour on PVA.

In the case of the PVA/SiO₂ system, the assumed formation of hydrogen bonds between the PVA hydroxyl groups and the silanols on the SiO₂ nanoparticles reduces the number of available OH groups at the interface between the grid dielectric and the active layer (DH4T layer). There should thus be an even lower electron trap density which is indeed confirmed by the shift of the potential towards positive values as revealed by the KPFM potential image (figure 4d). This reduction of the electron trap density may reinforce the ambipolar behaviour of the OTFTs. This effect combined with the larger capacitance of the PVA/SiO₂ layer which should allow an increased charge accumulation may explain the improved on–off ratio and output currents. Nonetheless, an obstacle assisted with such systems is increasing leakage currents by adding nanoparticles that causes additional leakage paths. One possible description of appearing such leakage paths is the attraction between charge carriers injected from the channel to dielectric layer and polarized nanoparticles. The attracted carriers flow along the direction of the gate electric field through successive attraction repulsion interactions with positive and negative sides of nanoparticles as has been proposed in [18]. Altogether, controlling the dielectric surface chemistry thus allows tuning the behaviour and performances of OTFTs.

5. Conclusions

In this study, neat PVA and PVA/SiO₂ nanocomposite films were fabricated as dielectric layers and their structure and electrical properties were investigated in the form of transistors based on DH4T as an active layer. Both types of devices showed ambipolar behaviour with hole and electron mobilities in the low-voltage range. The reduction of the electron

trap density at the interface due to the ‘neutralization’ of the PVA hydroxyl groups allowed us to obtain an ambipolar behaviour with DH4T-based transistors. The nanocomposite dielectric films with PVA and SiO₂ nanoparticles showed a larger capacitance, a less hydrophilic and rougher surface. The transistors fabricated with this nanocomposite dielectric showed considerable leakage currents in spite of improving some other electrical properties such as on–off ratio and output conduction. KPFM measurements revealed a shift of the surface potential in the channel towards positive values. All these observations show that modifying both the dielectric properties of the organic films and its surface chemical properties allows us to tune the performance of the OTFTs. On the bright side, polymeric materials might have the potential to be considered as gate dielectrics in optoelectronic devices because of the simple processing and the capability of enhanced capacitance by adding nanoparticles. However, on the not-so-bright side, the leakage problem assisted with polymeric nanocomposites would bring about constraints to their commercialization and is a challenge, which needs to overcome.

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References

- [1] Lee W H, Choi H H, Kim D H and Cho K 2014 *Adv. Mater.* **26** 1660
- [2] Lin Y H, Faber H, Zhao K, Wang Q, Amassian A, McLachlan M *et al* 2013 *Adv. Mater.* **25** 4340
- [3] An T K, Park S J, Ahn E S, Jang S H, Kim Y, Kim K *et al* 2015 *Dyes Pigm.* **114** 60
- [4] Dodabalapur A 2006 *Mater. Today* **9** 24
- [5] Torsi L, Tafuri A, Cioffi N, Gallazzi M C, Sassella A, Sabbatini L *et al* 2003 *Sens. Actuators B* **93** 257
- [6] Kim G, Kang S J, Dutta G K, Han Y K, Shin T J, Noh Y Y *et al* 2014 *J. Am. Chem. Soc.* **136** 9477
- [7] Kim M J, An T K, Kim S O, Cha H, Xiofeng T, Park C E *et al* 2015 *Mater. Chem. Phys.* **153** 63
- [8] Tan X, Park J M, Ma J Y and Kim Y H 2015 *J. Nanosci. Nanotechnol.* **15** 1396
- [9] Park K H, Yun H J, Lu W, Chung D S, Kwon S K and Kim Y H 2014 *Dyes Pigm.* **103** 214
- [10] Chua L L, Zaumseil J, Chang J F, Ou E C W, Ho P K H, Sirringhaus H *et al* 2005 *Nature* **434** 194
- [11] Yoo H, Ghittorelli M, Smits E C P, Gelinck G H, Lee H K, Torricelli F *et al* 2016 *Sci. Rep.* **6** 35585
- [12] Yang S Y, Shin K and Park C E 2005 *Adv. Funct. Mater.* **15** 1806
- [13] Nunes J G, Zane S G and Meth J S 2005 *J. Appl. Phys.* **98** 104503

- [14] Klauk H, Halik M, Zschieschang U, Schmid G, Radlik W and Weber W 2002 *J. Appl. Phys.* **92** 5259
- [15] Schroeder R, Majewski L A and Grell M 2005 *Adv. Mater.* **17** 1535
- [16] Chang J W, Hsu W L, Wu C Y, Guo T F and Wen T C 2010 *Org. Electron.* **11** 1613
- [17] Yang C Y, Cheng S S, Ou C W, Chuang Y C, Wu M C and Dhananjay C W C 2008 *J. Appl. Phys.* **103** 094519
- [18] Kwon J H, Zhang X, Piao S, Choi H, Bae J H and Park J 2016 *Polymers* **8** 88
- [19] Chen F C, Chu C W, He J, Yang Y and Lin J L 2004 *Appl. Phys. Lett.* **85** 3295
- [20] Maliakal A, Katz H, Cotts P M, Subramoney S and Mirau P 2005 *J. Am. Chem. Soc.* **127** 14655
- [21] Jung C, Maliakal A, Sidorenko A and Siegrist T 2007 *Appl. Phys. Lett.* **90** 062111
- [22] Choi J W, Yoon H G and Kim J K 2010 *Org. Electron.* **11** 1145
- [23] Singh T B, Meghdadi F, Günes S, Marjanovic N, Horowitz G, Lang P *et al* 2005 *Adv. Mater.* **17** 2315
- [24] Park S J and Lee D G 2006 *Curr. Appl. Phys.* **6** e182
- [25] Leroy J, Boucher N, Sergeyev S, Sferrazza M and Geerts Y H 2007 *Eur. J. Org. Chem.* **8** 1256
- [26] Afsharimani N and Nysten B 2013 *Thin Solid Films* **536** 295
- [27] Duca M D, Plosceanu C L and Pop T 1998 *Polym. Degrad. Stab.* **61** 65
- [28] Jeong S, Kim D, Lee S, Park B K and Moon J 2006 *Appl. Phys. Lett.* **89** 092101
- [29] Ogura A, Saitoh H, Kosemura D, Kakemura Y T Y, Takei M, Koganezawa T *et al* 2009 *Solid State Lett.* **12** H117
- [30] Wang J, Lee W H, Ho J and Hu T 2009 *J. Mater. Sci.: Mater. Electron.* **20** 355
- [31] Joshi D P, Pant G, Arora N and Nainwal S 2017 *Heliyon* **3** e00253
- [32] Wrachien N, Lago N, Rizzo A, D'Alpaos R, Stefani A, Turatti G *et al* 2015 *Microelectron. Reliab.* **55** 1790
- [33] Serban D A, Kilchytska V, Vlad A, Martin-Hoyas A, Nysten B, Jonas A M *et al* 2008 *Appl. Phys. Lett.* **92** 143503
- [34] Singh T, Senkarabacak P, Sariciftci N S, Tanda A, Lackner C, Hagelauer R *et al* 2006 *Appl. Phys. Lett.* **89** 033512
- [35] Kepler R 1960 *Phys. Rev.* **119** 1226
- [36] Singh T B, Günes S, Marjanovic N and Sariciftci N S 2005 *J. Appl. Phys.* **97** 114508
- [37] Mojumdar S C and Raki L 2005 *J. Therm. Anal. Calorim.* **82** 89
- [38] Chrissafis K, Paraskevopoulos K M, Papageorgiou G Z and Bikiaris D N 2008 *J. Appl. Polym. Sci.* **110** 1739
- [39] Alexandrova S and Szekeres A 2001 *Phys. Status Solidi A* **187** 499
- [40] Olthuis W and Bergveld P 1992 *IEEE Trans. Dielectr. Electr. Insul.* **27** 691
- [41] Duong D L, Leeb S M and Lee Y H 2012 *J. Mater. Chem.* **22** 1994