Sol–Gel PMMA–ZrO₂ Hybrid Layers as Gate Dielectric for Low-Temperature ZnO-Based Thin-Film Transistors

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ABSTRACT: We report a simple sol–gel process for the deposition of poly(methyl methacrylate) (PMMA)–ZrO₂ organic–inorganic hybrid films at low temperature and studied their properties as a function of the molar ratios of the precursors in the hybrid sol–gel solution, which included zirconium propoxide as the inorganic (zirconia) source, methyl methacrylate as the organic source, and 3-trimethoxy-silyl-propyl-methacrylate (TMSPM) as the coupling agent to enhance the compatibility between the organic and inorganic phases. The hybrid thin-film deposition was done on glass slide substrates by the dip-coating method. After deposition, the films were heat-treated at 100 °C for 24 h. The analysis of the hybrid films included Fourier transform infrared spectroscopy to identify their chemical groups and thermogravimetric analysis to determine the content of their organic and inorganic components. In addition, capacitance–voltage (C–V) and current–voltage (I–V) curves in metal–insulator–metal structures, using gold as metal contacts, were measured to find the dielectric constant and leakage current of the PMMA–ZrO₂ hybrid films. Finally, because of their adequate dielectric characteristics, single hybrid layers were deposited on indium tin oxide-coated glass substrates and were tested as gate dielectric in thin-film transistors (TFTs), using sputtered ZnO layers as the semiconductor active channel. We measured the output electrical response and transfer characteristics of these hybrid dielectric gate-based devices and determined their main electrical parameters as a function of the TMSPM content in the hybrid dielectric gate layer. The better TFT electrical behavior presents field effect mobility of 0.48 cm²/V s, low threshold voltage of 3.3 V, and on/off current ratio of 10⁵, and it was obtained by using PMMA–ZrO₂ with 0.3 TMSPM content as the gate dielectric layer. The values obtained for the electrical parameters show that PMMA–ZrO₂ hybrid films are quite suitable for dielectric gate applications in TFTs.

1. INTRODUCTION

Thin-film transistors (TFTs) are currently of great interest for potential applications in different portable devices,¹ sensors,² radio-frequency (RF) identification tags,³ smart cards, and backplane circuits for active matrix displays,⁴ among others. In recent years, the technology has been focused on the development of new materials, which can be easily processed at low temperature in large-area applications,⁵ low power consumption, low operating voltage,²–⁹ and are compatible with different substrates for transparent electronics.¹⁰ The organic–inorganic hybrid gate dielectrics address many of these requirements and therefore represent promising materials.²–⁵,¹⁰ The transistor parameters critically depend not only on the thickness and the dielectric properties of the gate insulator but also on the interface formed between dielectric and semiconductor layers, where the trapped charge has strong influence on the device electrical behavior.² For a better interface, the hybrid gate dielectrics must have good compatibility with either organic or inorganic semiconductors.¹¹ The organic materials for dielectric gate applications offer a smooth surface and good compatibility to grow organic semiconductors, but they have low dielectric constant and therefore low capacitance.¹² On the other hand, the high processing temperature of inorganic dielectric materials with high k values is not compatible with deposition on flexible plastic substrates required in flexible electronics.⁵,¹³ The appropriated combination of organic and inorganic phases in organic–inorganic hybrid materials, strongly bonded at the molecular level, is a new approach in the development of materials with reinforced specific properties for a wide variety of applications.¹⁴ Many of these hybrid materials can be obtained by the sol–gel method, which is a solution, low-
temperature process compatible with deposition on large-area substrates.10,15 Hybrid materials have been investigated for dielectric gate applications in organic or hybrid electronic devices.4,5,16 A simple configuration, frequently employed, is a hybrid organic–inorganic bilayer dielectric gate, where the inorganic layer increases the capacitance and the organic one provides a smooth surface at the dielectric–semiconductor interface.1 High-k inorganic nanoparticles, such as ZrO2 and TiO2,13 embedded in a polymeric matrix, such as poly(methyl methacrylate) (PMMA) and poly(vinylpyrrolidone), have also been tested as dielectric gate layers in TFTs. In this configuration, the inorganic nanoparticles tend to agglomerate, increasing the surface roughness of the hybrid layers, and the inorganic–organic interaction is rather low, producing devices with rough interface and poor electrical parameters, such as high gate leakage current1 and low on/off current ratio.1

Another less-explored approach is the employment of class II inorganic–organic hybrid layers,16–20 where the phases are much stronger linked through covalent and/or ionic bonds at the molecular level. This class of hybrid materials forms a cross-linked inorganic–organic network, the properties of which depend on the amount of both phases and the chemical interaction between them.18 If a proper link is attained between organic and inorganic phases, the precipitation of phases can be avoided, yielding a homogeneous hybrid network with high optical transparency and smooth surface at the macroscopic level.16 In previous studies, we have reported a low-temperature sol–gel process to deposit PMMA-based hybrid films.16–20 In this deposition process, the cross-linking between PMMA and some inorganic oxides, such as SiO2, TiO2, and ZrO2, is attained through trimethoxy-silyl-propyl-methacrylate (TMSPM) molecules as a coupling agent, with chemical groups compatible with both inorganic and organic phases. The formation of the hybrid material consists in the in situ polymerization of the organic phase during hydrolysis and condensation of the inorganic one. Furthermore, the properties of these hybrid films, including the dielectric ones, can be adjusted by controlling the contents of organic and inorganic components. This way, the PMMA–SiO2 and PMMA–TiO2 hybrid layers with good dielectric characteristics were obtained and tested as dielectric gates in n-type ZnO-based TFTs with promising results.16,19

In this work, we report the sol–gel process to obtain highly transparent, cross-linked, PMMA–ZrO2 hybrid films deposited on glass and indium tin oxide (ITO)-coated glass substrates at low temperature. The development of such a process was based on our previous experience on the film deposition of PMMA–inorganic oxide hybrid systems described above. The hybrid films were characterized by several experimental techniques as a function of their organic to inorganic content, and it was found, according to their dielectric properties, that they were good candidates for dielectric gate layers in TFTs. Therefore, TFTs with a ZnO/PMMA–ZrO2/ITO/glass structure were assembled in a clean-room environment and analyzed to determine their main electrical parameters, such as threshold voltage, channel mobility, and $I_{on}/I_{off}$ current ratio as functions of the composition of the hybrid dielectric gates.

2. RESULTS AND DISCUSSION

The formation of the hybrid network was verified by identifying the chemical groups of both phases, PMMA and ZrO2, in the Fourier transform infrared (FTIR) spectra of the three samples, shown in Figure 1. The three spectra display the same absorption bands with the intensity increasing with the coupling agent content, which is basically due to the increasing hybrid film thickness. In this graph, some absorption bands, the most representative, are labeled with their corresponding vibration modes. Centered at about 3350 cm$^{-1}$ is observed the typical wide and strong band attributed to O–H stretching modes, mainly from Zr–OH groups resulting from the incomplete inorganic condensation in the sol–gel process, which is due to the low temperature of the curing process.4 The groups of the organic phase produce the absorption bands observed at 2945, 1730, and 1640 cm$^{-1}$, which can be related to the stretching vibration modes of the C–H, C=O, and C=C groups, respectively.15 The C=C signal arises from these groups, which are contained in the coupling agent and MMA monomer molecules. These groups are the bonding point for the polymerization of PMMA by free radicals,18 and their presence in the hybrid material evidences some incomplete polymerization of the MMA monomer. In the wavenumber region between 950 and 1230 cm$^{-1}$, several vibration modes overlap: at 1075 cm$^{-1}$, there is a band related to Si–C bonds of the coupling agent molecules, at 1060–1100 cm$^{-1}$, there is another band produced by the C–O–C stretching vibration modes of the acrylic groups, and at 1126 cm$^{-1}$, there is a band assigned to Zr–O–C, which evidences some incomplete hydrolysis of zirconium propoxide (ZP). In the lower wavenumber region, the absorption due to vibration modes of Si–O–Zr groups, at 930 cm$^{-1}$, has been reported due to the interaction between the inorganic phase and the organic one through the coupling agent molecule.17 The formation of the inorganic ZrO2 phase in the hybrid material is corroborated by the signals observed at ~490 and ~630 cm$^{-1}$ related to the vibration modes of the Zr–O groups.21 Thus, FTIR analysis of the hybrid material shows the formation of both organic and inorganic phases and evidences their interaction through the coupling agent molecules. Furthermore, this analysis also provides some insight into the reaction mechanism for the formation of the organic–inorganic hybrid network. In the beginning, Si–OH groups result from the hydrolysis of TMSPM molecules. These bind with the Zr–OH groups of the inorganic precursor, forming Si–O–Zr groups and then Zr–O–Zr ones.17

![Figure 1. FTIR spectra of the PMMA–ZrO2 hybrid layers with different TMSPM content.](image-url)
The high reactivity of the inorganic precursor (ZP) in the aqueous medium results in fast hydrolysis, favoring the growth of the inorganic amorphous network constituted by Zr–O–Zr groups, which have affinity with one extreme of the coupling agent hydrolyzed molecules. On the other hand, the free radical method results in the polymerization of PMMA organic phase by breaking the C=C groups of the MMA monomer molecules and binding to the other extreme of the coupling agent molecules. This process occurs with some selectivity due to the hydrophobicity of the organic molecules.

Figure 2 shows the thermogravimetric analysis (TGA) measurements for the three types of PMMA–ZrO2 hybrid materials with different contents of TMSPM showing three temperature regions for the weight loss of the samples.

*Figure 2.* TGA analysis of the PMMA–ZrO2 hybrid materials with different contents of TMSPM showing three temperature regions for the weight loss of the samples.

films, where three stages of weight loss were clearly identified for all of the samples. At low temperature, up to 230 °C, there is a gradual, small weight loss between 4 and 6%, corresponding to the release of residual solvents from the amorphous hybrid network and due the condensation of hydrolyzed species, which remains inside (as previously observed in the FTIR spectra), which is a nonoxidative degradation. At a higher temperature, up to 400 °C, there is a large and abrupt weight loss followed by a small and gradual weight loss, which continues up to about 650 °C. The abrupt weight loss is related to the oxidative degradation of PMMA as the organic component in the hybrid material. The subsequent gradual weight loss is associated with the thermal decomposition of PMMA chains, which require more energy to break due to their interaction with the inorganic phase through the coupling agent. This portion of PMMA is that which decomposes in the 400–650 °C temperature range, and it is about 7–8% in the three hybrid samples. This is because pure PMMA completely decomposes at 385–400 °C, as reported in the literature. In the 600–1100 °C temperature range, there is no more weight loss because the remaining solvents and organic phase have been completely removed and only the inorganic phase remains. The remainder material is constituted by the inorganic components: ZrO2 from the inorganic source and SiO2 from the coupling agent component. On the basis of this analysis, the weight content (%) of each phase and remaining solvents in each sample were calculated, and the results are shown in the inset of Figure 2 and plotted in Figure 3. The observed trend in these results is that the increasing TMSPM molar ratio decreases the inorganic content in the hybrid samples, increasing the organic one on the other hand. This is because the coupling agent added to the hybrid network contains an acrylic group, which has a larger mass than that of the silicate one. The same trend with the TMSPM content was reported for the PMMA–SiO2 hybrid system.

*Figure 3.* Content of the components (solvents, organic, and inorganic) of the PMMA–ZrO2 hybrid materials as a function of the TMSPM molar ratio, determined by TGA analysis.

contribution of the inorganic components. For this, the values of C, measured at several frequencies, were substituted in the formula C = ϵrA/d, where ϵr is the dielectric constant, A is the area of the capacitor, and d is the thickness of the hybrid dielectric layer. The dielectric constant values, at different frequencies, as a function of the TMSPM content in the PMMA–ZrO2 hybrid films are plotted in Figure 5. The dielectric constant values are in the 4–12 range, larger than the dielectric constant of the organic component PMMA, which is 3.9, due to the contribution of the inorganic components. The remaining solvents, due to the low-temperature processing, with high-polarizability groups and the presence of monomers because of the incomplete polymerization also have some influence on these rather larger values of the dielectric constants. The observed trend is that the dielectric constant decreases with the TMSPM content in
the hybrid films; therefore, according to TGA measurements, it decreases with the increasing organic phase content, as expected due to its lower dielectric constant. On the other hand, the dielectric constant increases when the frequency is reduced because at a lower frequency, there are more contributions to the polarizability in the hybrid materials. The enhancement in the dielectric response is another evidence of the appropriate integration of the organic−inorganic components in the hybrid dielectric layers.20 These results show that the PMMA−ZrO2 hybrid layers have appropriate dielectric characteristics to be employed as a dielectric gate layer in devices like TFTs, where low leakage current and high dielectric constant can enhance the device performance.

Figure 6 displays (a) the schematic cross section of the common-bottom PMMA−ZrO2 dielectric gate, ZnO-based TFT configuration, and (b) the corresponding scanning electron microscopy (SEM) image of the device cross section, where the PMMA−ZrO2 layer was deposited with 1:0.3:1 molar ratio.
in semilog scales, measured at $V_{ds} = 6 \text{ V}$, as a function of $V_{gs}$ for the three types of devices. In these curves, it is observed that the current measured at both lowest and highest applied $V_{gs}$ voltages increases for the devices with the hybrid dielectric gate containing more TMSPM, which could be due to the influence of the leakage current on the dielectric layers. In Figure 8a, for the device with dielectric gate containing 0.3 M TMSPM, the lowest value of the current, corresponding to the off-state of this device, is on the order of $10^{-12} \text{ A}$. This is a rather low off current ($I_{off}$), and it is comparable to the values observed in ZnO-based TFTs with inorganic dielectric layers. Meanwhile, the current values at the highest applied voltage correspond to the on-current ($I_{on}$) of the device, which is on the order of $10^{-7} \text{ A}$ (almost $10^{-6} \text{ A}$); therefore, the $I_{on}/I_{off}$ current ratio for this device is on the order of $10^{5}$ (almost $10^{6}$). In Figure 8b,c, the off-currents are not completely defined; however, we can estimate the $I_{on}/I_{off}$ current ratios for these devices by taking the lowest currents as the off-currents to obtain $10^{4}$ and $10^{2}$ for the devices with hybrid dielectric gates with 0.5 and 0.75 M TMSPM contents, respectively.

In the right $y$ axis of Figure 8 are plotted the sqrt $I_{ds}$ versus $V_{gs}$ curves of the three devices, which were fit to eq 1 to determine their threshold voltage and channel mobility. The scattered plots correspond to experimental data, and the solid lines represent the best linear fits to eq 1. A clear trend is observed in the interception of the solid straight line with the $V_{gs}$ axis, which is the threshold voltage of the device; it decreases with the increasing TMSPM content in the hybrid dielectric gate of the devices. Because the threshold voltages are positive, the TFT devices operate in the enhancement mode, that is, at $V_{gs} = 0 \text{ V}$, the devices are in the off-state and a positive gate bias is required to form a conductive channel and thus turn on the devices. This is an important advantage for low power consumption. It is also observed that when negative voltage is applied, the current remains low, unlike some devices reported in the literature in which undesirable leakage current is observed. On the other hand, the channel mobility in our devices, on the order of $10^{-1} \text{ cm}^2/\text{V s}$, shown in Table 1, has similar values to those of others reported in the literature for ZnO-based TFTs. The highest value of mobility, $0.48 \text{ cm}^2/\text{V s}$, was obtained in the devices with hybrid dielectric gates with the lowest content of TMSPM (0.3 M).

| TMSPM content | thickness (nm) | $\kappa$ (1 MHz) | $V_{t}$ (V) | $\mu_{FET}$ (cm$^2$/V s) | $I_{on/off}$ | $SS$ (mV/dec) |
|---------------|----------------|-----------------|-------------|-------------------------|-------------|-------------|
| 0.3           | 65             | 6.8             | 3.3         | 0.48                    | 10$^7$      | 476         |
| 0.5           | 110            | 7.2             | 2.3         | 0.35                    | 10$^7$      | 640         |
| 0.75          | 209            | 4.2             | 0.9         | 0.34                    | 10$^2$      | 733         |
\[ S = \left( \frac{d(\log_{10} I_d)}{dV_{ds}} \right)^{-1} \]

From these results, describing the electrical response of the ZnO-based TFT devices, it can be concluded that the PMMA–ZrO₂ hybrid dielectric layers, processed by the sol–gel method at low temperature, have very good performance as a dielectric gate in the TFT devices. The hybrid dielectric gate with 0.3 M content produced the best device with 0.48 cm²/V s channel mobility and 3.3 V threshold voltage. The on/off current ratio is 10⁵, which is higher than the values reported in other works for ZnO-based devices using hybrid dielectric gates. ²⁻⁹⁻⁻² Several factors concur in this hybrid dielectric gate layer to enhance the device performance. The smooth hybrid layer with more inorganic content produces a better dielectric–semiconductor interface, which favors the low density of traps. ²⁻⁹⁻⁻² The small thickness increases the capacitance, and the lowest content of remnant solvent reduces the leakage current.

3. CONCLUSIONS

We reported here the low-temperature deposition process and the characteristics of hybrid dielectric layers obtained from a sol–gel reaction among ZP, TMSPM, and MMA precursors at variable TMSPM molar ratios. The resulting PMMA–ZrO₂ hybrid layers were employed as dielectric layers in MIM to determine the influence of the TMSPM content on their dielectric characteristics. Furthermore, the hybrid dielectric layers were tested as dielectric gate layers in ZnO-based TFTs and the influence of TMSPM concentration on the TFT electrical performance was also investigated. All of the TFTs had low threshold voltage, channel mobility on the order of 10⁻¹⁰ cm²/V s, and on/off current ratio in the range 10⁻⁵⁻¹⁰⁴. The latter parameter had more influence of the leakage current in the hybrid dielectric gate layers. The devices with best performance parameters were obtained with the hybrid dielectric gate layer containing the lowest amount, 0.3 M, of TMSPM, which had 3.3 V, 0.48 × 10⁻¹⁰ cm²/V s, and 10⁵ values for threshold voltage, channel mobility, and on/off current ratio, respectively. The characteristics of larger amount of inorganic component, smaller thickness, and less amount of remnant solvents in this hybrid dielectric layer yield to improve its dielectric properties and then the better performance of the ZnO-based TFT devices. The electrical parameters of this device are comparable to those of other ZnO-based TFTs reported in the literature, which uses organic, hybrid, and even inorganic dielectric gate layers. That is why the PMMA–ZrO₂ cross-linked hybrid system represents a novel hybrid organic–inorganic dielectric material, the low-temperature processing of which is quite appropriate for its deposition on rigid or flexible substrates for the development of flexible electronic devices.

4. EXPERIMENTAL SECTION

To prepare the sol–gel hybrid solution, zirconium propoxide (ZP) was used as the inorganic precursor, methyl methacrylate (MMA) as the source of the organic component, and 3-trimethoxy-silyl-propyl-methacrylate (TMSPM) as the coupling agent. Benzoyl peroxide was used to start the MMA polymerization. To hydrolyze the inorganic ZP precursor, ethyl alcohol was used as the solvent and HNO₃ as the catalyst. The TMSPM coupling agent solution was prepared using deionized water and HCl. Three types of PMMA–ZrO₂ hybrid films were deposited from three hybrid precursor solutions obtained by mixing the ZP, TMSPM, and MMA solutions with molar ratios of 1:0.3:1, 1:0.5:1, and 1:0.75:1, respectively, where the TMSPM content is increased from 0.3 to 0.75, keeping the same ZP¹ and MMA¹ molar ratios. The hybrid films were deposited by dip-coating on conventional glass slide substrates. Before deposition, the substrates were ultrasonically cleaned in acetone (5 min), isopropyl alcohol (5 min), and deionized water (5 min) and finally dried with nitrogen. The velocity for both immersion and removal during the dipping process was 3 cm/min. After deposition, the wet hybrid films were baked at 100 °C in air (atmospheric pressure) for 24 h in a conventional oven. The resulting hybrid films in the three cases were homogeneous, highly transparent, and very well adhered to the glass substrates. The PMMA–ZrO₂ hybrid films were analyzed by Fourier transform infrared (FTIR) spectroscopy, thermogravimetric analysis (TGA), and scanning electron microscopy (SEM) measurements. The FTIR spectra of the hybrid samples deposited on Cr/Au (10/100 nm)-coated glass substrates were obtained using a Gx PerkinElmer system in the reflectance mode (GA-ATR) with a resolution of 4 cm⁻¹, in the wavenumber range of 400–4000 cm⁻¹. The TGA experiments were performed using a TA Instruments SDT-Q600 TGA–differential scanning calorimetry, from room temperature to 1100 °C under synthetic air flow (50 mL/min) using a heating rate of 10 °C/min. The measurements were carried out using PMMA–ZrO₂ powders obtained by using the same temperature and baking time conditions as those used for the curing of the hybrid films. Cross-sectional SEM images of the hybrid films were obtained in a Zeiss Supra-40. The hybrid samples were coated with a thin gold layer before the SEM measurements.

The dielectric characterization of the PMMA–ZrO₂ layers was done in MIM structures fabricated by depositing the hybrid layers on gold-coated glass substrates. The top gold electrodes (100 nm) were evaporated using a shadow mask to define top circular contacts (50, 100, 200, 300, and 500 μm diameters). The current versus voltage curves of the MIM structures were measured at room temperature using a semiconductor parameter analyzer (Keithley 4200), and a C–V analyzer (HP4284A; Hewlett Packard) was used to measure their capacitance versus voltage characteristics, at low and high frequencies (1 kHz to 1 MHz). For the fabrication of TFT devices, a single layer of PMMA–ZrO₂ was deposited as a dielectric gate on ITO-coated glass substrates previously cleaned in the same way as the glass substrates. Next, as the active semiconductor layer, an n-type ZnO thin film (40 nm) was sputtered on the hybrid dielectric gate using a commercially sintered ZnO target (99.999%). The ZnO RF sputtering process was performed at room temperature and using 50 W power. To complete the TFT structure, patterned source and drain Al (100 nm) electrodes were evaporated through a metallic shadow mask. The width of the Al electrodes was 500 μm, and the channel length of the devices was 40 μm. The fabrication of the devices was performed under clean-room environment (US-class 10 000) at the University of Texas at Dallas. A 4200 Keithley semiconductor parameter analyzer was used to carry out current–voltage (I–V) measurements at room temperature to determine the electrical characteristics of the hybrid gate-based TFT devices.
Organic Field-Effect Transistor With PMMA/ZrO$_2$ Bilayer.

REFERENCES

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REFERENCES

(1) Lu, A.; Sun, J.; Jiang, J.; Wan, Q. Low-Voltage Transparent Electric-Double-Layer ZnO-Based Thin-Film Transistors For Portable Transparent Electronics. Appl. Phys. Lett. 2010, 96, No. 043114.

(2) Choi, C. G.; Bae, B.-S. Organic–Inorganic Hybrid Materials As Solution Processible Gate Insulator For Organic Thin Film Transistors. Org. Electron. 2007, 8, 743–748.

(3) Zhang, C.; Wang, H.; Shi, Z.; Cui, Z.; Yan, D. UV-Directly Patternable Organic–Inorganic Hybrid Composite Dielectrics for Organics Thin-Films Transistors. Org. Electron. 2012, 13, 3302–3309.

(4) Wang, Y.; Kim, H. UV-Curable Organic–Inorganic Hybrid Gate Dielectrics For Organic Thin Film Transistors. Org. Electron. 2012, 13, 2997–3003.

(5) Ha, Y.-g.; Jeong, S.; Wu, J.; Kim, M.-G.; Dravid, V. P.; Facchetti, A.; Marks, T. J. Flexible Low-Voltage Organic Thin-Film Transistors Enabled by Low-Temperature, Ambient Solution-Processable Inorganic/Organic Hybrid Gate Dielectrics. J. Am. Chem. Soc. 2010, 132, 17426–17434.

(6) Kwon, J. S.; Park, H. W.; Kim, D. H.; Kwark, Y.-J. Solvent-Free Processable and Photo-Patternable Hybrid Gate Dielectric for Flexible Top-Gate Organic Field-Effect Transistors. ACS Appl. Mater. Interfaces 2017, 9, 5366–5374.

(7) Je, S. Y.; Son, B.-G.; Kim, H.-G.; Park, M.-Y.; Do, L.-M.; Choi, R.; Jeong, J. K. Solution-Processable LaZrO$_3$/SiO$_2$ Gate Dielectric at Low Temperature of 180 °C for High-Performance Metal Oxide Field-Effect Transistors. ACS Appl. Mater. Interfaces 2014, 6, 18693–18703.

(8) Choi, C. G.; Bae, B.-S. Organic–Inorganic Hybrid Gate Dielectrics for Low-Voltage Pentacene Organic Thin Film Transistors. Synth. Met. 2009, 159, 1288–1291.

(9) Shang, L.; Liu, M.; Tu, D.; Liu, G.; Liu, X.; Ji, Z. Low-Voltage Organic Field-Effect Transistor With PMMA/ZrO$_2$ Bilayer. IEEE Trans. Electron Devices 2009, 56, 370–372.

(10) Jung, Y.; Jun, T.; Kim, A.; Song, K.; Yeo, T. H.; Moon, J. Direct Photopatternable Organic–Inorganic Hybrid Gate Dielectric For Solution-Processed Flexible ZnO Thin Film Transistors. J. Mater. Chem. 2011, 21, 11879–11885.

(11) Kim, K.; Song, H. W.; Shin, K.; Kim, S. H.; Park, C. E. Photo-Cross-Linkable Organic–Inorganic Hybrid Gate Dielectric for High Performance Organic Thin Film Transistors. J. Phys. Chem. C 2016, 120, 5790–5796.

(12) Kim, G. H.; Yoon, S.-M.; Kang, S. Y.; Kim, C. A.; Ahn, S. D.; Suh, K. S. Organic Field-Effect Transistors With Thermal-Cured Polyacrylate Gate Dielectric Films. Thin Solid Films 2008, 516, 1574–1577.

(13) Lee, J.-W.; Ju, B.-K.; Jang, J.; Yoon, Y.-S.; Kim, J.-K. Highly Mobility Organic Transistor Patterned by Shadow Mask With All Structure on a Plastic Substrate. J. Mater. Sci. 2007, 42, 1026–1030.

(14) Liu, C.-T.; Lee, W.-H.; Su, J.-F. Pentacene-Based Thin Film Transistor With Inkjet-Printed Nanocomposite High-k Dielectrics. Act. Passive Electron. Compon. 2012, 2012, No. 921738.