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Universal Solution-Processed High-*k* Amorphous Oxide Dielectrics for High-Performance Organic Thin Film Transistors

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Solution-processed high-*k* dielectrics for organic thin-film transistors (OTFTs) have been widely studied with the objective of achieving high performance and low-cost OTFTs for next-generation electronics. In this study, we introduce a universal solution fabrication of high-*k* amorphous oxide dielectrics for high-performance OTFTs. The amorphous oxide films by solution processes are featured with smooth surface, uniform composition and excellent dielectric properties. With $ZrTiO_x$ as a typical example, the *k* value and capacitance as high as 53 and 467 nF/cm^2 could be achieved. The polystyrene (PS) modification of $ZrTiO_x$ dielectric films results in a leakage current as low as $4\times10^{-8} A/cm^2$. Based on their implementation as a gate insulator, the solution-processed high-*k* $ZrTiO_x$ dielectric films realize high and stable performances performance OTFTs during operation at a low voltage. A carrier mobility of 0.58 $cm^2V^{-1}s^{-1}$, an on/off current ratio of 10^4, and a low operating voltage of 6 V were achieved through simple control of annealing temperature. Our results show the possibility of the solution-processed high-*k* amorphous oxide dielectric layer as a gate insulator for OTFTs. We believe that this high-*k* amorphous oxide dielectric films offer great potentials for next-generation high-performance organic electronics, especially low-voltage electronics.

Introduction

Organic thin film transistors (OTFTs) have attracted considerable interests for low cost electronic devices such as radio-frequency identification (RFID) tags, sensors, and electronic paper.[1-4] For organic semiconductors, the intrinsically low carrier motilities could limit the performance of OTFTs. To achieve desirable channel currents, the relatively high operating voltages (＞20 V) are mostly required, which may lead to high power consumption in organic electronics. To overcome this key drawback, a feasible solution is to use a high dielectric constant (*k*) layer in OTFT devices.[5] In the last years, various high-*k* oxide dielectrics, such as Al_2O_3[6], HfO_2[7], ZrO_2[8], Y_2O_3[9] and La_2O_3[10], have attracted extensive research efforts for low-voltage organic devices. To obtain a high *k* value, the highly crystalline oxides are usually indispensable. Nevertheless, the grain boundary in polycrystalline films could become the channel of leakage current, limiting the performance of TFTs. Moreover, the mechanical stability, films smoothness and device uniformity of polycrystalline thin films are less desired for high-performance electronic devices. In contrast, recent results suggested that the corresponding amorphous oxides with smooth surface and high density offer intriguing and potentially useful properties for the high-performance TFTs.[11-13] In particular, complex oxides, such as alloys of Zr, Hf and La oxides, are often used to have better dielectric properties.[14-16] However, the low *k* (＜10) values of most amorphous oxides greatly limit the large-scale fabrication of high-performance OTFTs.

In the last years, various vapor routes, such as chemical vapor deposition,[17] atomic layer deposition,[18] sputtering,[19] and pulsed laser deposition,[20] are usually used for the preparation of oxide dielectric layers. For example, $Zr_xTi_yO_z$ thin films by atomic layer deposition possess the high *k* of 45-65, and the leakage current of 10^{-4} A/cm^2 at 0.2 MV/cm.[21] However, these vapor growth techniques are mostly high cost due to the high vacuum equipment, leading to limited production and throughout. In contrast, solution methods, like sol-gel process, have attracted intensive interests due to the inexpensive equipment, large-scale production, and high throughput deposition. Recently, ultraviolent (UV) irradiation of sol-gel films have been shown as an effective method for the room-temperature fabrication of high *k* oxide dielectric films.[22,23] Amorphous $ZrTiO_x$ films can achieve a high *k* of 18.9.[23] Nevertheless, the high cost of short wavelength UV equipment may limit the mass production. Therefore, it is of great significance to facilely and low-lost fabricate high-*k* amorphous oxide dielectrics for high-performance electronics.

In this paper, we report a general strategy on the scalable fabrication of high-*k* amorphous complex oxide dielectric films $ZrTiO_x$ by a simple sol-gel spin-coating method. We found the suppressed crystallization and significantly high density of complex oxides lead to high dielectric properties, such as a high dielectric constant of 53 and high capacitance of 467 nF/cm^2. As a proof of concept, a high-performance low-voltage pentacene-based OTFT with a low operating voltage of 6 V, high carrier mobility of 0.58 $cm^2V^{-1}s^{-1}$, and on/off current ratio of 10^4, was realized with the polystyrene (PS) modified high-*k* $ZrTiO_x$ dielectric layer. Consequently, the solution-processed high-*k* amorphous oxide dielectric films provide an effective approach for high-performance organic electronics, especially low-voltage transistors and memories.

Experimental

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Preparation of amorphous oxide dielectric films

In a typical process for $ZrTiO_x$ film, 0.4 mmol zirconium oxychloride octahydrate ($ZrOCl_2{\cdot}8H_2O$) was dissolved in 1 ml 2-methoxyethanol ($HOCH_2CH_2OCH_3$) to form zirconium precursor solution, while 0.4 mmol tetrabutyl titanate ($Cl_6H_{36}O_4Ti$) stabilized by acetylacetone was dissolved in 1 ml 2-methoxyethanol to form titanium precursor solution. Then the $ZrTiO_x$ precursor solution was obtained by thoroughly mixing the two solutions and stirring at 100 ℃ for 60 min to obtain a transparent precursor solution. The precursor solution was spin-coated on p-type silicon substrate at 2500 rpm for 60 s to form a uniform $ZrTiO_x$ film at room temperature. Then the sample was preheated at 300 ℃ for 10 min to remove the volatile organic components. These processes were repeated several times to obtain desired thickness. The as-deposited $ZrTiO_x$ films were then annealed at 400 to 750 ℃ in an air furnace for 2 hours. For comparison, the pure ZrO_x films were also prepared with the same process. The preparation of $LaTiO_x$ film is similar to that of $ZrTiO_x$ film by just changing the metal precursor ($La(NO_3)_3$ was selected as precursor).

Fabrication of OTFTs

To fabricate the pentacene-based OTFTs, a heavily doped n-type Si (100) wafer was used as the substrate. All of the substrates were carefully cleaned in acetone, followed with ethanol and deionized water using ultrasonication for 10 minutes each time. After that, the substrates were spin-coated with $ZrTiO_x$ precursor solution and annealed at setting temperature to obtain a smooth and dense layer. To improve the interface compatibility of amorphous oxide and organic active layers, a thin polystyrene (PS) layer was prepared on the $ZrTiO_x$ films by a simple spin coating process. 0.1 g PS was dissolved in 1 ml toluene to form a transparent solution. Then the PS solution was spin coated on the $ZrTiO_x$ films at 3000 rpm for 60 s and vacuum dried at 120 ℃ for 60 min to form PS layer. After that, the pentacene active layer and silver source/drain were deposited by thermal evaporation process. In a typical process, 50 nm thick pentacene was evaporated onto the PS/$ZrTiO_x$ dielectrics at a rate of 0.2 Å/s in a base pressure of 2.5×10^{-4} Pa. Then 80 nm silver source and drain electrodes were evaporated onto the pentacene through a shadow mask. The thickness of pentacene and silver layer were controlled by a calibrated quartz crystal monitor. The channel length (L) and the width (W) were fixed at 100 and 2000 μm, respectively.

Characterizations

The crystalline and phase were examined by Rigaku D/max-2500X-ray diffraction (XRD) using Cu K_α radiation analysis. Hitachi S-4800 scanning electron microscopy (SEM) and Vecco Dimension 3100 atomic force microscope (AFM) system were used to study the microstructure and surface. For leakage and capacitance characterization, a metal-insulator-metal (MIM) structure was fabricated by depositing 50 nm thick gold dots with diameter of 200 μm onto the dielectric layer through a shadow mask. The frequency dependence of capacitance density was tested by 7600 Plus Precision LCR Meter at a frequency of 100 Hz~100 KHz. The leakage properties were measured by Keithley 2400 Source-Meter. OTFTs were characterized using two Keithley 2400 Source-Meter units. All characterizations were carried out at room temperature in ambient air.

Results and discussion

Microstructure of amorphous oxide dielectric films

The crystalline and phase condition of $ZrTiO_x$ films annealed at temperatures from 600 to 750 ℃ were first studied, and the x-ray diffraction pattern (XRD) patterns are shown in Figure 1a. The results revealed that the $ZrTiO_x$ films are amorphous when annealed below 600 ℃, and begin to crystallize at about 650 ℃. When the annealed temperature increased to 750 ℃, the strong diffraction peaks are indexed to single $ZrTiO_4$ phase with high crystalline state. For comparison, the XRD patterns of the pure ZrO_2 film by sol-gel process are shown in Figure 1b. It can be seen that the pure ZrO_2 films remained amorphous state annealed at 400 ℃, and crystallize at 450 ℃. These results clearly suggested that the crystallization onset temperature of the $ZrTiO_x$ complex oxide films was delayed at least 200 ℃ with the incorporation of TiO_2.

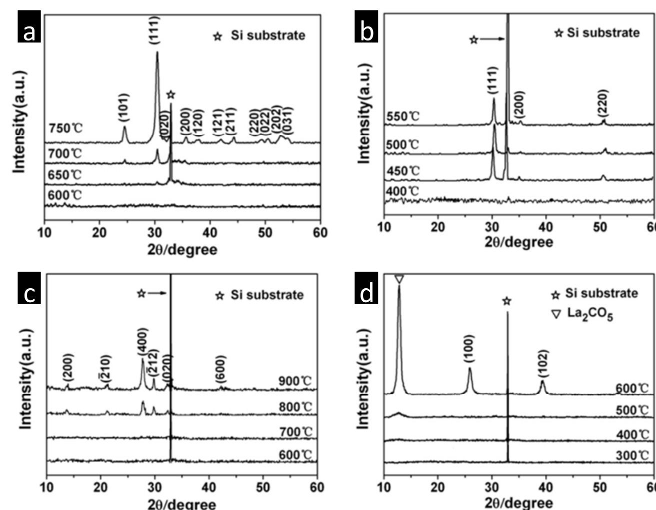

Figure 1. XRD patterns of (a) $ZrTiO_x$, (b) ZrO_x (c) $LaTiO_x$, and (d) La_2O_3 thin films at different annealing temperatures.

To demonstrate the universality of our strategy, we also prepared $LaTiO_x$ films with the addition of TiO_2. The XRD patterns in Figure 1c revealed the single $La_2Ti_2O_7$ phase formed with the crystallization temperature about 800 ℃. In contrast, La_2O_3 films with minor La_2CO_5 begin to crystallize between 500 to 600 ℃, as shown in Figure 1d. Therefore, the introduction of TiO_2 offers an effective route to delay the crystallization onset temperature of metal oxides. For oxide dielectrics used in OTFTs, amorphous oxide dielectrics are expected to avoid the usually large leakage current caused by the grain boundary of polycrystalline oxide, benefiting the high performance OTFT devices. The suppressed crystallization of oxide provided us novel routes to amorphous materials with potentially higher density, important for high *k* dielectric materials.

The amorphous state of $ZrTiO_x$ films is proved by the microstructure characterization. Figure 2a-c displays scanning electron micrograph (SEM) images of amorphous $ZrTiO_x$ films deposited on Si substrates. The surface of $ZrTiO_x$ films is quite smooth due to the amorphous state, regardless of different annealing temperatures of 400~600 ℃. The cross-section SEM image in the inset of Figure 2b shows the thickness of $ZrTiO_x$ film annealed at 500 ℃ is about 100 nm. Figure 2d-f shows the tapping mode atomic force micrograph (AFM) images of sol-gel $ZrTiO_x$ films annealed at 400-600 ℃. The scanning area is 3 μm×3 μm. As can been seen, all the $ZrTiO_x$ films exhibits a uniform and smooth surface without any surface defects and pinholes. The film root mean square (RMS) roughness decreased with the increase of annealing temperature. The RMS values of $ZrTiO_x$ films annealed at 400-600 ℃ are 0.34, 0.26, and 0.25 nm, respectively. These small RMS values are comparable

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to that of thermal SiO_2 (Rms~0.19 nm). The smooth surface and high quality $ZrTiO_x$ are beneficial for the charge carrier transport of organic semiconductors, since rough dielectric surface could induce the physical traps, or disturb the growth of organic semiconductors.[23]

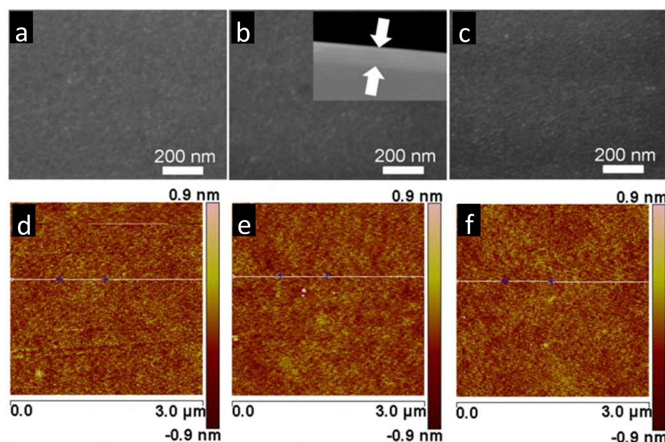

Figure 2. SEM images of ZrTiOx thin films at different annealing temperature of (a) 400, (b) 500, and (c) 600 ℃, and AFM images of $ZrTiO_x$ thin films at different annealing temperatures (d) 400, (e) 500, and (f) 600 ℃. The inset in Figure (b) is the cross-section image.

Dielectric properties of amorphous oxide dielectric films

To prove the high dielectric properties of the complex oxide films with the addition of TiO_2, we measured the capacitance for the $ZrTiO_x$ and $LaTiO_x$ film, as shown in Figure 3. The thickness of all

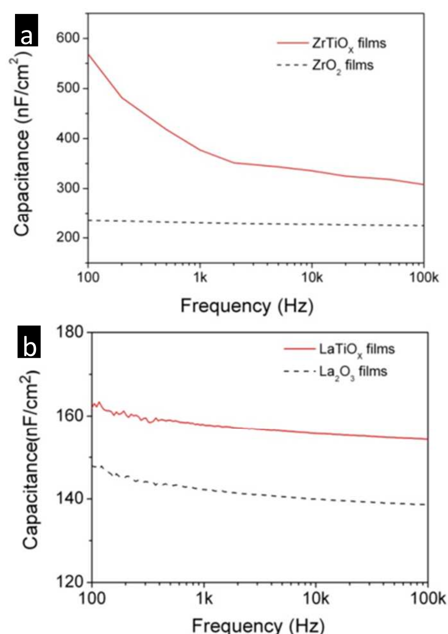

Figure 3. The capacitance of dielectric films (a) $ZrTiO_x$ and ZrO_2, (b) $LaTiO_x$ and La_2O_3.

the films annealed at 500 ℃ was controlled to be 100 nm. As shown in Figure 3a, the capacitance of pure ZrO_2 films is about 230 nF/cm^2 at the frequency range from 100 Hz to 100 KHz. With the addition of TiO_2, the capacitance of $ZrTiO_x$ increases dramatically, and the capacitance is as high as 560 nF/cm^2 at the low frequency of 100 Hz. At the high frequency, the capacitance decreases to about 350 nF/cm^2. This large dielectric dispersion of the ZrTiOx film at low frequency is similar to the previous results.[24, 25] At present the exact explanation for this large dielectric dispersion is not clear. It may be related to the charged structural defects, such as the oxygen vacancy $V_{\ddot{O}}$, which forman electric dipole and lead to the dielectric dispersion.

Since OTFTs are normally worked in low frequency circuits, this high capacitance value of the $ZrTiO_x$ dielectrics obtained at low frequency can meet the requirement of most OTFT applications.[26] For the La_2O_3 films, the addition of TiO_2 can also increase the capacitance from 148 to 162 nF/cm^2 at 100 Hz, as shown in Figure 3b. Among the various high-*k* oxides, titanium dioxide (TiO_2) is promising because of its high dielectric constant of approximately 80. However, its poor conduction band offsets with Si makes it difficult to act as an effective dielectric material. The improved capacitance for the $ZrTiO_x$ and $LaTiO_x$ films indicated that the complex oxide by the addition of TiO_2 is an effective strategy for the enhancement of dielectric properties.[21,23]

In the following part, we will focus on the $ZrTiO_x$ films to show high-performance amorphous oxide dielectric films for organic electronic applications. The dielectric properties of $ZrTiO_x$ films annealed at different temperatures were characterized by capacitance-frequency (C-f) curve, as shown in Figure 4a. The capacitance density of the $ZrTiO_x$ films decreases with the increasing frequency. At 1 KHz, the capacitance density of the $ZrTiO_x$ films annealed at 500 ℃ is about 380 nF/cm^2 while those annealed at 400 and 600 ℃ are 443 and 467 nF/cm^2, respectively. The effective *k* value is calculated by the following formula :

$$C_i = \varepsilon_0 \cdot k / t \quad (1)$$

where *k* is the dielectric constant, ε_0 is the permittivity in vacuum, and t is the thickness of a$ZrTiO_x$ film. The effective *k* values were calculated to be 50, 42, and 53 for $ZrTiO_x$ films annealed at 400, 500, and 600 ℃, respectively. These values are higher than or comparable to the previous $ZrTiO_x$ films. [21,23]

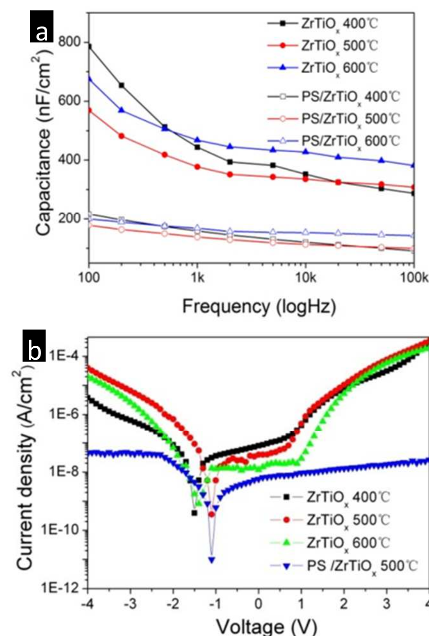

Figure 4. Dielectric characteristics (a) capacitance property and (b) leakage current of $ZrTiO_x$ thin films at different annealing temperatures.

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To improve the interface compatibility of amorphous oxide and organic active layers, $ZrTiO_x$ films were modified with 10 nm PS layer. Due to the low permittivity of PS layer (k ~2.5) and the increased thickness of $ZrTiO_x$/PS dielectric layer, the equivalent capacitance of the hybrid dielectrics layer decreases to 158, 137 and 168 nF/cm^2 at 1 KHz for $ZrTiO_x$ films annealed at 400, 500 and 600 ℃, respectively, as shown in Figure 4a. Although the total capacitance of PS/$ZrTiO_x$ hybrid dielectric films decrease, the remarkably decreased leakage current density was achieved. As shown in Figure 4b, the leakage current is about 5×10^{-6}A/cm^2 at -4 V for the $ZrTiO_x$ films annealed at 400 ℃, while it increased to be over 10^{-5}A/cm^2 for the $ZrTiO_x$ films annealed at 500 and 600 ℃. In contrast, with the modification of 10 nm PS layer, the leakage current of the 500 ℃ annealed $ZrTiO_x$ decreased to 4×10^{-8} A/cm^2, as shown in Figure 4b. The significantly decreased leakage current by the PS layer is highly desirable for high-performance OTFT devices.

OTFT applications of amorphous oxide dielectric films

To demonstrate the effectiveness of our high k amorphous complex oxide dielectrics, bottom-gate, top-contact pentacene-based OTFTs devices with silver source-drain electrode were fabricated with PS modified $ZrTiO_x$ dielectric films. The schematic structure is shown in the inset part of Figure 5a. All the device measurements were performed in ambient air. The output and transfer characteristics curves of OTFTs are shown in Figure 5. The V_G varies from 0 to -6V with a step of 1 V. All the devices have typical p-type OTFT characteristics with a linear region at small source-drain voltages (V_{DS}) and an excellent saturation region when the V_{DS} values near the gate voltages. In transfer characteristic curves, the V_{DS} was kept at −6V, the field-effect mobility (μ) in the saturation region is obtained by the following equation:

$$I_{ds} = \frac{\mu W C_i}{2L}(V_g - V_{th})^2 \qquad (2)$$

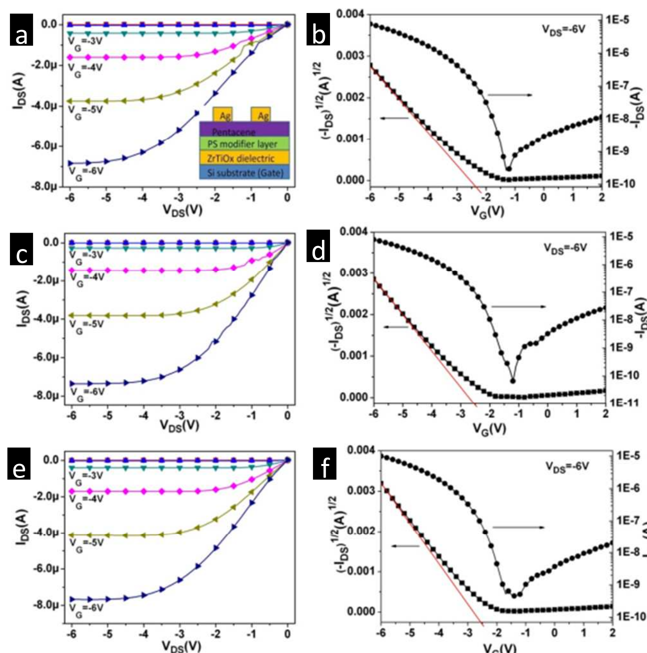

Figure 5. Electrical characteristics of OTFTs with the PS/$ZrTiO_x$ dielectric films at different annealing temperatures: (a) output curves at 400 ℃, (b) transfer curves at 400 ℃, (c) output curves at 500 ℃, (d) transfer curves at 500 ℃, (e) output curves at 600 ℃, (f) transfer curves at 600 ℃.

where I_{DS} is the drain-source current, C_i is the capacitance per unit area of the hybrid PS/$ZrTiO_x$ dielectric layer, and V_{th} is the threshold voltage. The threshold voltage of the devices is determined by the x-axis intercept of the tangent of $I_{DS}^{0.5}$ against V_G curve. Table 1 summarizes the dielectric properties, the corresponding capacitance per unit area and the electrical characteristics of pentacene OTFT devices with different PS/$ZrTiO_x$ dielectric layers. We have measured five devices for every annealing temperature. All the devices demonstrate the high on/off current ratio of 10^4. The highest field effect mobility of 0.58 cm^2/Vs was obtained for OTFTs with 500 ℃ annealed $ZrTiO_x$ dielectric layer.

Table 1. Electrical characteristics of OTFTs with the $ZrTiO_x$ dielectric films modified with PS.

Temperature (℃)	C_i (nF/cm^2)	k	Mobility (cm^{2}V^{-1}s^{-1})	$I_{on/off}$ ($\times10^4$)	V_{th} (V)
400	443	50±0.5	0.35±0.02	2.90	-2.27
500	380	42±0.3	0.47±0.03	5.57	-2.50
600	467	53±0.5	0.58±0.03	1.99	-2.54

The improved mobility of $ZrTiO_x$ dielectric layers with the annealing temperature can be attributed to the synthetic effect of the decreased surface roughness and the decreased traps densities. First, with the increase of the annealing temperature, the surface roughness of the $ZrTiO_x$ dielectric layer decreases from 0.34 to 0.25 nm, as shown in Figure 2. The decreased surface roughness of the dielectric layer leads to the increased grain size of the semiconductor, and the ordered growth of semiconductor.[27] The morphology and crystallite of the pentacene deposited onto the PS/$ZrTiO_x$ dielectric layer were investigated. As shown in Figure 6, all these pentacene films exhibit a dendritic structure with well-formed terraces. For $ZrTiO_x$ dielectrics annealed at 400 ℃, some small and round grain dispersed between large pentacene grains, and the average grain size of pentacene is about 1.2 μm. When the annealing temperature increased to 500 and 600 ℃, the average grain size of pentacene increased to 1.5 μm, with less amount of small grains marked with the red circles in Figure 6. As a result, the enhanced order and the reduced grain-boundary-induced scattering of pentacene will benefit the charge transport between the source and drain, and improve the performance of OTFTs. Moreover, the higher annealing temperature leads to a more dense structure and less defects of $ZrTiO_x$ dielectric layer, enhancing the charge carrier transport.[28]

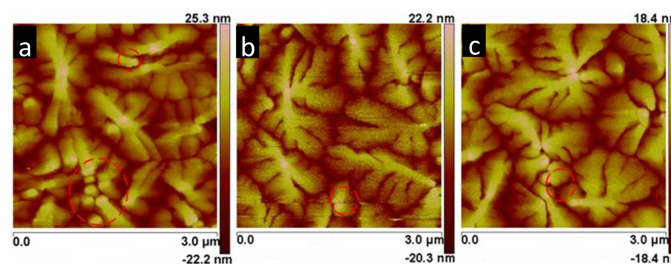

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Figure 6. AFM Surface morphology of the pentacene film deposited on the hybrid PS/$ZrTiO_x$ dielectric films at different annealing temperatures of (a) 400, (b) 500, and (c) 600 ℃.

Conclusions

In summary, a universal solution fabrication of high-*k* amorphous oxide dielectric layers was demonstrated for high-performance OTFTs. A high dielectric constant of 53 and high capacitance of 467 nF/cm^2 were achieved for the ZrTiOx films through simple control of annealing temperature. The interface modification of amorphous oxide dielectric films with polymer layer lead to the leakage current as low as 4×10^{-8} A/cm^2. As a result, the polymer modified high-*k* amorphous oxide dielectric films realize high-performance OTFTs, such as a high carrier mobility of 0.58 $cm^2V^{-1}s^{-1}$, on/off current ratio of 10^4 and low operating voltage of 6 V. Our simple solution preparation of high-*k* amorphous oxide dielectric films could provides novel routes for high-performance organic electronics, especially low-voltage organic transistor and memory devices.

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Notes and references

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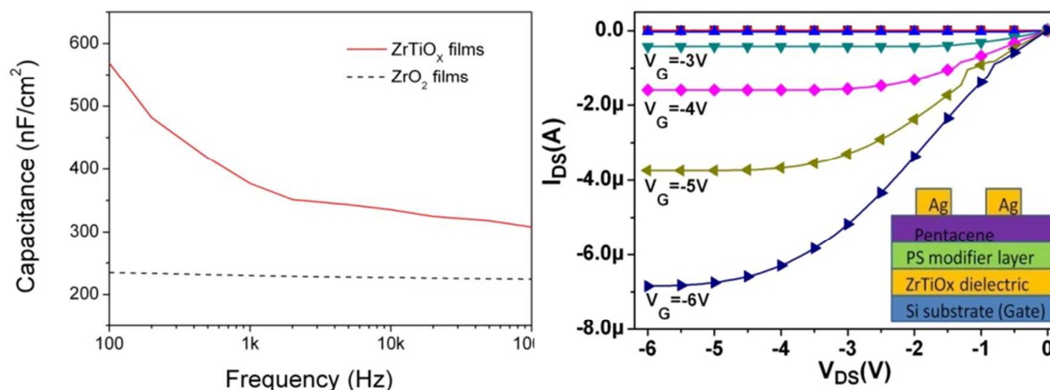

A facile and universal solution fabrication of high-*k* amorphous oxide dielectrics has been achieved for high-performance organic thin-film transistors (OTFTs). With $ZrTiO_x$ as a typical example, a *k* value and capacitance as high as 53 and 467 nF/cm^2, and a low leakage current of $4\times10^{-8} A/cm^2$ after polystyrene (PS) modification could be achieved. OTFTs with PS modified $ZrTiO_x$ films achieve a carrier mobility of 0.58 $cm^2V^{-1}s^{-1}$, an on/off current ratio of 10^4, and a low operating voltage of 6 V through simple control of annealing temperature of $ZrTiO_x$ films.

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