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Low leakage current gate dielectrics prepared by ion beam assisted deposition for organic thin film transistors

Chang Su Kim, Sung Jin Jo, Jong Bok Kim, Seung Yoon Ryu,
Joo Hyon Noh, and Hong Koo Baik^{a)}

Department of Materials Science and Engineering, Yonsei University, Seoul 120-749, Korea

Se Jong Lee

Department of Materials Science and Engineering, Kyungsoong University, Busan 608-736, Korea

Youn Sang Kim

Center for Intelligent Nano-Bio Materials and Division of Nano Science (BK21), Ewha Womans University, Seoul 120-750, Korea

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This communication reports on the fabrication of low operating voltage pentacene thin-film transistors with high- k gate dielectrics by ion beam assisted deposition (IBAD). These densely packed dielectric layers by IBAD show a much lower level of leakage current than those created by e -beam evaporation. These results, from the fact that those thin films deposited with low adatom mobility, have an open structure, consisting of spherical grains with pores in between, that acts as a significant path for leakage current. By contrast, our results demonstrate the potential to limit this leakage. The field effect mobility, on/off current ratio, and subthreshold slope obtained from pentacene thin-film transistors (TFTs) were $1.14 \text{ cm}^2/\text{V s}$, 10^5 , and 0.41 V/dec , respectively. Thus, the high- k gate dielectrics obtained by IBAD show promise in realizing low leakage current, low voltage, and high mobility pentacene TFTs. © 2007 American Institute of Physics.

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Organic thin-film transistors (OTFTs) based on conjugated polymers, oligomers, or other organic molecules have long been envisioned as a viable alternative to more traditional, mainstream TFTs based on inorganic materials. Potential applications include disposable radio frequency identification tags, large area gas sensors, photovoltaic cells, and low cost displays.¹⁻⁷ However, both organic and inorganic materials have been used as the dielectric layers for OTFTs and the choice of material has shown a significant impact on OTFTs performance. Some studies have suggested that inorganic materials with a high dielectric constant may be desirable not only for their higher current output, but also for the fact that they allow induction of a higher charge carrier concentration at the same applied gate voltage. Thus, various inorganic materials such as TiO_2 , Ta_2O_5 , yttria-stabilized zirconia have been evaluated as gate dielectrics for low operating voltage OTFTs.⁸⁻¹⁰

In recent years, the most significant advance in the vapor deposition technology has been the introduction of the ion beam assisted deposition (IBAD) method, namely, vacuum deposition with simultaneous ion bombardment.¹¹⁻¹³ This method produces many advantages over the conventional methods; in particular, high quality films can be obtained at lower growth temperatures, whereas the flux and the energy of ions can be changed independently of the flux of the depositing atoms, thereby allowing for easy control of the stoichiometry of compound films.

In previous work, we have reported on low operating voltage pentacene TFTs with $\text{CeO}_2\text{-SiO}_2$ gate dielectrics by

e -beam evaporation.¹⁴ However, the performance of such devices still requires further improvement. In this work, therefore, we controlled the packing density of the $\text{CeO}_2\text{-SiO}_2$ dielectric layers using IBAD in order to achieve better device performance in the pentacene TFTs.

Prior to the deposition of the dielectric layers on indium tin oxide (ITO) covered glass, this substrate was cleaned with acetone, ethanol, and deionized water in that order. The thickness and sheet resistance of the ITO films were 80 nm and $30 \text{ } \Omega/\text{sq}$, respectively. Subsequently, the 50 nm $\text{CeO}_2\text{-SiO}_2$ dielectric layers were deposited by e -beam evaporation and an IBAD process, respectively, using a 5 cm Kaufman source operating at 200 eV and processing an incident angle of 55° with respect to a normal substrate. An effective k value of 13-14 remained for the 50 nm $\text{CeO}_2\text{-SiO}_2$ dielectric layers regardless of the deposition method.¹⁵ The ion current density was measured by a Faraday cup mounted in front of the sample holder, whereas the deposition rate was maintained at $1 \text{ } \text{Å}/\text{s}$ by using a quartz-crystal monitor; the film thickness was fixed at 50 nm. The base pressure of the deposition chamber was 5×10^{-7} Torr, a value which increased to 3×10^{-4} Torr when Ar and O_2 flowed into the ion source. The O_2 flow rate was set at 15% of the Ar flow to maintain O stoichiometry in the $\text{CeO}_2\text{-SiO}_2$ dielectric layers during growth. A constant ion beam was generated for all depositions by setting the beam voltage to 200 eV, the discharge voltage to 40 eV and the acceleration voltage to 200 eV. An attractive alternative approach to modifying the dielectric surfaces would be to deposit a second layer, such as a spin-on polymer.^{16,17} Poly (4-vinylphenol) (PVP) film was then prepared by using solu-

^{a)}Electronic mail: thinfilm@yonsei.ac.kr

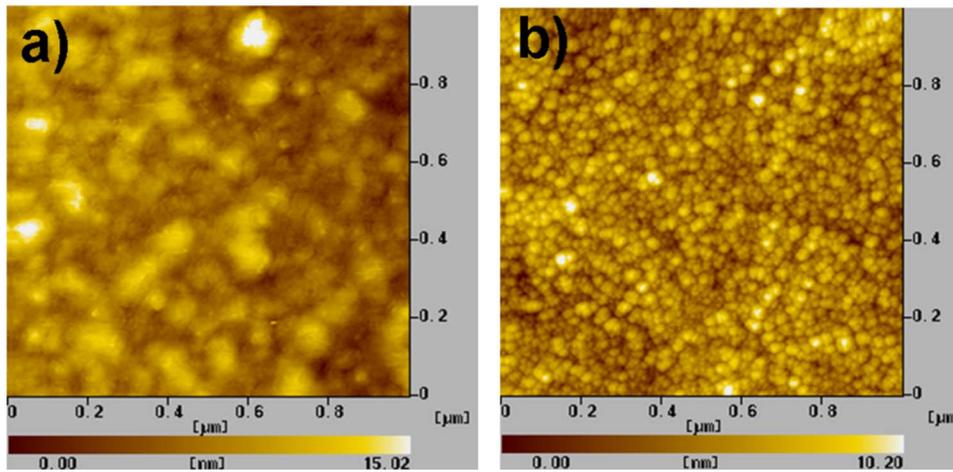


FIG. 1. (Color online) AFM images of the $\text{CeO}_2\text{-SiO}_2$ dielectric layers coated on the ITO glass by (a) *e*-beam evaporation and (b) IBAD, respectively.

tions of PVP and poly (melamine-co-formaldehyde), as a cross-linking agents, in the propylene glycol monomethyl ether acetate, by spin coating and subsequently cross linking (curing) at 175 °C for 1 h in a vacuum oven. The final thickness of the PVP film was approximately 45 nm as measured by the surface profiler. By applying the PVP coating, the reduced roughness induced uniform pentacene grains and OH groups on the $\text{CeO}_2\text{-SiO}_2$ are terminated as C_6H_5 , resulting in a larger hydrophobic surface. Enhanced pentacene quality and decreased hysteresis were observed in the current–voltage (I – V) measurements for the PVP-coated pentacene TFTs.¹⁸ For the hybrid double layer, an effective k value of 9.8 was also achieved. Pentacene (Sigma-Aldrich) channels and source/drain Au films were then sequentially patterned through shadow masks at a substrate temperature of 20 °C [room temperature] and by thermal evaporation in a vacuum chamber with a base pressure of 8.0×10^{-7} Torr. The deposition rates were fixed to 0.5 and 5 Å/s for the pentacene and Au electrodes, respectively, whereas the thicknesses of the pentacene and Au films were 50 and 100 nm, respectively, as monitored by the crystal quartz and confirmed by the surface profiler. Our device produced a nominal pentacene TFT channel length (L) of 150 μm and width (W) of 1000 μm .

All I – V measurements for our pentacene TFTs were performed with a semiconductor parameter analyzer (Agilent 4155C), whereas the current density–voltage (J – V) measurements of the $\text{CeO}_2\text{-SiO}_2$ dielectric layers were also evaluated with the Au/ $\text{CeO}_2\text{-SiO}_2$ /ITO structure. The refractive index for thin films deposited on the Si(100) substrates was measured at 633 nm by an L117 ellipsometer (Gaertner Scientific Corporation). The surface morphology of the films on the ITO was examined by atomic force microscopy (AFM) with a Digital Instruments NanoscopeII.

Figures 1(a) and 1(b) show the respective AFM images of the $\text{CeO}_2\text{-SiO}_2$ dielectric layers by *e*-beam evaporation and IBAD at the ion beam current density of 60 $\mu\text{A}/\text{cm}^2$. The $\text{CeO}_2\text{-SiO}_2$ dielectric layers in Fig. 1(a) indicate a loose microstructure consisting of large spherical particles with a root mean square (rms) of 1.4 nm, resulting from the low adatom mobility of the *e*-beam evaporation. However, in Fig. 1(b), ion bombardment induces many nucleation sites and enhances adatom mobility, thus producing a densely packed

structure composed of fine particles with a rms of 0.8 nm; the pore size becomes extremely small, in contrast to that of the spherical particles. Therefore, it is thought that the uniformly distributed small grains of the ion assisted films are attributed to the reduction in the island size and increase of the number of islands due to the ion bombardment.^{19–21}

Packing density dictates the porosity of thin films and is closely related to the relative refractive index value. Therefore, as ion bombardment during IBAD deposition promotes film densification, this increases the refractive index.^{22,23} Assuming that the pores within the films are accessible to and filled with molecules from the ambient atmosphere and setting the refractive index of the pores to 1, packing density can be simply calculated using the following equation:

$$n_f = pn_s + (1 - p)n_v, \quad (1)$$

where p , n_f , n_s , and n_v are the packing density and the index of the film, the solid part of the film (i.e., the grains or columns), and pores, respectively. Variations in the refractive index of the $\text{CeO}_2\text{-SiO}_2$ dielectric layers by *e*-beam evaporation and IBAD are displayed in Fig. 2(a). The $\text{CeO}_2\text{-SiO}_2$ dielectric layers produced *e*-beam evaporation show a refractive index of 1.88, whereas that produced during ion bombardment are 2.06. Therefore, the packing density of the $\text{CeO}_2\text{-SiO}_2$ dielectric layers produced IBAD increased up to 92% from 86% as obtained by *e*-beam evaporation, as presented in Fig. 2(a). As shown in Fig. 2(b), the leakage current density of the IBAD dielectric layers decrease by nearly one order of magnitude. This is related to the influence of film porosity on the leakage current as those thin films deposited with low adatom mobility have an open structure consisting of spherical grains, with the pores in between acting as a significant path for leakage current.^{24,25}

Field effect mobility was determined from the inset $\sqrt{I_D - V_G}$ curves of Fig. 3. The pentacene TFTs fabricated with IBAD $\text{CeO}_2\text{-SiO}_2$ dielectric layers exhibit a higher field mobility of 1.14 $\text{cm}^2/\text{V s}$ than those produced by *e*-beam evaporation dielectric layer, which also appears to have quite a good mobility of 0.91 $\text{cm}^2/\text{V s}$. According to the plotting of $\log_{10}(I_D) - V_G$ in Fig. 3, the on/off current ratios for the two devices with *e*-beam evaporation and IBAD $\text{CeO}_2\text{-SiO}_2$ dielectric layers were 10^4 and 10^5 , respectively, and their subthreshold slopes were 0.58 and 0.41

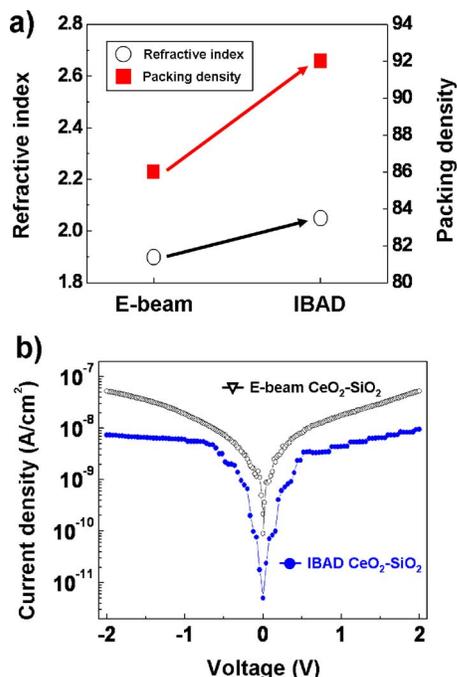


FIG. 2. (Color online) (a) The refractive index (left axis) and the packing density (right axis) of the CeO₂-SiO₂ dielectric layers by *e*-beam evaporation and IBAD. (b) Plots of the J - V characteristics of both dielectrics measured from the Au/dielectrics/ITO structure.

V/dec, respectively. Moreover, it is worth noting that the off-state I_D of the TFTs with the IBAD dielectric layer is a few times lower than that of the *e*-beam evaporation dielectric layer, reflecting the lower level of leakage current of the dielectric in Fig. 2(b). Thus, it is now assured that our dielectric scheme is very effective in fabricating high quality pentacene TFTs with exceptionally low leakage current at low voltages.

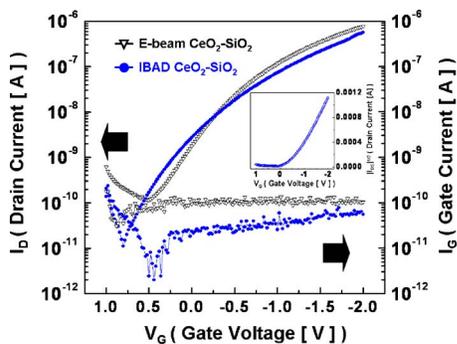


FIG. 3. (Color online) $\log_{10} I_D$ - V_G and $\sqrt{I_D}$ - V_G curves obtained from our pentacene TFTs with both dielectrics under a drain saturation condition ($V_D = -2$ V).

In summary, we have fabricated low operating voltage pentacene TFTs with CeO₂-SiO₂ dielectric layers by ion beam assisted deposition. These dielectric layers are readily deposited at room temperature and show a high capacitance with low a leakage current density. In particular, the field effect mobility, on/off current ratio, and subthreshold slope obtained from the pentacene TFTs were 1.14 cm²/V s, 10⁵, and 0.41 V/dec, respectively. We thus conclude that our IBAD CeO₂-SiO₂ dielectric layers show promise for realizing a low operating voltage pentacene TFTs.

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