

## Development of Organic-Inorganic Hybrid Dielectric for Organic Thin Film Transistors

Sunho Jeong, Dongjo Kim, Sul Lee, Bong-Kyun Park, and Jooho Moon\*

Department of Materials Science and Engineering

Yonsei University, Seoul 120-749, Korea

Phone: 82-2-2123-2855, E-mail: jmoon@yonsei.ac.kr

### Abstract

*Using a thermally-crosslinkable organosiloxane-based organic-inorganic hybrid material, solution-processable gate dielectric layer for organic thin-film transistors (OTFTs) have been fabricated. The hybrid dielectrics are synthesized by the sol-gel process, followed by the heat-treatment at 190 °C. To investigate the electrical property of hybrid dielectric, leakage current behavior and capacitance were measured. To fabricate coplanar-type OTFTs, Au/Cr electrode was deposited onto the heavily doped silicon substrate with the organic-inorganic hybrid dielectric layer and then  $\alpha,\omega$ -dihexylquaterthiophene was drop-cast between source and drain electrode. I-V character was measured to investigate an electrical performance of the fabricated transistor.*

### 1. Introduction

Organic thin-film transistors have recently received great attention because of their light weight, low-cost, flexibility, and easy processability. They are considered as potential candidates for a wide variety of applications such as large-area displays and low-end electronic devices like smart cards, electronic identification tags as well as large-area sensing devices [1-4]. However, besides the great advances that have made towards improvement of the OTFTs, there has been relatively little works on the gate dielectrics that critically determine the electrical performance of OTFTs. Most gate dielectrics reported so far are based on thermally grown silicon dioxide, PECVD deposited silicon nitride, sputtered aluminum oxide, or sputtered tantalum pentoxide and hence are usually brittle and expensive to prepare [5-10]. The poor mechanical properties of these materials make it highly challenging to be realistic in the flexible electronics. In addition, the preparation requires a high temperature vacuum process, which is not compatible with the plastic substrates. Consequently, it is necessary to develop a cheap and easy way (e.g.,

a solution-processable method) to fabricate gate insulators with requisite dielectric properties and flexibility.

In this letter, we report novel organosiloxane-based organic-inorganic hybrid dielectric materials synthesized by a sol-gel reaction. Since our materials are composed of a cross-linked network formed by hydrolysis and condensation reactions between alkoxides and alkyl(aryl)-substituted alkoxyisilane, the organic character that imparts low temperature- and solution-processabilities could be easily incorporated into the inorganic siloxane matrix of good dielectric stability.

### 2. Experiment

The precursor solutions of organic-inorganic hybrid dielectrics were started from a combination of methyltriethoxysilane and tetraethylorthosilicate. To initiate the hydrolysis and condensation reactions, DI-water and acid-catalyst were added in the mixed precursor solution. After the synthesized precursor solution was diluted, it was coated on a heavily doped silicon substrate using a spin-coating technique. The obtained films were pre-baked at 190 °C to evaporate the solvent and complete sol-gel reaction.

To fabricate coplanar-type transistors, Au/Cr electrodes were patterned by a shadow mask on top of the hybrid dielectric deposited on a heavily doped silicon substrate. The width and the length of the channel are 3 mm and 100  $\mu$ m; respectively.  $\alpha,\omega$ -dihexylquaterthiophene (DH4T, Syncom B.V.) was then deposited as an organic semiconductor between the source and drain electrodes by drop-casting a 0.8 wt% solution of DH4T in chlorobenzene. Figure 1 shows the schematic depiction and top view of the fabricated transistor.

All I-V and C-V measurements were performed in air using an Agilent 5263A source-measure unit and an Agilent 4284A precision LCR meter, respectively. All samples were dehydrated in a vacuum atmosphere

prior to the measurements. Atomic force microscopy (AFM, SPA 400, Seiko) was performed to investigate the surface morphology.

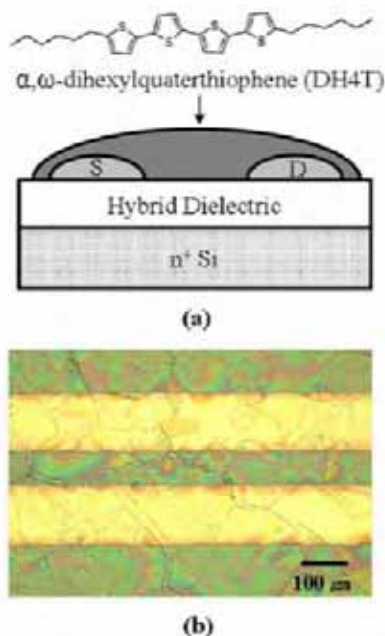


Fig. 1 (a) Schematic depiction and (b) optical microscopy image of the top view of the transistor fabricated using hybrid dielectric.

### 3. Results

It is observed that the dielectric strength of the hybrid dielectrics is about 1.65 MV/cm (defined as an electric field at the current density of  $10^{-6}$  A/cm<sup>2</sup>) and the capacitance of the hybrid dielectric is 13.5 nF/cm<sup>2</sup> at the frequency of 1 MHz, as shown in Fig. 2. The dielectric constant is calculated to 4.0, considering the thickness of the hybrid dielectric film is 260 nm.

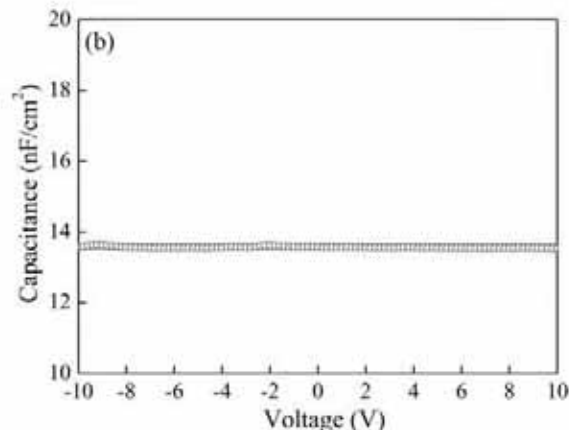
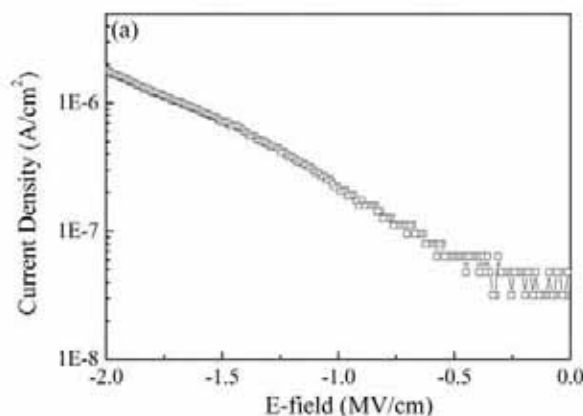


Fig. 2 (a)  $J$ - $E$  and (b)  $C$ - $V$  characteristic of the organic-inorganic hybrid dielectric material.

The roughness of a gate dielectric layer is a significant issue in the fabrication of OTFTs. Rough surface likely induces defect and voids at the semiconductor-dielectric interface during the deposition of the semiconducting layer and consequently the field-effect mobility could be lowered by the interfacial charge trap and poor molecular ordering of semiconductor. As shown in Fig. 3, the surface of the hybrid dielectric is very smooth, representing a rms surface roughness of 6 nm. In our sol-gel derived hybrid materials, the organic molecules are homogeneously incorporated into the inorganic matrix at the molecular-scale. Thus the films with the superior smoothness are easily achieved by simple coating methods such as spin-coating and dip-coating, while physical incorporation of inorganic particles in the organic matrix leads to form the film with rough surface [11].

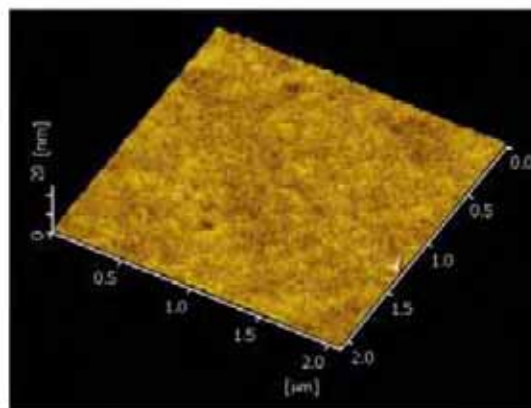


Fig. 3 Atomic force microscopy image of surface of the organic-inorganic hybrid dielectric film.

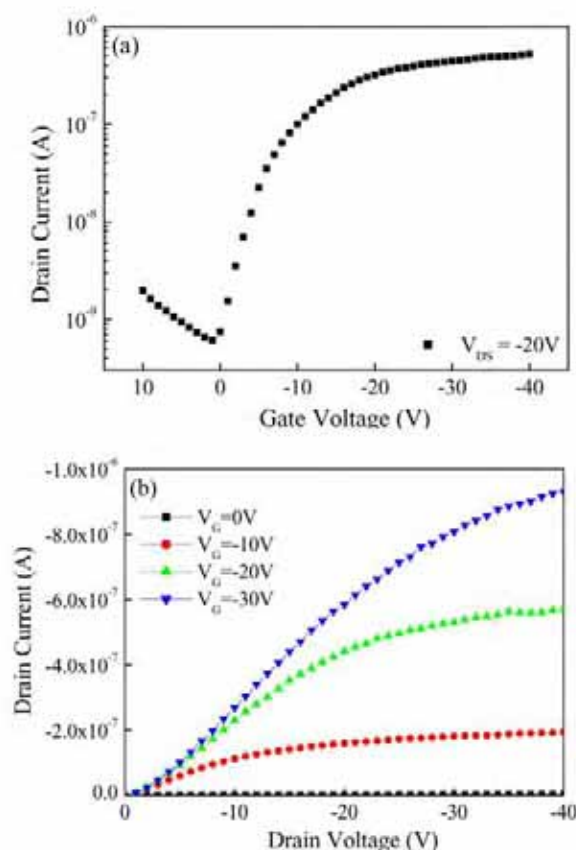


Fig. 4 Transfer (a) and output (b) characteristics of the organic thin-film transistor with a hybrid material as a dielectric layer.

Figure 4 shows the transfer and output characteristic of the transistor fabricated using the sol-gel hybrid film as a dielectric layer. The fabricated transistor displays the off-current of  $6 \cdot 10^{-10}$  A, field-effect mobility of  $5 \cdot 10^{-3}$   $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ , and threshold voltage of -0.3 V. The field-effect mobility and threshold voltage are calculated from the slopes and intercepts of plots at  $V_{DS} = -20\text{V}$ . It is interestingly observed that positively higher threshold voltage in our transistor, which might be attributed to the density of bulk trap states inside the hybrid dielectric. The silanol group can act as a bulk trap site, because the electrons injected from gate electrode are trapped under negative gate bias, forming the negative charge product ( $\text{SiO}^-$ ) [12]. The presence of the negative traps inside the dielectric layer would assist the hole easily accumulated at the interface between the gate dielectric and the semiconductor, inducing the positive shift of threshold voltage.

### 3. Conclusions

We have demonstrated the synthesis of organosiloxane-based gate dielectric materials. A sol-gel reaction between methyltriethoxysilane and tetraethylorthosilicate led to the formation of solution-processable dielectric materials. Using this dielectric material, high quality organic-inorganic hybrid films exhibiting a dielectric strength of 1.65 MV/cm and the capacitance of 13.5 nF/cm<sup>2</sup> were obtained at 190°C. It is observed that the roughness of hybrid dielectric formed by simple spin-coating method was  $6 \cdot 10^{-3}$  μm. The transistor fabricated using our hybrid dielectric and DH4T shows behavior of saturation and linear regime and it is considered that positively higher threshold voltage results from the bulk trap states due to silanol group.

### 4. Acknowledgements

This work was supported by the National Research Laboratory (NRL) Program from the Korea Science and Engineering Foundation.

### 5. References

- [1] C. D. Dimitrakopoulos, D. J. Masearo, IBM J. Res. Dev. 45, 11 (2001).
- [2] H. Edzer, A. Huitema, G. H. Gelinck, J. Bas, P. H. Van der Putten, K. E. Kuijk, K.M. Hart, E. Cantatore, and D. M. De Leeuw, Adv. Mater. (Weinheim, Ger.) 14, 1201 (2002).
- [3] B. Crone, A. Dodabalapur, Y. Y. Lin, R. W. Filas, A. Bao, A. Laduca, R. Sarpeshkar, H. E. Katz and W. Li, Nature (London) 403, 521 (2000).
- [4] B. K. Crone, A. Dodabalapur, R. Sarpeshkar, A. Gelperin, H.E. Katz, and Z. N. Bao, J. Appl. Phys. 91, 10140 (2002).
- [5] D. Knipp, R. A. Street, A. Vokel, and J. Ho, J. Appl. Phys. 93, 347 (2003).
- [6] R. A. Street, D. Knipp, and A. R. Vokel, Appl. Phys. Lett. 80, 1658 (2002).
- [7] G. Wang, D. Moses, A. Heeger, H. Zhang, M. Naransimhan, and R. E. Demaray, J. Appl. Phys. 95, 316 (2004).
- [8] J. Wang, X. Yan, Y. Xu, J. Zhang, and D. Yan, Appl. Phys. Lett. 85, 5424 (2004).
- [9] J. Lee, J. H. Kim, and S. Im, J. Appl. Phys. 95, 3733 (2004).

[10] A. Facchetti, M.-H. Yoon, and T. J. Marks, *Adv. Mater.* 17, 1705 (2005).

[11] L. Chua, J. Zaumseil, J. Chang, E.W. Ou, P.H. Ho, H.

Sirringhaus, and R. Friend, *Nature* 434, 194 (2005).

[12] F. Chen, C. Chu, J. He, Y. Yang, and J. Lin: *Appl. Phys. Lett.* 85, 3295 (2004).