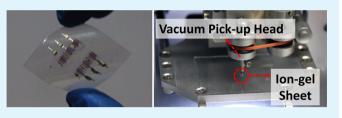
Low-Voltage Large-Current Ion Gel Gated Polymer Transistors Fabricated by a "Cut and Bond" Process

Xianyi Shao,[†] Bei Bao,[‡] Jiaqing Zhao,[†] Wei Tang,[†] Shun Wang,^{*,‡} and Xiaojun Guo^{*,†}

[†]Department of Electronic Engineering, and [‡]Key Laboratory of Artificial Structures and Quantum Control (Ministry of Education), Department of Physics and Astronomy, Shanghai Jiao Tong University, Shanghai 200240, P. R. China

ABSTRACT: A "cut and bond" process using a commercial die bonder was developed for fabricating ion gel gated organic thin-film transistors (OTFTs). It addresses the issues of damaging or contaminating the channel layer when depositing the ion gel layer on top in conventional fabrication processes. The formed isolated dielectric regions can help to eliminate possible lateral electric field coupling through the dielectric layer when several devices are integrated to construct



functional circuits. The fabricated OTFTs provide mA-level ON current, and an ON/OFF current ratio higher than 10^5 with the gate swing voltage of less than 3 V. With the developed process, the ion gel OTFTs are integrated with inorganic light emitting diodes (LEDs) of different colors on plastic substrate using the same die bonder, and the light emission of the LEDs can be modulated in a wide range from dark to high brightness with change of the gate voltage less than 3 V.

KEYWORDS: organic thin film transistor, ion gel, low voltage, large current, light emitting diode

INTRODUCTION

Organic thin film transistors (OTFTs) are promising for low cost and flexible electronics applications owing to their advantages such as compatibility with high throughput printing or coating processes, excellent mechanical flexibility, and so on.^{1–6} In the past decade significant progresses have been achieved for OTFTs with soluble organic semiconductor materials, device technologies, and circuit integration.^{7–10} However, OTFTs still suffer high operation voltage and low current driving capability, which limit OTFTs for many practical applications.

To reduce the operation voltage and improve the current driving capability, an effective approach is enlarging the gate dielectric capacitance with an ultrathin or high-dielectricconstant (high-k) gate dielectric layer.¹¹⁻¹⁵ However, an ultrathin dielectric layer is difficult to be processed via simple printing or coating processes over large area and may also present severe reliability issues. Using a high-k dielectric layer would be a better choice, but there is lack of proper dielectric materials of large enough k value to be compatible with the solution processes for OTFTs and induce enough charges for large driving current. Recently, a type of solid polymer electrolyte, so-called ion gel, has been used as gate dielectric for OTFTs.^{16,17} The ion gel is obtained by blending ionic liquids with a gelating triblock copolymer to form a physically cross-linked network and features very large specific capacitance exceeding 1 μ F/cm² with thickness of about 1 μ m, so that it can help to substantially reduce the operation voltage and simultaneously achieve very high ON currents.¹⁸ Attributed to these features, the ion gel OTFTs were able to drive organic light emitting diodes (OLEDs) at low operating voltages and extremely small device dimensions with respect to the OLED

active area.¹⁹ Although the ion gel layer can be processed by spin coating or printing from solvents, the processes might damage or contaminate the channel layer for a top gate structure transistor.²⁰ To address this issue, an approach was developed by cutting the formed free-standing ion gel film of high tensile strength and laminating it on the organic semiconductor layer using tweezers.²¹ This manual approach, however, has several limitations in process control for device manufacturing.

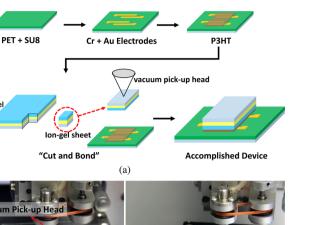
In this work, a "cut and bond" process is developed for ion gel gated OTFT fabrication by using a commercially available die bonder, which enables accurate control of alignment and pressure during the lamination process. With the developed process, the ion gel gated OTFTs can be integrated with red, green, and blue inorganic light emitting diodes (LEDs) on plastic substrate by using the same die bonder. It was demonstrated that the light emission of the LEDs can be modulated in a wide range from dark to high brightness with a small gate voltage swing less than 3 V.

EXPERIMENTAL SECTION

The fabrication processes and the cross-sectional structure of the ion gel gated OTFT are illustrated in Figure 1a. A 125 μ m thick poly(ethylene terephthalate) (PET) film was used as the substrate. A 90 nm thick SU8 film was deposited by spin coating as the smoothing layer before the following processes. A stack of 4 nm thick chromium (Cr) and 22 nm thick gold (Au) was thermally evaporated with a shadow mask to define the channel length and width of 60 and 1200 μ m, respectively. The channel layer of about 32 nm thickness was

Received:December 2, 2014Accepted:January 23, 2015Published:January 23, 2015

ITO <



TET Channel Re

(b)

Ion-gel Sheet

Figure 1. (a) Illustration of the cross-sectional structure and the fabrication processes of the top gate bottom contact ion gel OTFT. (b) Photographic images of the cut and bond process for laminating ion gel/ITO/PET film on the channel region with the help of the facility "die bonder".

formed by spin-coating of a poly(3-hexylthiophene) (P3HT) (BASF, SepiolidTM P200) solution in chloroform (CHCl₃) with a concentration of 5 mg/mL in a N₂ filled glovebox, followed by an annealing process at 120 °C for 30 min. The P3HT film out of the channel region was carefully wiped off with cotton sticks.

To form the ion gel, poly(vinylidene fluoride-*co*-hexafluoropropylene) (P(VDF-HFP)) and 1-ethyl-3-methylimidazolium bis-(trifluoromethylsulfonyl)imide ([EMIM][TFSI]) ionic liquid were dissolved in acetone with a weight ratio of 1:4:7. The ion gel was spincoated onto an ITO coated PET film, forming an about 1 μ m thick film after being heated at 70 °C for 24 h in a N₂ filled glovebox to remove the residual solvent.

A cut and bond process was developed to selectively deposit the ion gel film onto the P3HT channel for each device. As illustrated in Figure 1a, the prefabricated ion gel/ITO/PET film was cut into small size sheets with a paper cutter. The vacuum pick-up head in a commercial die bonder machine (CaiNa, RF500) was used to pick up the small sheets. The machine positioned these picked sheets above the proper device region with a micropositioner and then released and laminated them onto the channel layer with a baking process at 80 °C for 15 min in atmosphere. With the well automatically controlled processes, these small ion gel sheets can be placed over the channel regions of different devices very accurately and consistently to form isolated dielectric regions. The snapshot photos of the die bonder during cut and bond process are given in Figure 1b, with a supplementary video provided. LED chips were also bonded on to substrate using the same die bonder. Peripheral interconnects to connect the LEDs and the transistors were formed by dispensing silver conductive epoxy with a robot dispenser (Pioneer, PTC RD331).

The ITO/ion gel/ITO capacitor test structure was characterized using a SOLARTRON 1260 impedance analyzer with the frequency from 1 Hz to 1 MHz. The electrical characteristics of the fabricated transistors were measured with a Keithley 4200 semiconductor parameter analyzer.

RESULTS AND DISCUSSION

The measured specific capacitance of the ITO/ion gel/ITO test structure film as a function of frequency is inserted in Figure 2a. In the low-frequency range, the specific capacitance is about 3.1 μ F/cm², which is a typical value for ion gel dielectric capacitor.¹⁵ With such a large specific capacitance, low-voltage

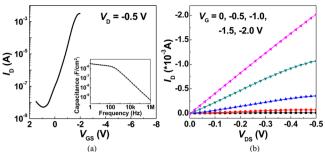


Figure 2. Measured-representative (a) transfer characteristics (I_D vs V_{GS}) and (b) output characteristics (I_D vs V_{DS}) of fabricated OTFTs with ion gel gate insulator. Inset shows the frequency dependence of the specific capacitance of ion gel film based on P(VDF-HFP) and [EMIM][TFSI].

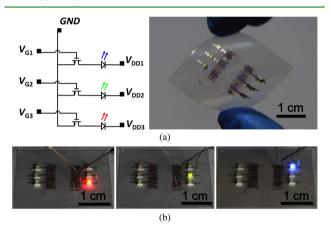


Figure 3. (a) Circuit diagram of this LED driving circuit and the images of the completed flexible LED driving circuit with ion gel OTFT devices. (b) Red, green, and blue LEDs were lightened with supply voltages (V_{DD}) of 2.2, 2, and 3 V, respectively.

and large-current OTFTs were achieved. The measured transfer $(I_{\rm D}-V_{\rm GS})$ and output $(I_{\rm D}-V_{\rm DS})$ characteristics are shown in Figure 2 a and b, respectively. With a gate voltage swing of less than 3 V, the device can be switched from the OFF state to the ON state with an ON/OFF current ratio higher than 10⁵, and ON current reaching the level of mA. The extracted subthreshold swing of the device is about 360 mV/decade, which is larger than the value of about 250 mV/decade in previous work.²¹ The field effect property should be able to be further improved by optimizing the material preparation and film coating processes or choosing different semiconductor or ion gel materials. The developed cut and bond process, without concern of damaging or contaminating the channel layer when depositing the ion gel layer, would provide more freedom to try different types of semiconductor and ion gel materials for this purpose.

The large gate dielectric specific capacitance helps to achieve large current and low operation voltage but might also induce lateral electrical field coupling through the dielectric layer. As a result, when several devices are integrated together, one device's operation could influence the neighboring devices. With the developed cut and bond process, isolated dielectric regions are formed for individual devices to suppress the electric field coupling.

As a simple demonstration, OTFT driven LEDs in different color (red, green, blue) were fabricated on PET substrate, with the circuit diagram and the photo of the fabricated sample given

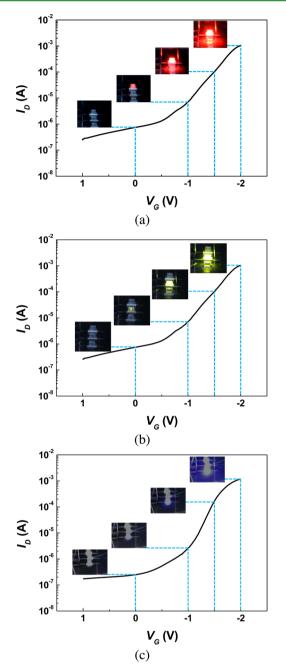


Figure 4. Measured-representative I-V curve of driving (a) red, (b) green, and (c) blue inorganic LEDs. Insets are photographs of the LEDs under different voltages of 0, -1, -1.5, and -2 V.

in Figure 3a. The brightness of each LED is modulated by changing the gate voltage of its driving OTFT. Figure 3b shows that with the driving OTFTs of isolated dielectric regions, the operation of each OTFT driven LED cell will not affect others. This will be even more important for high density integration of OTFTs.

As shown in Figure 4, the light emission of the red, green, and blue LEDs can be modulated in a wide range from dark to high brightness with change of the gate voltage from 1 V to -2 V and supply voltage ($V_{\rm DD}$) of 2.2, 2, and 3 V, respectively, which are determined by the different driving voltage requirements of the different color LEDs. With the large driving current to the level of mA and low voltage modulation, this ion gel gated OTFT is promising for driving the inorganic

LEDs, which has never been achieved by other OTFT technologies.

CONCLUSION

In this work, a cut and bond process with a commercial die bonder was developed for fabricating ion gel gated OTFTs. It addresses the issues of damaging or contaminating the channel layer when depositing the ion gel layer on top in conventional fabrication processes. The formed isolated dielectric regions can help to eliminate the possible lateral electric field coupling through the dielectric layer when several devices are integrated to construct functional circuits. The fabricated OTFTs provide mA-level ON current and an ON/OFF current ratio higher than 10⁵ with the gate swing voltage of less than 3 V. Referring to the previous work, the field effect property should be able to be further improved by optimizing the material preparation and film coating processes or choosing different materials. The developed cut and bond process could provide more freedom to try different types of semiconductor and ion gel materials for this purpose. As an application example, inorganic LEDs in different colors were integrated with the OTFTs using the same die bonder on plastic substrate. It is shown that, with the large driving current and low voltage modulation, this ion gel gated OTFT is promising for driving the inorganic LEDs, which has never been achieved by other OTFT technologies. With the manufacturing base in microelectronics industry on semiconductor wafer dicing and die bonding, the cut and bond process would be capable of being scaled up to achieve largearea and high-density OTFT integration for many large-current and low-voltage plastic electronics applications.

AUTHOR INFORMATION

Corresponding Authors

*E-mail: x.guo@sjtu.edu.cn.

*E-mail: shunwang@sjtu.edu.cn.

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Funding

The NSFC of China (Grant No. 61274083, 61334008), 863 Program (Grant No. 2014AA032702), Ph.D. Programs Foundation of Ministry of Education of China (20120073110093) is ackowledged.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the NSFC of China (Grant No. 61274083, 61334008), 863 Program (Grant No. 2014AA032702), Ph.D. Programs Foundation of Ministry of Education of China (20120073110093).

REFERENCES

(1) Noh, Y.-Y.; Zhao, N.; Caironi, M.; Sirringhaus, H. Downscaling of Self-Aligned, All-Printed Polymer Thin-Film Transistors. *Nat. Nanotechnol.* **2007**, *2*, 784–789.

(2) Berggren, M.; Nilsson, D.; Robinson, N. D. Organic Materials for Printed Electronics. *Nat. Mater.* **2007**, *6*, 3–5.

(3) Xia, Y.; Friend, R. H. Nonlithographic Patterning through Inkjet Printing via Holes. *Appl. Phys. Lett.* **200**7, *90*, 253513–253513–3.

ACS Applied Materials & Interfaces

(4) Liu, Y.; Cui, T.; Varahramyan, K. All-Polymer Capacitor Fabricated with Inkjet Printing Technique. *Solid-State Electron.* **2003**, *47*, 1543–1548.

(5) Crone, B.; Dodabalapur, A.; Lin, Y.-Y.; Filas, R.; Bao, Z.; LaDuca, A.; Sarpeshkar, R.; Katz, H.; Li, W. Large-Scale Complementary Integrated Circuits Based on Organic Transistors. *Nature* **2000**, *403*, 521–523.

(6) Guo, X.; Feng, L.; Cui, Q.; Xu, X. Low Voltage Organic/ Inorganic Hybrid Complementary Inverter with Low Temperature All Solution Processed Semiconductor and Dielectric Layers. *IEEE Electron Device Lett.* **2014**, 35, 542–544.

(7) Feng, L.; Tang, W.; Xu, X.; Cui, Q.; Guo, X. Ultralow-Voltage Solution-Processed Organic Transistors with Small Gate Dielectric Capacitance. *IEEE Electron Device Lett.* **2013**, *34*, 129–131.

(8) Wöbkenberg, P. H.; Ball, J.; Kooistra, F. B.; Hummelen, J. C.; de Leeuw, D. M.; Bradley, D. D.; Anthopoulos, T. D. Low-Voltage Organic Transistors Based on Solution Processed Semiconductors and Self-Assembled Monolayer Gate Dielectrics. *Appl. Phys. Lett.* **2008**, *93*, 013303–013303–3.

(9) Dimitrakopoulos, C. D.; Malenfant, P. R. Organic Thin Film Transistors for Large Area Electronics. *Adv. Mater.* 2002, *14*, 99–117.

(10) Halik, M.; Klauk, H.; Zschieschang, U.; Schmid, G.; Dehm, C.; Schütz, M.; Maisch, S.; Effenberger, F.; Brunnbauer, M.; Stellacci, F. Low-Voltage Organic Transistors with An Amorphous Molecular Gate Dielectric. *Nature* **2004**, *431*, 963–966.

(11) Cheng, X.; Caironi, M.; Noh, Y.-Y.; Wang, J.; Newman, C.; Yan, H.; Facchetti, A.; Sirringhaus, H. Air Stable Cross-Linked Cytop Ultrathin Gate Dielectric for High Yield Low-Voltage Top-Gate Organic Field-Effect Transistors. *Chem. Mater.* **2010**, *22*, 1559–1566.

(12) Jang, Y.; Kim, D. H.; Park, Y. D.; Cho, J. H.; Hwang, M.; Cho, K. Low-Voltage and High-Field-Effect Mobility Organic Transistors with a Polymer Insulator. *Appl. Phys. Lett.* **2006**, *88*, 072101–072101–3.

(13) Li, J.; Sun, Z.; Yan, F. Solution Processable Lowvoltage Organic Thin Film Transistors with High-k Relaxor Ferroelectric Polymer as Gate Insulator. *Adv. Mater.* **2012**, *24*, 88–93.

(14) Zirkl, M.; Haase, A.; Fian, A.; Schön, H.; Sommer, C.; Jakopic, G.; Leising, G.; Stadlober, B.; Graz, I.; Gaar, N. Low-Voltage Organic Thin-Film Transistors with High–k Nanocomposite Gate Dielectrics for Flexible Electronics and Optothermal Sensors. *Adv. Mater.* **2007**, *19*, 2241–2245.

(15) Facchetti, A.; Yoon, M. H.; Marks, T. J. Gate Dielectrics for Organic Field-Effect Transistors: New Opportunities for Organic Electronics. *Adv. Mater.* **2005**, *17*, 1705–1725.

(16) Cho, J. H.; Lee, J.; Xia, Y.; Kim, B.; He, Y.; Renn, M. J.; Lodge, T. P.; Frisbie, C. D. Printable Ion-Gel Gate Dielectrics for Low-Voltage Polymer Thin-Film Transistors on Plastic. *Nat. Mater.* **2008**, *7*, 900–906.

(17) Zhang, S.; Lee, K. H.; Sun, J.; Frisbie, C. D.; Lodge, T. P. Viscoelastic Properties, Ionic Conductivity, and Materials Design Considerations for Poly(styrene-*b*-ethylene oxide-*b*-styrene)-Based Ion Gel Electrolytes. *Macromolecules* **2011**, *44*, 8981–8989.

(18) Lee, J.; Panzer, M. J.; He, Y.; Lodge, T. P.; Frisbie, C. D. Ion Gel Gated Polymer Thin-Film Transistors. J. Am. Chem. Soc. 2007, 129, 4532–4533.

(19) Braga, D.; Erickson, N. C.; Renn, M. J.; Holmes, R. J.; Frisbie, C. D. High-Transconductance Organic Thin-Film Electrochemical Transistors for Driving Low-Voltage Red–Green–Blue Active Matrix Organic Light-Emitting Devices. *Adv. Funct. Mater.* **2012**, *22*, 1623–1631.

(20) Lee, K. H.; Zhang, S.; Lodge, T. P.; Frisbie, C. D. Electrical Impedance of Spin-Coatable Ion Gel Films. J. Phys. Chem. B 2011, 115, 3315–3321.

(21) Lee, K. H.; Kang, M. S.; Zhang, S.; Gu, Y.; Lodge, T. P.; Frisbie, C. D. Cut and Stick Rubbery Ion Gels as High Capacitance Gate Dielectrics. *Adv. Mater.* **2012**, *24*, 4457–4462.