

# RSC Advances



This is an *Accepted Manuscript*, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

*Accepted Manuscripts* are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. This *Accepted Manuscript* will be replaced by the edited, formatted and paginated article as soon as this is available.

You can find more information about *Accepted Manuscripts* in the [Information for Authors](#).

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard [Terms & Conditions](#) and the [Ethical guidelines](#) still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this *Accepted Manuscript* or any consequences arising from the use of any information it contains.

Cite this: DOI: 10.1039/c0xx00000x

www.rsc.org/xxxxxx

ARTICLE TYPE

# Printable Poly(methyl silsesquioxane) Dielectric Ink and Its Application in All Solution Processed Metal Oxide Thin-Film Transistors

Xinzhou Wu, Zheng Chen\*, Teng Zhou, Shuangshuang Shao, Meilan Xie, Minshun Song, Zheng Cui\*

Received (in XXX, XXX) Xth XXXXXXXXX 20XX, Accepted Xth XXXXXXXXX 20XX

DOI: 10.1039/b000000x

Thermally cross-linkable poly(methyl silsesquioxane) (PMSQ) has been investigated as a printable dielectric ink to make the gate insulator by aerosol jet printing for solution processed metal oxide (IGZO) thin-film transistors. It was found that with the increase of curing temperatures from 150 to 200 °C the dielectric constant and loss tangent of printed PMSQ layer reduces dramatically. The mobility, leakage current and gate current of PMSQ enabled thin-film transistor reduces accordingly, while the on/off ratio increases with the increase of curing temperature. An interfacial layer is introduced to further improve the on/off ratio to  $3 \times 10^5$  and reduce the leakage current to  $2.6 \times 10^{-10}$  A, which is the best result for solution processed IGZO thin-film transistors using the PMSQ as gate insulator at the curing temperature of only 150 °C. The work has demonstrated the feasibility of fabricating IGZO thin-film transistors by all solution process.

## Introduction

Printable conductors, semiconductors, and dielectric materials are all crucial to the fabrication of large area, flexible and low cost electronics.<sup>1-6</sup> Considerable progress has been made in printable semiconductors for thin film transistors (TFTs) and the mobility has been achieved within the last few years to a level comparable to that of hydrogenated amorphous silicon transistors.<sup>7-9</sup> Recently, printable gate dielectrics (polyvinyl phenol<sup>10-15</sup> and polyimide<sup>16</sup>, etc) have attracted much attention, as it is well known that the electrical performances of transistors are crucially depend on the properties of gate dielectrics. However, most of accessible solution type of gate dielectrics such as poly(methylmethacrylate) (PMMA) have relatively poor chemical resistance to organic solvent, which greatly restrict their application in solution processing of TFTs. On the other hand, some thermally curable polymer dielectrics such as solution type of polyimide (PI)<sup>17</sup> and polyvinyl phenol (PVP)<sup>18</sup> require much too high curing temperatures (>200 °C) to allow for the use of conventional cheap plastic substrates whose glass transition temperatures are usually lower than 150 °C. Besides, additional cross-linking agents are required for the cross-linking reactions. In contrast, the poly(methyl silsesquioxane) (PMSQ) has emerged recently as the most attractive organic-inorganic hybrid dielectrics.<sup>8, 9, 17, 19, 20</sup> Firstly, compact PMSQ films can be obtained at curing temperature as low as 150 °C through polycondensation reactions of Si-OH groups without any cross-linking agents; Secondly, PMSQ solution can be readily synthesized by a sol-gel method without poisonous and harmful chemicals; Thirdly, the surface morphology and dielectric properties of PMSQ films can be easily tuned by altering their organic side groups. Finally, PMSQ is chemically resistant to common organic solvents (such as methylbenzene, alcohol,

chloroform, etc), making it stable during solution deposition of organic semiconductors.

Though PMSQ has been widely used in solution processed organic thin-film transistors (OTFTs), these transistors suffered from high off current, leading to low on/off ratios. In 2008, Yamazaki and his coworkers<sup>17</sup> fabricated poly(3-hexylthiophene)-based (P3HT) field effect transistors, using the PMSQ as the gate insulators cured at 150 °C. The fabricated OTFTs exhibited on/off ratio less than  $10^3$  and off current of  $10^{-8}$  A. It was suggested that the high off current was caused by residual silanol groups due to incomplete polycondensation of PMSQ<sup>21</sup>. It was reduced to  $6 \times 10^{-10}$  A in another work by increasing the curing temperature from 150 to 190 °C in  $\alpha,\omega$ -dihexylquaterthiophene (DH4T)-based OTFTs<sup>22</sup>. Nagase and his coworkers<sup>9</sup> systematically investigated the synthesis condition of PMSQ solution. They found that PMSQ with low silanol concentration could be obtained by using PGMEA as the solvent during synthesis and the polycondensation reaction could take place at 70 °C temperature. They managed to achieve on/off ratio of  $10^4$  and leakage current of  $3 \times 10^{-9}$  A in their P3HT-based TFTs using PMSQ as gate insulator. Though improved, these values are still not good enough for practical use of printed TFTs.

While solution type of organic semiconductors still suffer from low mobility and environmental instability issues, inorganic semiconductors, particularly metal oxide semiconductors such as Indium-gallium-zinc oxide (IGZO) have found their place in TFTs due to their high mobility, high environmental stability, good uniformity and low cost. They have already been used to make backplane driving electronics in flat-panel display applications<sup>23</sup>. High performance IGZO TFTs can be fabricated either through vacuum sputtering deposition or by solution/printing deposition process<sup>24-30</sup>. For solution processing of metal oxide TFTs, high performance gate dielectrics is needed,

which should ideally be deposited by printing or solution process. In this work, we investigated the electrical properties of PMSQ film under different curing temperatures and applied the dielectric films as gate insulators in IGZO thin-film transistors by solution process. An interfacial passivation layer was introduced, prior to solution deposition of PMSQ, which improved the on/off ratio to  $3 \times 10^5$  and reduced the leakage current to  $2.6 \times 10^{-10}$  A. This is the best result using solution deposited PMSQ as gate insulator with low curing temperature at 150 °C. Top-gate and top-contact IGZO TFTs were also fabricated by printing PMSQ as the gate insulator, instead of spin coating, and the devices showed on current up to  $3.4 \times 10^{-5}$  A, effective mobility up to  $0.75 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ , leakage current  $1.66 \times 10^{-9}$  A and on/off ratio of  $2 \times 10^4$ .

## Experimental Section

### Materials

$\text{In}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{Ga}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  and  $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  were purchased from Acros. 3-glycidioxypropyltrimethoxysilane (GPTMS), formic acid and propylene glycol monomethyl ether acetate (PGMEA) were purchased from Sinopharm Chemical Reagent Co., Ltd. All solvents were analytical grade and used without further purification.

### Synthesis of PMSQ ink and deposition of PMSQ films

Poly(methylsilsequioxane) (PMSQ) solution was synthesized according to the previously reported procedure.<sup>9</sup> Deionized water with an acid catalyst was slowly added to the methyltrimethoxysilane PGMEA solution. The mixture was stirred for 30 min at room temperature and then heated up to 70 °C for 1 h to initiate polycondensation reaction. Highly viscous PMSQ solution was obtained after vacuum distillation at 70 °C to remove small molecules in solution and then diluted with PGMEA to designated concentration (0.45 g/mL). For preparation of PMSQ ink, the viscous PMSQ was dissolved in acetone to the concentration of 15 mg/ml. PMSQ films are formed by either spin coating (2000 rpm, 60 s) or aerosol jet printing (M3D-103, Optomec), followed by curing in muffle furnace at designated temperatures for one hour in air.

### Fabrication of metal oxide TFTs

Two types of IGZO-based TFTs were fabricated according to the reported procedure<sup>31</sup>. One was bottom-gated TFT using  $\text{SiO}_2$  as the gate insulator and another was the top-gated TFT using solution deposited PMSQ as the gate insulator. For the bottom-gated TFT, the IGZO film was prepared by spin coating the precursor solution on a silicon wafer with 300 nm-thick thermally grown  $\text{SiO}_2$  and annealed at 450 °C in air for 60 min. The IGZO precursor solution was prepared by dissolving  $\text{In}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{Ga}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  and  $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (molar ratio In:Ga:Zn = 3:1.5:2) with ethanolamine in 5 mL 2-methoxyethanol. The final concentration of metallic salts was 0.05 mol/L. The source/drain electrodes were made by thermally evaporating 120 nm thick Al onto the IGZO film via a shadow mask. The heavily doped silicon substrate served as the gate electrode. The channel length (L) and width (W) were 50 and 1000  $\mu\text{m}$ , respectively. A surface modification of IGZO film was carried out by immersing the transistor into 0.5 mM GPTMS solution in toluene (with 4.5 mM water) for 24 h, if necessary. For the top-gated TFT, a PMSQ layer was deposited on top of IGZO film either by spin-coating or aerosol jet printing. Al gate

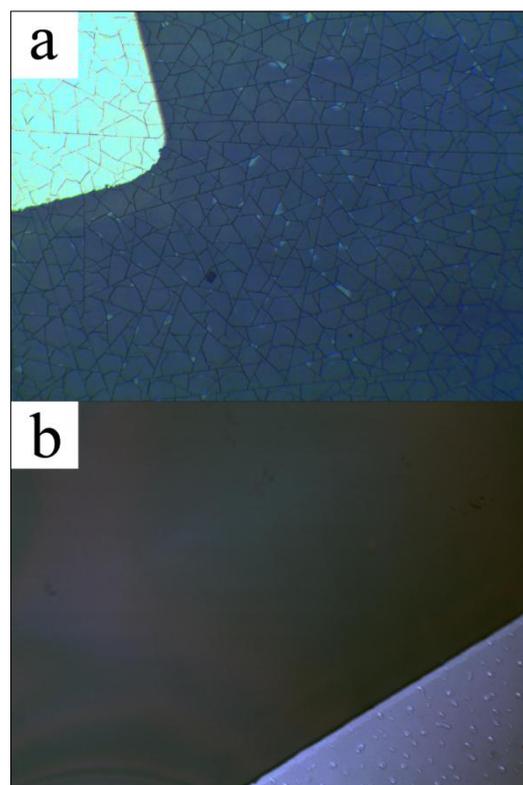


Figure 1 The morphology of PMSQ film cured (a) directly at 450 °C hotplate, (b) a ramp up heating at the rate of 5 °C/min in a muffle furnace for 1 h.

electrode was thermally evaporated on top of the PMSQ film, in addition to the Al source/drain electrodes as described above. For dielectric characterisation of PMSQ films, an Al/PMSQ/ITO structure was utilized. The PMSQ films were prepared by spin-coating the precursor solution on ITO glass. Al electrodes were then deposited on top of spin-coated PMSQ films by thermal evaporation.

### Characterization

The wettability of the surfaces was characterized via static water contact angle goniometry at room temperature (Kino, SL200C). Transmittances of glass substrate, glass/IGZO, glass/PMSQ and glass/IGZO/GPTMS/PMSQ were characterized using a UV-Vis spectroscope (Perkin Elmer Lambda 750 spectrometer). Frequency-dependent loss tangent (dissipation factor) of PMSQ films was measured by Keithley 4200 Capacitance Voltage Unit (from 10 kHz to 1 MHz at 6 V) at room temperature. A step profiler (Veeco, Dektak 150) was used to measure the thicknesses of PMSQ films. The thickness of PMSQ films for the measurement of dielectric constant and leakage current density is  $2 \pm 0.2 \mu\text{m}$ . The dielectric constants were calculated from the measured capacitances, thicknesses and areas of top electrode<sup>1</sup>. At least seven individual measurements were made to determine the dielectric parameters of PMSQ films. All the TFTs were characterized by Keithley Instruments Model 4200-SCS in atmospheric condition. The saturation mobility was calculated from the Formula (1) as follows:

$$\mu_{\text{sat}} = \left( \frac{\partial \sqrt{I_D}}{\partial V_{GS}} \right)^2 \frac{2L}{WC_i} \quad (1)$$

where  $C_i$  is the capacitance of the gate dielectrics per unit area,  $W$  and  $L$  are the channel width and length, respectively,  $V_{GS}$  is the gate voltage, and  $I_D$  is the drain current.

## Results and discussion

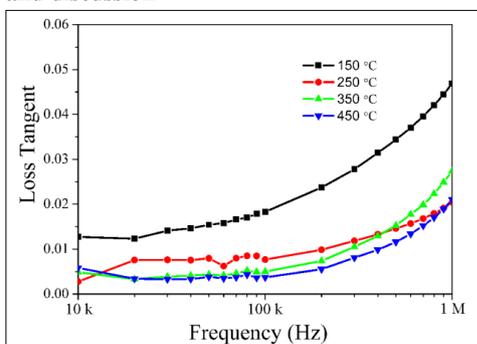


Figure 2 Frequency dependence of loss tangent for the PMSQ films cured at different temperatures.

Table 1 The permittivity and leakage current density of the PMSQ film with different curing temperatures

Curing temperature (°C)	Leakage current density* (nA/cm <sup>2</sup> )	Dielectric constant (10 MHz)
150	48-80	4.0
250	24-40	3.0
350	8-32	2.5
450	<10	2.4

\* Applied voltage is 20 V/μm

High temperature curing of PMSQ is necessary to ensure fully reaction of silanol groups in the PMSQ films. There are few reports on the electrical properties of PMSQ film cured at temperature more than 250 °C. As can be seen in Figure 1, the spin-coated PMSQ film chapped if directly cured it on 450 °C hotplate, as seen in Figure 1a, due to internal stress. A ramp up heating at the rate of 5 °C/min in a muffle furnace can eliminate the cracks as seen in Figure 1b without any crack.

Table 1 and Figure 2 show the leakage current density, dielectric constant and loss tangent of the PMSQ films cured at different temperatures, which were measured with the Al-PMSQ-ITO (Metal-Insulator-Metal) structure. As the curing temperature increased, they decreased accordingly. Note that with 20 V/μm and loss tangent (<0.005) over frequency range of 10 kHz to 1 MHz, the films cured at 350 and 450 °C show very low leakage current density (<10 nA/cm<sup>2</sup>). It is attributed to the extremely low concentration of polar silanol groups and residuals of small molecules in the high temperature treated PMSQ films. Simultaneously, the dielectric constant decreased with low content of polar groups and molecule as seen in Table 1, due to the decrease of polarizability.

PMSQ has been used as dielectric layer in organic thin-film transistors in the past, but suffered from low on/off ratios and high off current<sup>8, 21, 22, 32-34</sup>. Jeong and his coworkers found that increasing the curing temperature of PMSQ-insulator could increase the on/off ratio and reduce the leakage current<sup>34</sup>. The similar trend has been found for inorganic TFTs. Figure 3 shows the transfer characteristics of top-gate top-contact IGZO-TFTs with spin-coated PMSQ dielectric layers cured at 150 and 200 °C. The insert in Figure 3a is the schematic of device. The 150 °C

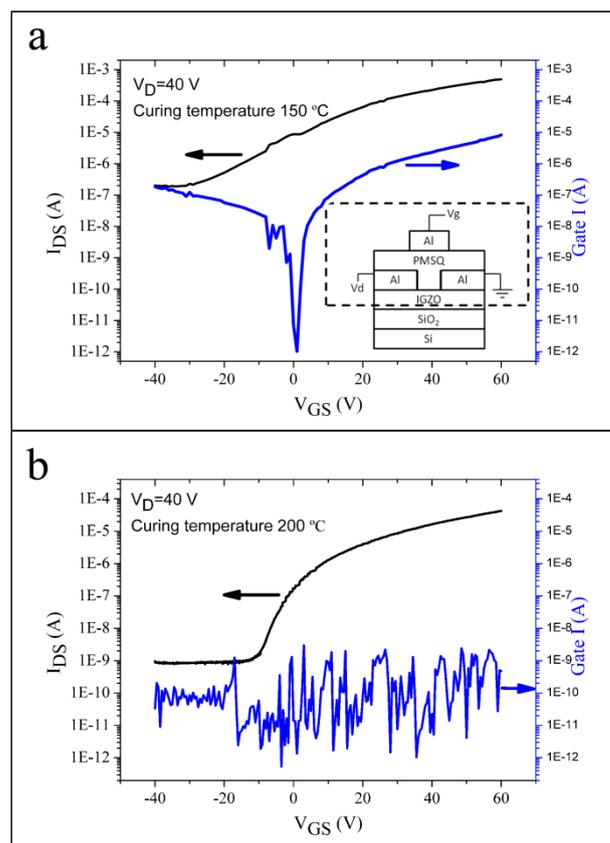


Figure 3 (a) Transfer characteristics of IGZO TFTs with spin-coated PMSQ dielectric layer cured at temperature of (a) 150 °C; (b) 200 °C. (The insert image is the schematic cross-sectional view of a fabricated top-gate, top-contact PMSQ-insulator IGZO TFT).

Table 2 The characteristic results of PMSQ-insulator IGZO TFTs

Curing temperature (°C)	Mobility (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	On/off ratio	Leakage current (10 <sup>-9</sup> A)
150	1.8	2.5 × 10 <sup>2</sup>	100-1000
200	0.4	5 × 10 <sup>4</sup>	0.8-5
150*	1.0	3 × 10 <sup>5</sup>	0.26

\*IGZO and Al electrodes modified with GPTMS

cured PMSQ-insulator TFT (Figure 3a, Table 2) showed on current up to 2 × 10<sup>-4</sup> A, saturation off current 10<sup>-6</sup>-10<sup>-7</sup> A and on/off ratio of 2.5 × 10<sup>3</sup> when the voltage between the drain and the source ( $V_{DS}$ ) was 40 V. With the PMSQ-insulator curing temperature up to 200 °C (Figure 3b, Table 2), the on/off ratio increased to 5 × 10<sup>4</sup> and the off current decreased to 0.8-5 × 10<sup>-9</sup> A. Unfortunately, the mobility also reduced from 1.8 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup> at 150 °C to 0.4 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup> at 200 °C curing. The mobility degradation is due to the poor contact between Al electrode and IGZO semiconductor at high curing temperature. Further increasing the curing temperature of PMSQ (250, 350 and 450 °C) has resulted in the damage of Al electrodes. And we also fabricated PMSQ-insulator TFTs on the glass substrate (See Supplementary Information, Figure S1), and it showed on current up to 3.8 × 10<sup>-4</sup> A, mobility of 0.7 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup> and on/off ratio of 8.5 × 10<sup>4</sup> while the thickness of PMSQ (200 °C) is 600 nm.

Figure 4 shows the transfer characteristics of top-gate, top-contact IGZO transistors before and after coating the PMSQ layer.

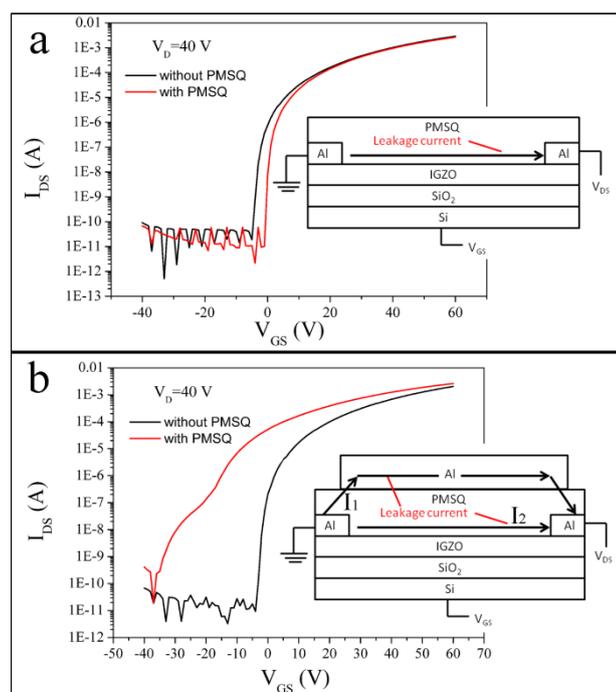


Figure 4 Transfer characteristics of the IGZO-based SiO<sub>2</sub>-insulator transistor with (a) PMSQ, (b) PMSQ/Al electrode.

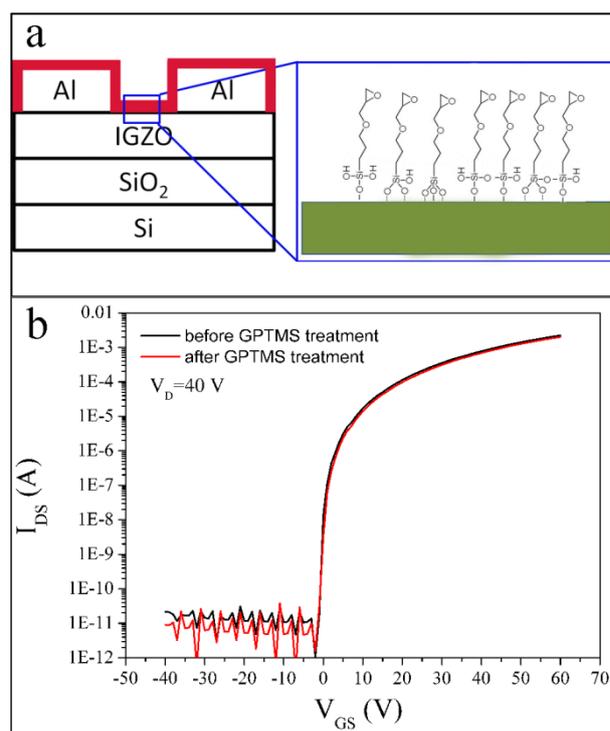


Figure 5 (a) Schematic illustrations of GPTMS modified IGZO TFT structure showing the chemical binding of GPTMS to IGZO. (b) Transfer characteristics of IGZO TFTs before and after the GPTMS treatment.

5 The results indicate that PMSQ layer itself does not increase the leakage current between the source and drain electrodes as seen in Figure 4a, which is in the range of  $10^{-10}$  to  $10^{-11}$  A with gate voltage varied from -40 to 0 V. However, when there is an Al electrode on top of the PMSQ layer, the leakage current increased sharply to  $10^{-8}$  A when the gate voltage is -30 V, as seen in Figure 4b. The high leakage current  $I_1$  (Figure 4b) has in fact included the contribution of current from drain electrode to top Al electrode and to Al source electrode.

15 In order to reduce the leakage current, a molecular passivation using GPTMS on both the Al electrode and the IGZO films was introduced prior to the deposition of PMSQ layer. By immersion in the GPTMS solution, the organic molecules were self-assembled onto Al and IGZO surfaces through hydrolysis of the triethoxy groups of GPTMS and condensation of the hydroxyl groups in GPTMS, as shown in Figure 5a. AFM images showed that the root mean square roughness (Rq) of IGZO films increase from 0.46 nm to 0.74 nm after GPTMS modification (See Supplementary Information, Figure S2), indicating formation of dense GPTMS layer on the IGZO films<sup>6</sup>. It has been experimentally proved that the GPTMS modification would not change the performance of IGZO-based TFT in the mobility, threshold voltage, off/on current and subthreshold swing, as shown in Figure 5b, because there is no polar group in the GPTMS molecule.

30 In contrast, other silane coupling agents were also applied to modify the IGZO surface. Dramatic reduction in the threshold voltage was noticed when agent with polar group was used, such as (3-aminopropyl)triethoxysilane (APTES, See Supplementary Information, Figure S3). Similar modification was also carried out by Seong and his coworkers. It was found the APTES molecules self-assembly led to n-type doping of ZnO because of the strong electron-donating characteristics of the amine group, and finally the threshold voltage and mobility of ZnO TFTs

45 changed accordingly.<sup>6</sup> Agents which contain Si-Cl group like phenyl trichlorosilane (M1) and 1H,1H,2H,2H-perfluorodecyltrichlorosilane (M2) will have great damage to the Al electrodes and IGZO film (See Supplementary Information, Figure S4). This is because plenty of HCl was generated during the hydrolysis of M1 and M2.

50 After GPTMS modification, PMSQ ink was spin-coated and cured at 150 °C, followed by deposition of Al gate electrode. The final TFT structure is shown in Figure 6a. The GPTMS modified TFT showed on current up to  $7.9 \times 10^{-5}$  A, leakage current  $2.6 \times 10^{-10}$  A, on/off current ratio of  $3 \times 10^5$ , and effective mobility up to  $1.0 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ , as seen in Table 2. Figure 6b is the transfer curve of TFT. The output characteristics is shown in Figure 6c, which demonstrates a typical n-channel transistor with clear pinch-off and excellent saturation, implying that the entire thickness of IGZO channel can be completely depleted of free electrons. Compared to the TFT without modification (Figure 3), the modified TFT has much lower gate current, leakage current, higher on/off ratio and acceptable mobility. There are two possible explanations for the improvement. Firstly, the GPTMS molecules form a passivation layer both on Al and IGZO surfaces. The conductive path from source/drain electrodes and IGZO to gate electrode is blocked. Secondly, the contact angle of IGZO and Al electrodes reduced from 88 ° and 83 ° after GPTMS treatment. The change of surface energy may lead to better quality of PMSQ deposition, which helps to reduce the leakage current.

75 Figure 7 (a) shows the photograph of IGZO-based TFT which has GPTMS modification and printed PMSQ ink as the dielectric layer. The TFT showed on current up to  $3.4 \times 10^{-5}$  A, leakage current  $1.66 \times 10^{-9}$  A, on/off ratio of  $2 \times 10^4$ , and effective mobility up to  $0.75 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ , as can be seen in Figure 7b.

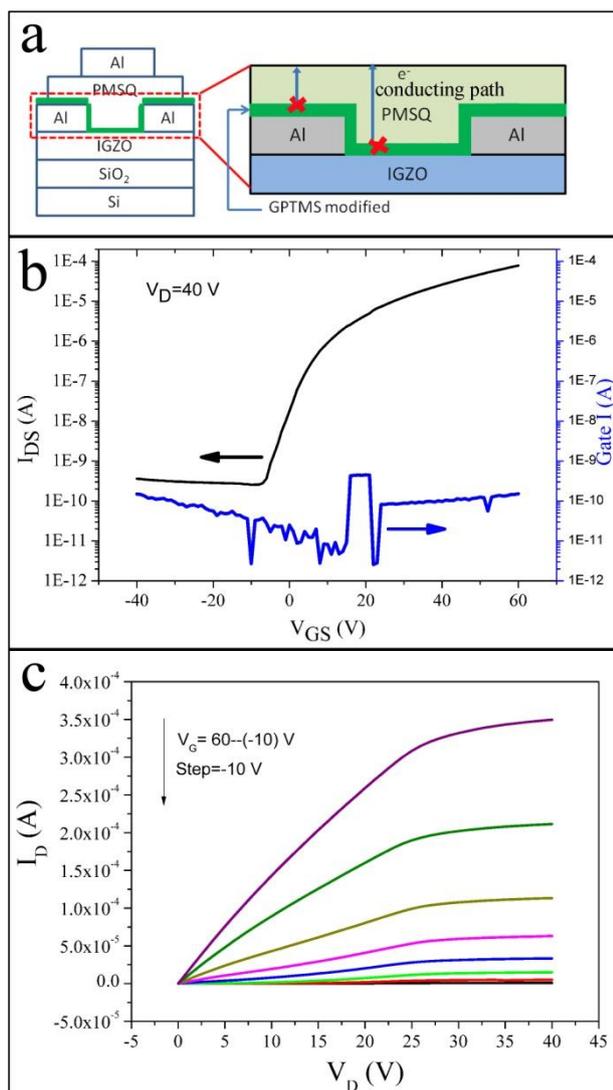


Figure 6 (a) The schematic cross-sectional view of a fabricated GPTMS modified bottom gate, top contact PMSQ-insulator IGZO TFT. (b) Transfer characteristics and (c) output characteristics of IGZO TFTs (curing temperature is 150 °C).

Compared to the TFT with spin-coated PMSQ, its performance is slightly reduced. It may be due to the thicker printed PMSQ film (1.8 μm) than the spin-coated one (1 ± 0.2 μm). It is hard to control the thickness of printed PMSQ layer by using the PMSQ-insulator ink with the present formulation method. The PMSQ ink needs to be further improved for printing thin layers.

As far as we know, the results of TFTs fabricated by PMSQ-insulator with such low leakage current and high on-off ratio have not been reported up to now, even the curing temperature is 190 °C<sup>22</sup>. P4VP insulator, the most popular solution-processable dielectric in the fabrication of transistor, has the leakage current of  $2 \times 10^{-10}$  to  $10^{-8}$  A.<sup>10, 35-39</sup> As mentioned above, comparatively large leakage current of PMSQ-insulator transistor has greatly restricted the dielectrics application of PMSQ. After GPTMS modification, the leakage current of PMSQ insulator has a similar magnitude equal to PVP.

In addition, IGZO, PMSQ and IGZO/GPTMS/PMSQ films on the glass substrate show very high transparency (about 95%)

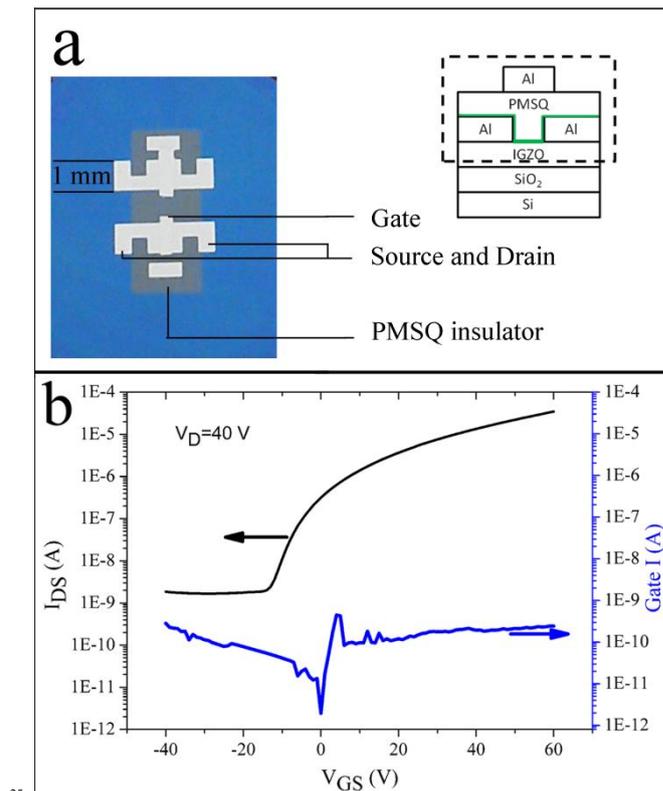


Figure 7 (a) The photograph and (b) transfer characteristics of GPTMS modified, printed PMSQ-insulator IGZO-based TFT (PMSQ curing temperature is 150 °C).

at visible region (400-800 nm wavelength) with no apparent drop compared to the glass substrate. High transparency of IGZO/GPTMS/PMSQ indicated their potential for transparent, high-performance electronics (See Supplementary Information, Figure S5).

### 35 Conclusions

The thermally cross-linked poly(methyl silsesquioxane) (PMSQ) have investigated as a printable dielectric ink and as a gate insulator in IGZO-based thin-film transistors by solution deposition under different curing temperatures. It is found that the dielectric constant, leakage current density and loss tangent of PMSQ layer reduces from 4.0, 48-80 nA/cm<sup>2</sup> and 0.015 to 2.4, <10 nA/cm<sup>2</sup> and 0.0025, respectively, with the curing temperature increases from 150 to 450 °C. When employing the PMSQ layer as a gate insulator in IGZO based TFTs, an interfacial passivation layer of GPTMS was introduced, prior to solution deposition of PMSQ, which improved the on/off ratio to  $3 \times 10^5$  and reduced the leakage current to  $2.6 \times 10^{-10}$  A. This is the best result using solution deposited PMSQ as gate insulator with low curing temperature at 150 °C. Top-gate and top-contact IGZO TFTs were also fabricated by printing PMSQ as the gate insulator, instead of spin coating, and the devices showed on current up to  $3.4 \times 10^{-5}$  A, effective mobility up to  $0.75 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ , leakage current  $1.6 \times 10^{-9}$  A and on/off ratio of  $2 \times 10^4$ .

### Acknowledgments:

This work was supported by the project of the Major Research plan of the National Natural Science Foundation of China (Grant No. 91123034) and the Strategic Priority Research Program of

Chinese Academy of Sciences (Grant No. XDA09020201).

## Notes and references

- \* Printable Electronics Research Centre, Suzhou Institute of Nano-Tech and Nano-Bionics, Chinese Academy of Sciences, 398 Ruoshui Road, Dushu Lake Higher Education Zone, Industrial Park, Suzhou, China. Fax: 62872628-8013; Tel: +86-0512-62872695; E-mail: zchen2007@sinano.ac.cn
- \* Printable Electronics Research Centre, Suzhou Institute of Nano-Tech and Nano-Bionics, Chinese Academy of Sciences, 398 Ruoshui Road, Dushu Lake Higher Education Zone, Industrial Park, Suzhou, China. Fax: 62872628-8013; Tel: +86-0512-62872695; E-mail: zcui2009@sinano.ac.cn
- 1 X. Wu, Z. Chen and Z. Cui, *Compos. Sci. Technol.*, 2013, **81**, 48-53.
  - 2 P. Liu, Y. Wu, Y. Li, B. S. Ong and S. Zhu, *J. Am. Chem. Soc.*, 2006, **128**, 4554-4555.
  - 3 W. Tang, L. Feng, J. Zhao, Q. Cui, S. Chen and X. Guo, *Journal of Materials Chemistry C*, 2014, **2**, 1995-2000.
  - 4 M. R. Beaulieu, J. K. Baral, N. R. Hendricks, Y. Tang, A. L. Briseño and J. J. Watkins, *ACS Appl Mater Inter*, 2013, **5**, 13096-13103.
  - 5 J. H. Park, Y. B. Yoo, K. H. Lee, W. S. Jang, J. Y. Oh, S. S. Chae and H. K. Baik, *ACS Appl Mater Inter*, 2013, **5**, 410-417.
  - 6 S. H. Yu, B. J. Kim, M. S. Kang, S. H. Kim, J. H. Han, J. Y. Lee and J. H. Cho, *ACS Appl Mater Inter*, 2013, **5**, 9765-9769.
  - 7 L. Qian, W. Xu, X. Fan, C. Wang, J. Zhang, J. Zhao and Z. Cui, *J Phys Chem C*, 2013, **117**, 18243-18250.
  - 8 S. Jeong, D. Kim and J. Moon, *J Phys Chem C*, 2008, **112**, 5245-5249.
  - 9 T. Nagase, T. Hamada, K. Tomatsu, S. Yamazaki, T. Kobayashi, S. Murakami, K. Matsukawa and H. Naito, *Adv. Mater.*, 2010, **22**, 4706-4710.
  - 10 Y. Jang, D. H. Kim, Y. D. Park, J. H. Cho, M. Hwang and K. W. Cho, *Appl. Phys. Lett.*, 2005, **87**, 152105.
  - 11 S. H. Ko, H. Pan, C. P. Grigoropoulos, C. K. Luscombe, J. M. J. Fréchet and D. Poulidakos, *Nanotechnology*, 2007, **18**, 345202.
  - 12 Y. Liu, K. Varshramyan and T. H. Cui, *Macromol. Rapid Commun.*, 2005, **26**, 1955-1959.
  - 13 C. Kim, Z. Wang, H.-J. Choi, Y.-G. Ha, A. Facchetti and T. J. Marks, *J. Am. Chem. Soc.*, 2008, **130**, 6867-6878.
  - 14 H. Kang, R. Kitsomboonloha, J. Jang and V. Subramanian, *Adv. Mater.*, 2012, **24**, 3065-3069.
  - 15 S. Chung, S. O. Kim, S. K. Kwon, C. Lee and Y. Hong, *IEEE Electron Device Lett.*, 2011, **32**, 1134-1136.
  - 16 Y. Noguchi, T. Sekitani and T. Someya, *Appl. Phys. Lett.*, 2006, **89**, 253507.
  - 17 S. Yamazaki, T. Hamada, K. Tomatsu, Y. Ueda, T. Nagase, T. Kobayashi, S. Murakami, K. Matsukawa and H. Naito, *Thin Solid Films*, 2008, **517**, 1343-1345.
  - 18 F.-Y. Yang, K.-J. Chang, M.-Y. Hsu and C.-C. Liu, *J. Mater. Chem.*, 2008, **18**, 5927-5932.
  - 19 S. J. Kim, D. L. Kim and H. J. Kim, *Thin Solid Films*, 2009, **517**, 4135-4137.
  - 20 M. Watanabe, K. Muro, T. Hamada, T. Tamai, A. Masuyama, H. Naito and K. Matsukawa, *Chem. Lett.*, 2009, **38**, 34-35.
  - 21 K. Tomatsu, T. Hamada, T. Nagase, S. Yamazaki, T. Kobayashi, S. Murakami, K. Matsukawa and H. Naito, *Jpn J Appl Phys*, 2008, **47**, 3196-3199.
  - 22 S. Jeong, D. Kim, S. Lee, B.-K. Park and J. Moona, *Appl. Phys. Lett.*, 2006, **89**, 092101-092103.
  - 23 S. Y. Lee, S. Chang and J.-S. Lee, *Thin Solid Films*, 2010, **518**, 3030-3032.
  - 24 K. Everaerts, L. Zeng, J. W. Hennek, D. I. Camacho, D. Jariwala, M. J. Bedzyk, M. C. Hersam and T. J. Marks, *ACS Appl Mater Inter*, 2013, **5**, 11884-11893.
  - 25 J. W. Hennek, Y. Xia, K. Everaerts, M. C. Hersam, A. Facchetti and T. J. Marks, *ACS Appl Mater Inter*, 2012, **4**, 1614-1619.
  - 26 G. H. Kim, H. S. Kim, H. S. Shin, B. D. Ahn, K. H. Kim and H. J. Kim, *Thin Solid Films*, 2009, **517**, 4007-4010.
  - 27 S. Y. Han, D. H. Lee, G. S. Herman and C. H. Chang, *J Disp Technol*, 2009, **5**, 520-524.
  - 28 S. Adachi and S. Okamura, *Adv. Mater.*, 2012, **24**, 2945-2986.
  - 29 S. Jeong, J.-Y. Lee, S. S. Lee, S.-W. Oh, H. H. Lee, Y.-H. Seo, B.-H. Ryu and Y. Choi, *J. Mater. Chem.*, 2011, **21**, 17066-17070.
  - 30 Y. Wang, X. W. Sun, G. K. L. Goh, H. V. Demir and H. Y. Yu, *Electron Devices, IEEE Transactions on*, 2011, **58**, 480-485.
  - 31 X. Wu, F. Fei, Z. Chen, W. Su and Z. Cui, *Compos. Sci. Technol.*, 2014, **94**, 117-122.
  - 32 Z. Bao, V. Kuck, J. A. Rogers and M. A. Paczkowski, *Adv. Funct. Mater.*, 2002, **12**, 526-531.
  - 33 Y. Wu, P. Liu and B. S. Ong, *Appl. Phys. Lett.*, 2006, **89**, 013505.
  - 34 S. Jeong, D. Kim, B. K. Park, S. Lee and J. Moon, *Nanotechnology*, 2007, **18**, 025204.
  - 35 M. Hwang, H. S. Lee, Y. Jang, J. H. Cho, S. Lee, D. H. Kim and K. Cho, *Macromolecular Research*, 2009, **17**, 436-440.
  - 36 P. K. Nayak, J. Kim, C. Lee and Y. Hong, *Physica Status Solidi a - Applications and Materials Science*, 2010, **207**, 1664-1667.
  - 37 J.-H. Kwon, M.-H. Chung, T.-Y. Oh, H.-S. Bae, J.-H. Park, B.-K. Ju and F. Yakuphanoglu, *Sensors and Actuators a-Physical*, 2009, **156**, 312-316.
  - 38 J. Kim, J. Jeong, H. D. Cho, C. Lee, S. O. Kim, S.-K. Kwon and Y. Hong, *Journal of Physics D-Applied Physics*, 2009, **42**, 115107.
  - 39 C. J. Chiu, S. P. Chang and S. J. Chang, *Thin Solid Films*, 2012, **520**, 5455-5458.