## CORRELATION BETWEEN THE STRUCTURE OF THE DIELECTRIC MONOLAYER AND THE PERFORMANCE OF LOW-VOLTAGE TRANSISTORS BASED ON PENTACENE

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#### > INTRODUCTION

Alkyl phosphonic acids (C<sub>n</sub>PA) are becoming a material of choice for passivation of high-k oxides in organic thin-film transistors with ultra-thin gate dielectrics. A monolayer of phosphonic acid inserted between the inorganic oxide and the organic semiconductor provides two main benefits: (i) the density of the charge carrier traps associated with the surface –OH groups of the oxide is reduced because these groups act as binding sites for the organic molecules; and (ii) the low surface energy of the organic monolayer may reduce the density of defects in the subsequently deposited conjugated polymer.

To date such monolayers have been assembled from solutions only. We have recently developed a vapour-phase self-assembly of n-octylphosphonic acid (C<sub>8</sub>PA) monolayer in vacuum that leads to a well chemisorbed monolayer of C<sub>8</sub>PA. When such a monolayer is attached to ~ 9-nm thick aluminium oxide to form an ultra-thin dielectric implemented in low-voltage organic thin-film transistors based on pentacene, the transistor performance exhibits measurable changes upon alteration of the structure of the C<sub>8</sub>PA monolayer.

#### > AIM

- Seek correlation between the structure of the dielectric and the short- and long-term performance of organic thin-film transistors •
- Optimize the vapour-phase self-assembly of n-octylphosphonic acid monolayer •





- Fully dry fabrication process
- AlO<sub>x</sub> is prepared by UV/ozone oxidation of aluminium
- All other layers are thermally evaporated in Minispectros vacuum system (K.J. Lesker)



# Samples for FTIR, AFM and WCA

FTIR – Fourier Transform Infrared Spectroscopy AFM – Atomic Force Microscopy WCA – Water Contact Angle Measurement

$\Delta V_t =  V_t(t) - V_t(0)  =  V_t(\infty) - V_t(0)  \cdot \left[1 - e^{-(t/\tau_{V_t})\beta_{V_t}}\right]$
$\Delta S(t) = S(t) - S(0) = (S(\infty) - S(0)) \cdot \left[ 1 - e^{-(t/\tau_S)\beta_S} \right]$
$\frac{\mu(t)}{\mu(0)} = \frac{\mu(\infty)}{\mu(0)} + \left[1 - \frac{\mu(\infty)}{\mu(0)}\right] e^{-(t/\tau_{\mu})\beta_{\mu}}$
$\frac{I_d(t)}{I_d(0)} = \frac{I_d(\infty)}{I_d(0)} + \left[1 - \frac{I_d(\infty)}{I_d(0)}\right] e^{-(t/\tau_{Id})\beta_{Id}}$

#### **BIAS STRESS**

- Performed on 3 sets of transistors with altered C<sub>8</sub>PA monolayer preparation:
- a) C<sub>8</sub>PA deposited at 25°C at 3 Å/s and annealed at 160°C for 25 210 minutes (Post-growth anneal series) 50-nm-thick pentacene deposited at 25°C at 0.24 Å/s, 50-nm-thick Au deposited at 3 Å/s
- b)  $C_8$ PA deposited at 25 150°C at 3 Å/s and annealed at 160°C for 180 minutes (Growth temperature series) - 50-nm-thick pentacene deposited at 70°C at 0.24 Å/s, , 50-nm-thick Au deposited at 3 Å/s
- c)  $C_8$ PA deposited at 25°C at 0.3 10 Å/s and annealed at 160°C for 180 minutes (Growth rate series) - 50-nm-thick pentacene deposited at 55°C at 0.24 Å/s, , 50-nm-thick Au deposited at 3 Å/s
- All transistors were bias-stressed up to 5000 seconds with  $V_{gs} = V_{ds} = -3$  V and the source electrode grounded
- Transfer characteristics measured at certain intervals to determine  $V_{t}$ , S,  $\mu$ , and  $I_{d}$
- Time constant  $\tau$  and stretching parameter  $\beta$  determined for each of  $\Delta V_{t}$   $\Delta S$ ,  $\mu/\mu$ (t=0), and  $I_{d}/I_{d}$ (t=0)

### **RESULTS**

## **POST-GROWTH ANNEAL SERIES**

#### **GROWTH TEMPERATURE SERIES**





-2.0 V

-1.75 V

-1.5 V

 $V_{\mathrm{DS}}\left(\mathrm{V}\right)$ 

 $V_{\rm GS} = -1.25 \ {\rm V}$ 

-2 -1.5 -1

|I<sub>D</sub>| (10<sup>-7</sup> A)

**FTIR** 





-0.5 0

**AS-FABRICATED TRANSISTORS** 

10-7

 $\widehat{\mathbf{E}}^{10^{-8}}$ 

<sup>-6</sup> <sup>10-9</sup>

10-10

10-11

 $V_{\rm DS} = -2.0 \ {\rm V}$ 

-0.1 V

-2 -1.5 -1 -0.5 0

 $V_{\mathrm{GS}}\left(\mathrm{V}
ight)$ 

**BIAS STRESS** 



C <sub>8</sub> PA annealing time (min)	$ au_{_{Vt}}$ (s)	$eta_{\scriptscriptstyle Vt}$	${ au}_{\scriptscriptstyle S}$ (s)	$\beta_{s}$	${ au}_{\mu}$ (s)	$eta_{\mu}$	${ au}_{_{Id}}$ (s)	$oldsymbol{eta}_{\it Id}$
25	3.3×10 <sup>6</sup>	0.15	4.0×10 <sup>4</sup>	0.35	4.5×10 <sup>2</sup>	0.61	3.0×10 <sup>2</sup>	0.29
60	8.8×10 <sup>3</sup>	0.23	7.0×10 <sup>4</sup>	0.36	5.7×10 <sup>2</sup>	0.61	3.7×10 <sup>2</sup>	0.30
90	1.6×10 <sup>3</sup>	0.26	5.3×10 <sup>6</sup>	0.42	5.7×10 <sup>2</sup>	0.61	1.8×10 <sup>2</sup>	0.33
210	7.5×10 <sup>2</sup>	0.28	3.7×10 <sup>10</sup>	0.72	7.0×10 <sup>2</sup>	0.65	1.4×10 <sup>2</sup>	0.34





#### **BIAS STRESS**

C <sub>8</sub> PA growth T (°C)	$ au_{_{Vt}}$ (s)	$\beta_{Vt}$	${ au}_{\scriptscriptstyle S}$ (s)	$\beta_{s}$	$ au_{\mu}$ (s)	$m{eta}_{\mu}$	${ au}_{_{Id}}$ (s)	$oldsymbol{eta}_{{\scriptscriptstyle Id}}$
25	2.6×10 <sup>2</sup>	0.30	9.5×10 <sup>10</sup>	0.70	3.6×10 <sup>2</sup>	0.65	2.2×10 <sup>2</sup>	0.34
75	6.7×10 <sup>2</sup>	0.27	1.1×10 <sup>4</sup>	0.60	3.8×10 <sup>2</sup>	0.53	2.2×10 <sup>2</sup>	0.35
125	6.9×10 <sup>2</sup>	0.25	1.7×10 <sup>4</sup>	0.59	5.6×10 <sup>2</sup>	0.53	2.6×10 <sup>2</sup>	0.37
150	2.9×10 <sup>3</sup>	0.25	5.0×10 <sup>3</sup>	0.51	5.7×10 <sup>2</sup>	0.50	2.6×10 <sup>2</sup>	0.38



- Long post-growth anneal leads to the lowest surface roughness, highest water contact angle and best molecular ordering of C<sub>8</sub>PA monolayer.
- Properties are similar to  $C_8$ PA monolayers assembled from solution.
- Long post-growth C<sub>8</sub>PA anneal also leads to the highest stretching factors for the threshold voltage and subthreshold slope.

#### **BIAS STRESS**

C <sub>8</sub> PA growth rate (Å/s)	$ au_{_{Vt}}$ (s)	$eta_{_{Vt}}$	${ au}_{\scriptscriptstyle S}$ (s)	$\beta_{s}$	${ au}_{\mu}$ (s)	$oldsymbol{eta}_{\mu}$	${ au}_{_{Id}}$ (s)	$oldsymbol{eta}_{\it Id}$
0.3	5.4×10 <sup>2</sup>	0.35	7.5×10 <sup>2</sup>	0.58	1.6×10 <sup>3</sup>	0.62	2.4×10 <sup>2</sup>	0.38
1	5.8×10 <sup>2</sup>	0.37	9.2×10 <sup>2</sup>	0.60	1.0×10 <sup>3</sup>	0.63	2.8×10 <sup>2</sup>	0.44
3	7.0×10 <sup>2</sup>	0.37	1.4×10 <sup>3</sup>	0.60	1.1×10 <sup>3</sup>	0.63	4.6×10 <sup>2</sup>	0.44
10	5.4×10 <sup>2</sup>	0.35	1.5×10 <sup>3</sup>	0.59	1.3×10 <sup>3</sup>	0.70	3.2×10 <sup>2</sup>	0.39

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#### > CONCLUSIONS

- Correlation between monolayer structure and long-term transistor stability was uncovered.
- C<sub>8</sub>PA annealing has the largest effect on the bias-induced transistor degradation, then the growth temperature and finally the evaporation rate.
- The best  $C_8PA$  properties were obtained at 25°C, 1-3 Å/s and 210-minute anneal. This correlates with improved transistor performance.
- These conditions lead to highest values of  $\beta$  for V<sub>t</sub> and S indicating narrower distribution of carrier traps.
- Pentacene growth also has a minor effect on the values of  $\beta$  and  $\tau$ .

