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**Electronic Supplementary Information** 

## Ladder-Like Polysilsesquioxane Dielectrics for Organic Field-Effect Transistor Applications

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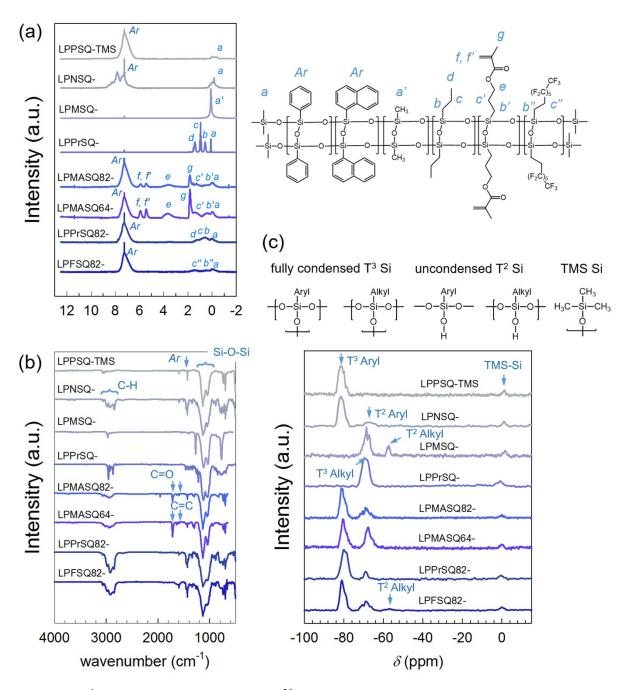


Fig. S1 (a)  $^{1}$ H NMR, (b) FT-IR, and (c)  $^{29}$ Si NMR spectra of LPSQ-TMS series studied in this paper.

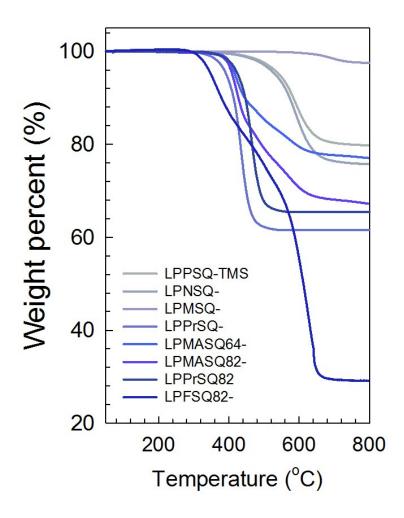


Fig. S2 TGA profiles of LPSQ-TMS series studied in this work.

	LPSQ-TMS treated SiO <sub>2</sub>								untreated
	LPPSQ-	LPM82-	LPM64-	LPMSQ-	LPPrSQ-	LPPrSQ82-	LPFSQ82-	LPNSQ-	SiO <sub>2</sub>
				Liquid	Contact				
H <sub>2</sub> O	6						0	<u> </u>	
$\theta_{\rm H_{2}O}(^{\circ})$	95.7	92.8	91.3	108.1	105.2	97.2	113.0	99.5	39.3
$CH_2I_2$				-					
$\theta_{\mathrm{CH}_2\mathrm{I}_2}$ (°)	37.5	45.5	53.9	75.2	71.7	40.3	81.4	(dissolved)	46.4
				Surface ener	gy ( <i>γ</i> , mJ m <sup>-2</sup> )				
non-polar (၇ <sup>d</sup> )	41.9	36.2	30.2	19.9	21.6	40.7	16.9	N/A	24.0
oolar (ፇ)	0.10	0.80	1.80	0.30	0.50	0.05	0.20	N/A	33.6
$\gamma (= \gamma^d + \gamma^p)$	42.0	37.0	32.0	20.2	22.1	40.75	17.1	N/A	57.6

Fig. S3  $\theta$ -based  $\gamma$  values of LPSQ-TMS treated and untreated SiO<sub>2</sub> dielectrics.

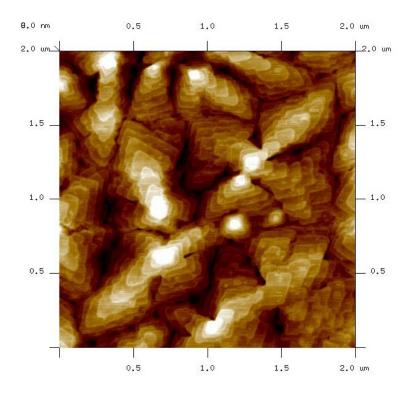


Fig. S4 AFM topography of 20 nm thick pentacene film on the LPMASQ82-TMS treated  $SiO_2$  surface.

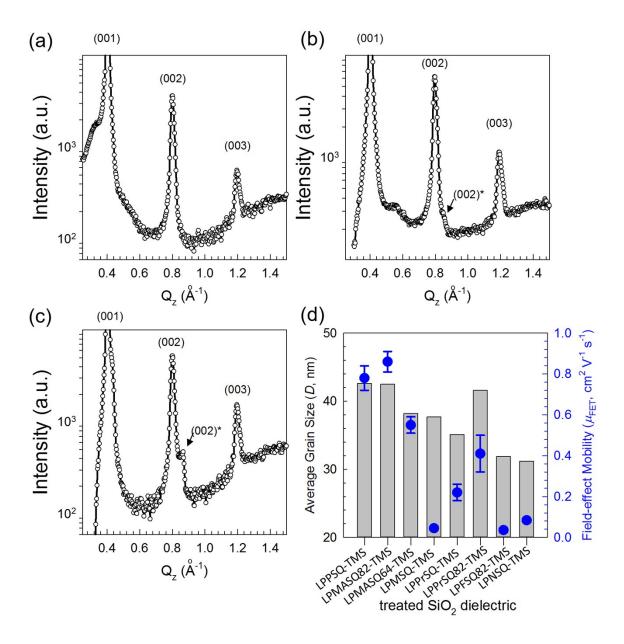


Fig. S5 (a–c) 1D out-of-plane X-ray diffraction profiles extracted along the  $Q_z$  axis from the 2D GIXD patterns of (a) LPMASQ82-, (b) LPPrSQ82-, (c) LPNSQ-TMS treated SiO<sub>2</sub> systems. (d) Variations in *D* and  $\mu_{\text{FET}}$  of 50 nm thick pentacene films on the LPSQ-treated SiO<sub>2</sub> dielectrics.

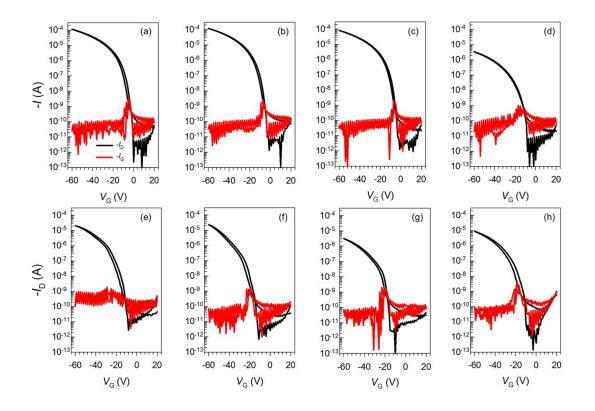


Fig. S6 Typical  $I_D-V_G$  transfer and  $I_G-V_G$  gate leakage curves of pentacene OFETs on the treated SiO<sub>2</sub> dielectrics including: (a) LPPSQ-, (b) LPMASQ82-, (c) LPMASQ64-, (d) LPMSQ-, (e) LPPrSQ-, (f) LPPrSQ82-, (g) LPFSQ-, and (h) LPNSQ-TMS layers.

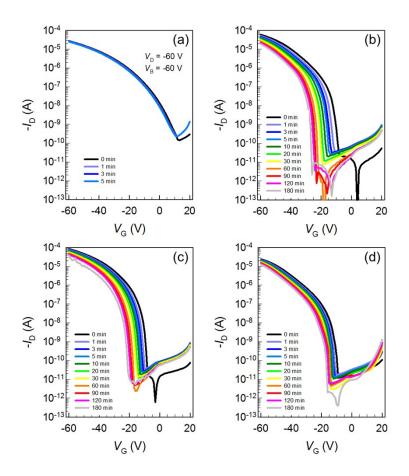


Fig. S7  $I_D-V_G$  transfer curves of 50 nm pentacene OFETs on the (a) untreated and (b–d) LPSQ-TMS treated SiO<sub>2</sub> dielectrics including: (b) LPPSQ-, (c) LPMASQ82-, and (d) LPPrSQ82-, under a sustained gate bias of = -60 V as a function of stress time (*t*).

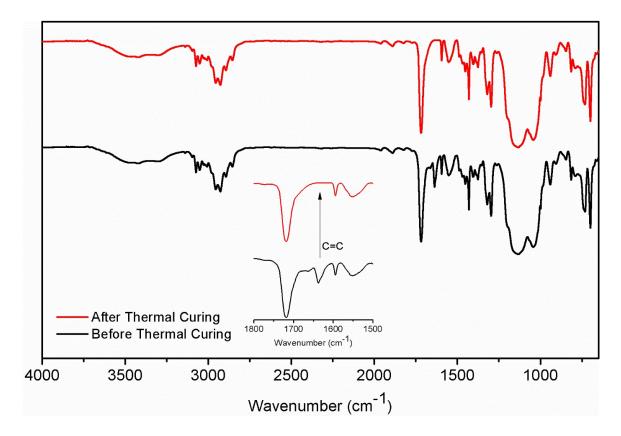


Fig. S8 FT-IR spectra of LPMASQ82-TMS/PMFM (95/5) before and after thermal curing.

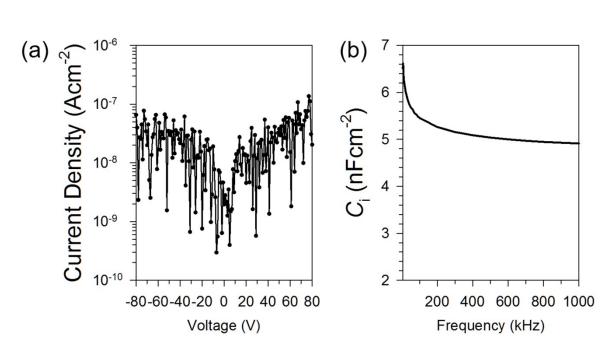


Fig. S9 (a) Current density and (b)  $C_i$  profile of 500 nm thick LPMASQ82-TMS film with  $\varepsilon_r$  of 2.77.

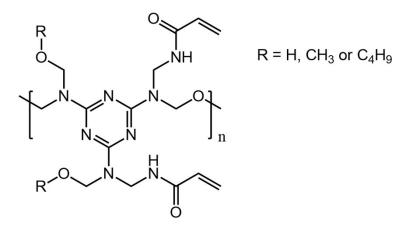


Fig. S10 Chemical structure of poly(melamine-co-formaldehyde), acrylated.