

Electronic Supplementary Information

Densely cross-linked polysiloxane dielectric for organic thin-film transistors with enhanced electrical stability

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Table S1 Summarized characteristics of organic-inorganic hybrid dielectrics and performance of the devices fabricated employing them as gate dielectric in literature

| Dielectric Material | Process condition | Thickness | ¹ J-V range | ² k | Active layer | SAM | Mobility | Hysteresis | ³ ΔV_{Th} | Ref. |
|---|-------------------|-----------|------------------------|----------------|---|----------------------------|---|------------|------------------------------|------|
| | | (nm) | (MV cm ⁻¹) | | | | (cm ² V ⁻¹ s ⁻¹) | (V) | (V) | |
| 3-methacryloxypropyltrimethoxysilane and zirconium propoxide | 170 °C | 190 - 265 | 2 | 5.5 | ^a DH α 4T | - | 1.0 \times 10 ⁻³ | N/A | N/A | 23 |
| DPSD (diphenylsilanediol) and MPTMS (3-(Trimethoxysilyl)propyl methacrylate) | 150°C, UV | 40 | 3 | 3.1 | Pentacene | - | 0.3 | Negligible | N/A | 24 |
| Anthryl-terminated alkyl-phosphonic acid on UV cured sol-gel HfOx | 200°C, UV | 4.8 | 2 | - | Pentacene / ^b TIPS-Pentacene | ^g PA derivative | 0.32 / 0.38 | Negligible | N/A | 25 |
| PMSQ (poly(methyl silsesquioxane)) | 150°C | 450 | 3 | 3.6 | ^c P3HT | - | 7.1 \times 10 ⁻³ | Negligible | on, -1 V (1,000 s) | 26 |
| ZrCl4 and α,ω -disilylalkane | 150°C, Vacuum | 20 - 43 | 2 | 5 - 10 | Pentacene | - | 0.1 - 1.6 | Negligible | N/A | 27 |
| Solution based ZrO ₂ (zirconium-(IV) acetylacetonate as a precursor) | UV | 5 - 6 | 3 | ~10 | ^d PBTTC-C14 | ^h ODPA | 0.2 | N/A | N/A | 28 |
| Cyclotetrasiloxane and melamine | 80°C | 400 | 1 | 3.79 | Pentacene | - | 0.36 | 3.3 | N/A | 29 |
| ZrTA (zirconium tetraacrylate) | 120°C | 50 - 60 | 2 | 5.48 | Pentacene | - | 0.5 | Negligible | N/A | 30 |
| PPMSQ (poly(phenyl-co-methacryl silsesquioxane)) | 200°C | 920 - 980 | 1 | 3.1 - 3.6 | Pentacene / ^e PTCDI-C8 | - | 0.53 / 0.17 | N/A | N/A | 31 |
| PMMS (poly[(mercaptopropyl)methyl-siloxane]) | R.T | 1,000 | 2 | 5.4 | ^f DPP-DTT / Pentacene | - | 5.5 \times 10 ⁻² / 1.3 \times 10 ⁻³ | Negligible | on/off, ~3 V (10,000 s) | 32 |
| LPSQ-TMS (trimethylsilyl-capped hybrid ladder-like polysilsesquioxane) | 110°C | 500 | 2 | - | Pentacene | - | 0.6 | Negligible | N/A | 33 |
| poly(azomethine) containing the isobutyl-substituted T8 cages | 100°C | - | N/A | - | P3HT | - | 4.3 \times 10 ⁻³ | Negligible | N/A | 34 |

¹Range of electric field applied for evaluating dielectric strength reported (leakage current), ²dielectric constant ³Shift of threshold voltage under the on/off bias stress condition

^a α,ω -Dihexylquaterthiophene, ^b 6,13-bis(triisopropyl-silylethynyl) pentacene, ^cpoly(3-hexylthiophene-2,5-diyl), ^dPoly(2,5-bis(3-tetradecylthiophen-2yl)(thieno[3,2-b]thiophene),

^eN,N'-dioctyl-3,4,9,10-perylenedicarboximide, ^fDiketopyrrolopyrrole-dithiophene-thienothiophene, ^ganthryl-terminated alkyl-phosphonic acid, ^hoctadecylphosphonic acid

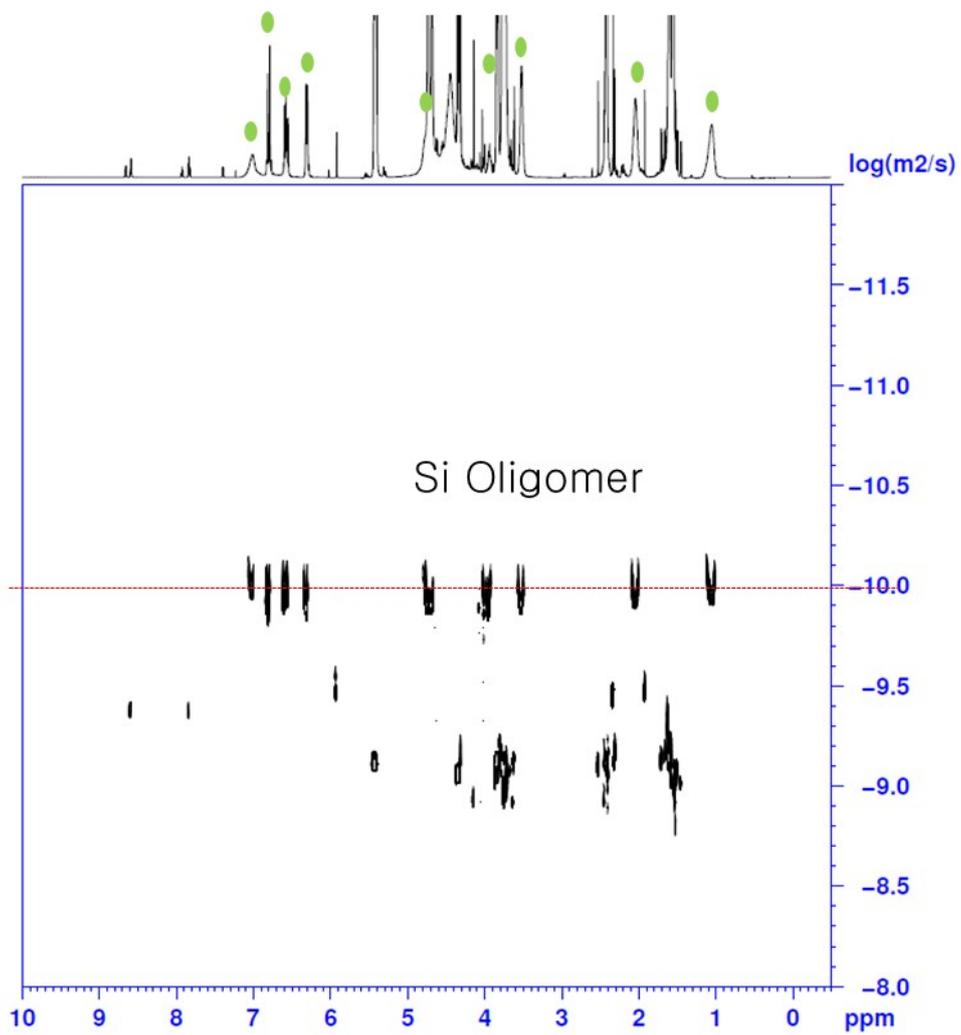


Fig. S1 2D-DOSY NMR of PSUA Solution

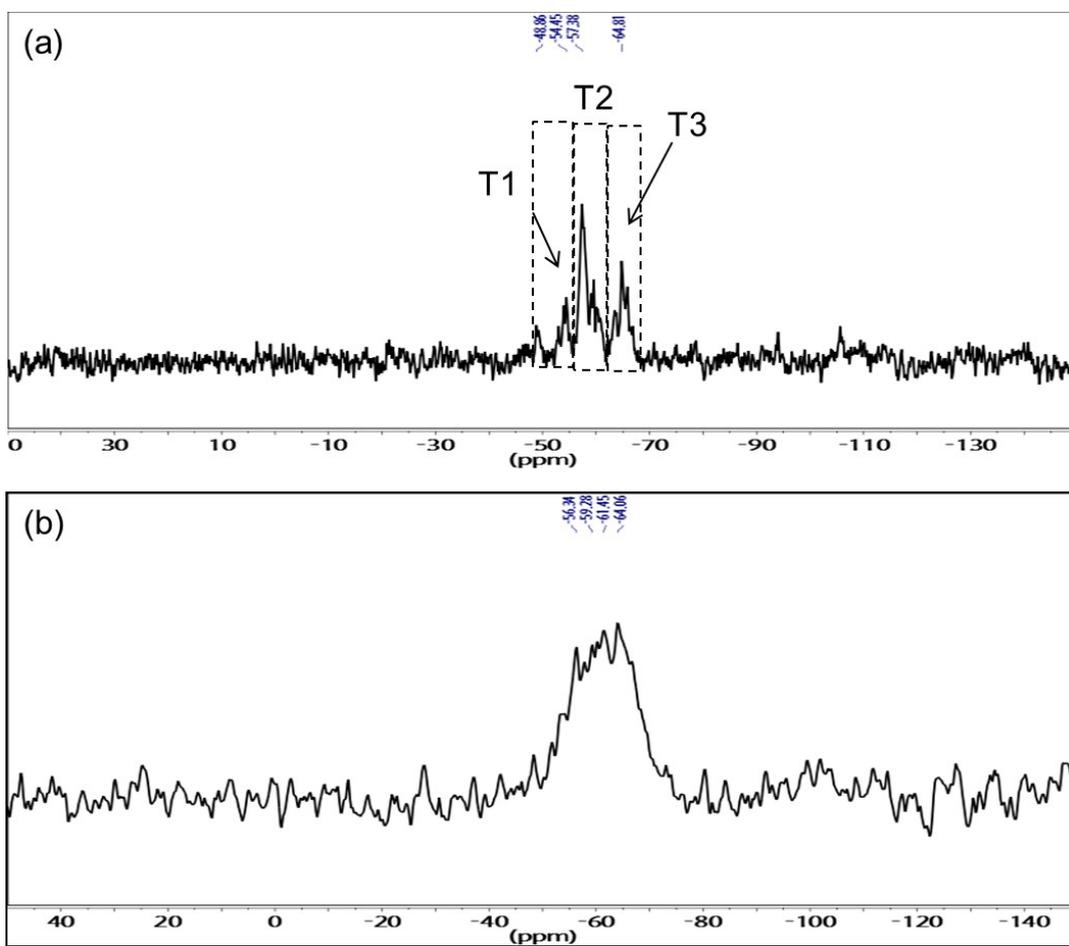


Fig. S2 (a) ^{29}Si NMR of PSUA solution (oligomer)
(b) ^{29}Si CP-MAS NMR of cross-linked PSUAC thin-film (solid)

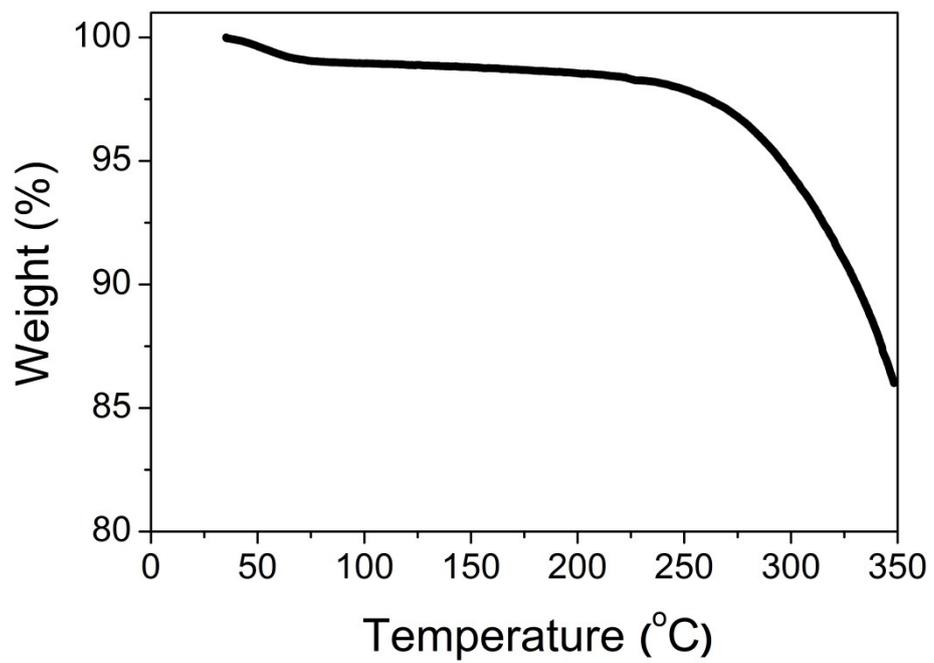


Fig. S3 TGA of cross-linked PSUAC thin-film

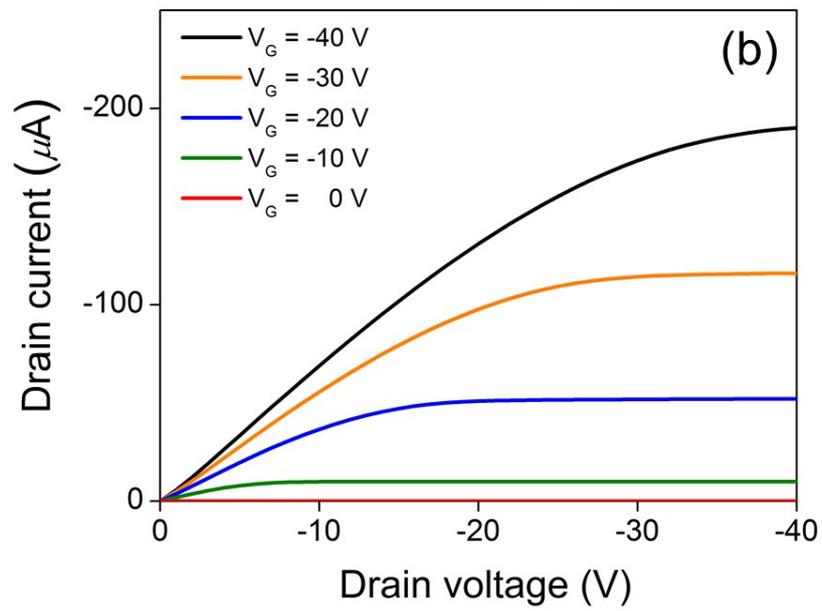
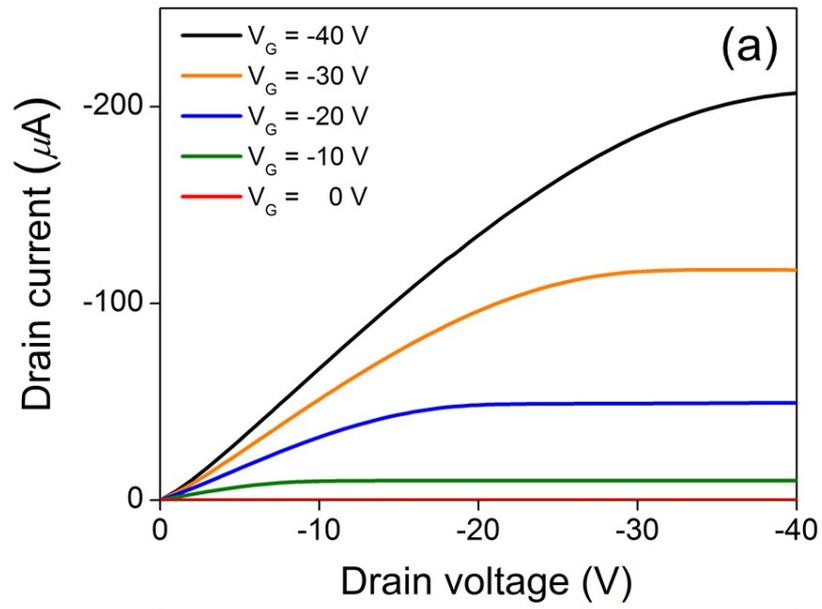


Fig. S4 Output curves of DBTTT TFTs fabricated on (a) PSUAC (b) PECVD SiO_2

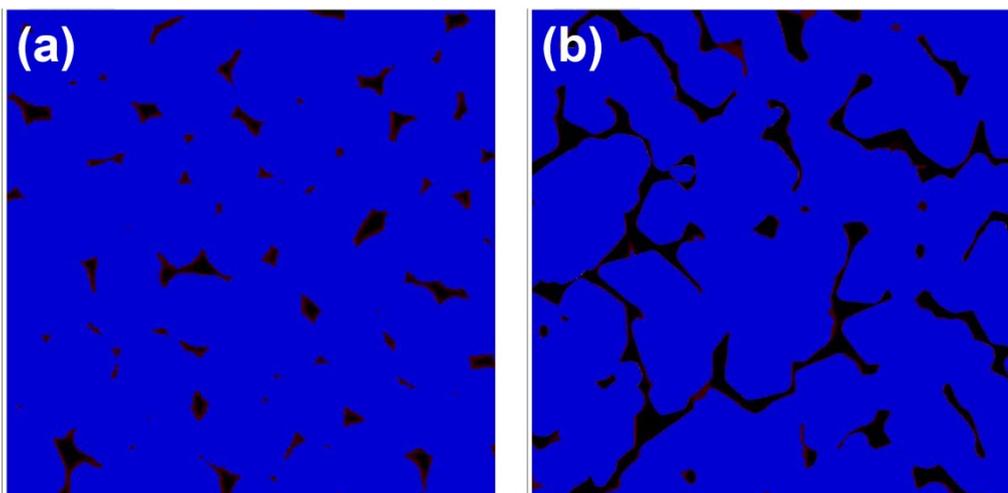


Fig. S5 AFM image analysis of DBTTT morphologies deposited on
(a) PSUAC (b) PECVD SiO₂