Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

A Facile Post-Peeling Modification Approach of Elastic Dielectrics for High-Performance Conformal Organic Thin-Film Transistors

Xiaoli Zhao, Hanbing Li, Qingxin Tang, Yanhong Tong and Yichun Liu*

Key Laboratory of UV Light Emitting Materials and Technology under Ministry of Education, Northeast Normal University, Changchun 130024, P. R. China

E-mail: tangqx@nenu.edu.cn

Tel./fax: +86-431-85099873.

Table S1. Detailed performance list for the reported elastic dielectric-based organic thin-film transistors.

Semiconductor	Elastic dielectric	Type	μ ($cm^2V^{-1}s^{-1}$)	I_{on}/I_{off}	Ref.
P3HT NW/PDMS	Crosslinked CTBN	p	0.00048±00013	-	[1]
CuPc	GR150	p	0.0026	2.5×10^2	[2]
α-6T	GR150	p	0.006	6×10^2	[2]
FCuPc	GR150	n	0.008	3×10^2	[2]
PHT	GR150	p	0.02	6	[2]
P3HT	PU	p	0.034±0.0163	$5.9\times10^2\pm4.6\times10^2$	[3]
DH-α-5T	GR150	p	0.045	6×10^3	[2]
PiI2T-Si	PDMS	p	0.09±0.0008	-	[4]
Pentacene	GR150	p	0.1	10^5	[1][2]
CNTs	TPU	p	0.18 ± 0.03	$1.3\times10^4\pm3.4\times10^3$	[5]
PSe-DPP/PII-2T	PVDF-HFP/PVP	p	0.199	10^5	[6]
TTA-DPP-polymer	$ZnCl_2$-PDMS	p	0.29 ± 0.06	1.2×10^4	[7]
TTA-DPP-polymer	$FeCl_2$-PDMS	p	0.35 ± 0.1	1.3×10^3	[7]
PTDPPTFT4	PDMS	p	0.4-0.1	-	[8]
C12-DPP	SEBS	p	0.463	-	[9]
PTDPPTFT4	ePVDF-HFP	p	0.48 ± 0.16	10^2–10^3	[10]
DPPT-TT:SEBS	SEBS	p	0.59	10^4	[11]
DPP-polymer	PDMS	p	0.6	$>10^5$	[12]
Pentacene	PDMS	p	0.65	10^4	[13]
29-DPP-SVS-(2)	SEBS	p	1.78	10^5	[14]
PTCDI-C13	DC 1-2577	n	1.03	10^6	Our work
DNTT	DC 1-2577	p	5.43	10^6	Our work
DNTT	PDMS	p	2.79	2.2×10^4	Our work
DNTT	DC 3-1965	p	2.61	7.5×10^6	Our work

Table S2. Detailed difference list of three kinds of organic silicone.

Elastic dielectric	Viscosity (cP)	Durometer Shore A	Ultimate enlongation (%)	Dielectric Strength (kV/mm)	Dielectric constant
DC 1-2577	950	80	77	16	2.76
DCS 184	3500	48	78	19	2.8
DC 3-1965	115	33	122	17	2.5

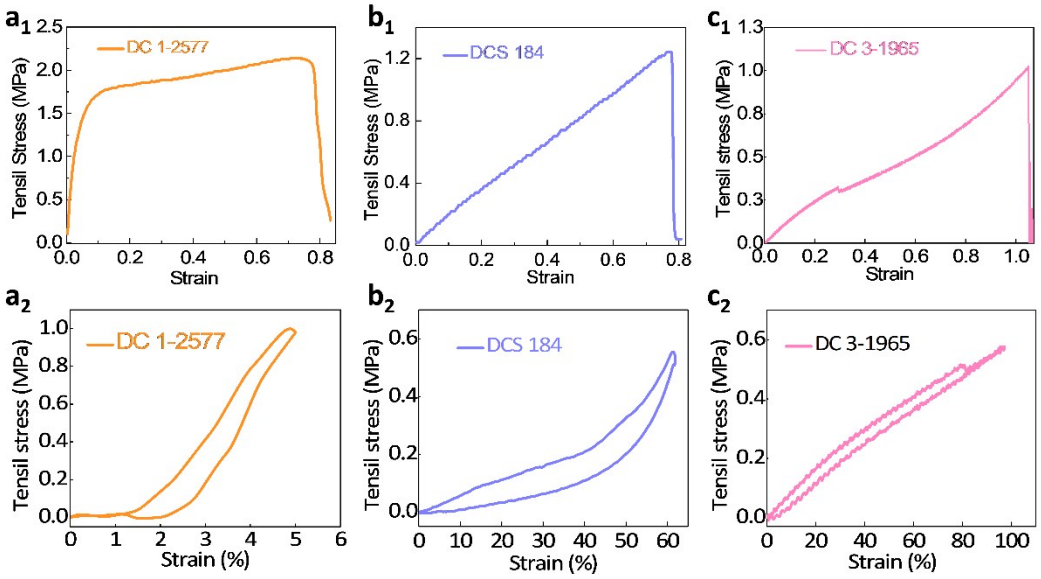

Fig. S1 Stress-strain curve and cyclic stress-strain hysteresis of DC 1-2577, DCS 184 and DC 3-1965. The ultimate enlongation of DC 1-2577, DCS 184 and DC 3-1965 is 77, 78, 122%, respectively. When the yield strain is higher than their ultimate enlongations, they will lose their dielectric performances. Cyclic stress-strain curves obviously show their stretched and restored features, which confirms these materials have elastic deformation in the corresponding strain range.

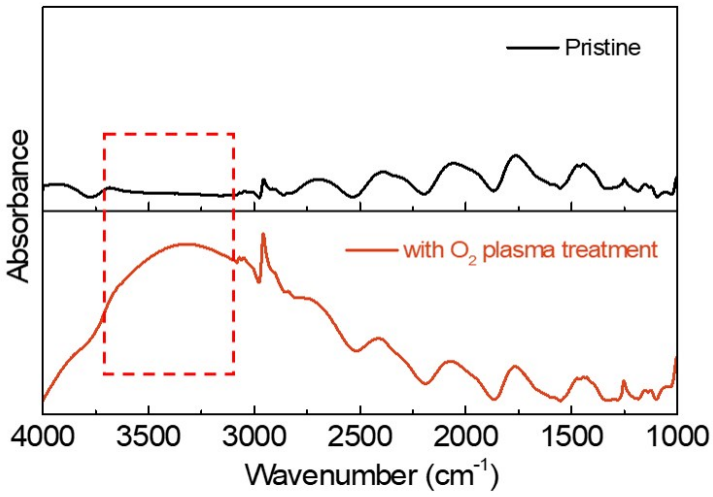

Fig. S2 FTIR spectra of bare and O_2 plasma-treated DC 1-2577 samples. DC 1-2577 elastic dielectric after O_2 plasma treatment shows obvious hydroxy peak.[15] The broad bands centered at around 3420 cm^{-1} correspond to the stretching vibrations of different hydroxyl groups.[16]

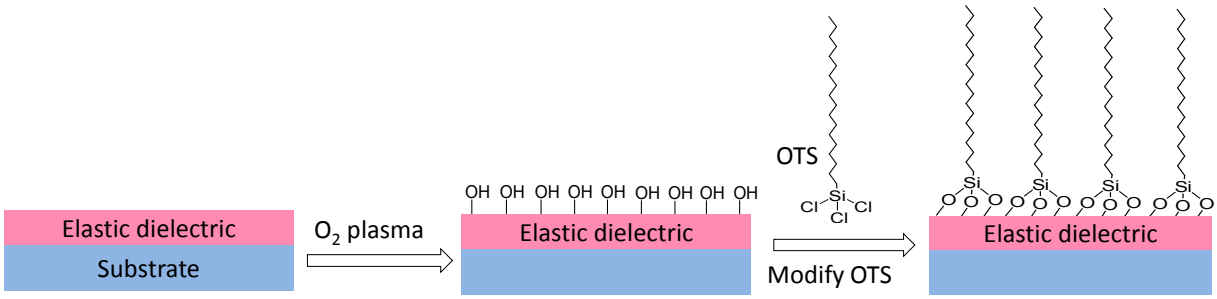

Fig. S3 Schematic images of OTS modification. The elastic dielectric is firstly treated with O_2 plasma to produce sufficient hydroxyl anchoring sites, and then chemically bonded with the chlorine terminal groups of OTS molecules to form the dense SAM via van der Waals forces.

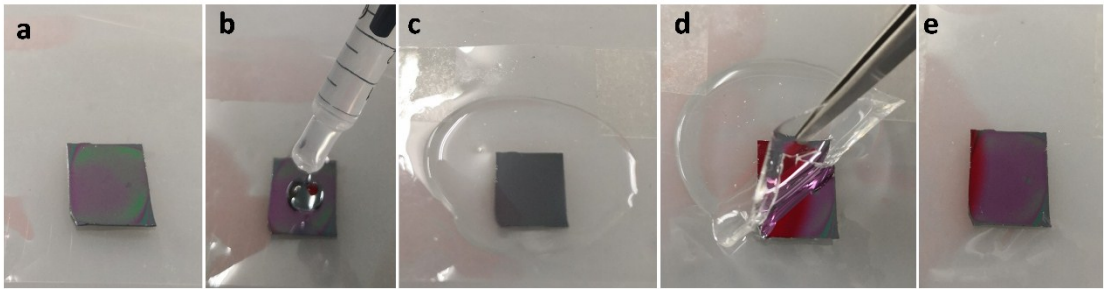

Fig. S4 Detailed PDMS post-peeling process. (a,b) The undiluted PDMS was dropped onto the OTS-modified elastic dielectric with a plastic injector. (c) PDMS gradually covered the entire substrate due to the self-leveling characteristic. (d) After PDMS curing in an oven, the PDMS layer was peeled off with a tweezer. (e) After peeling off PDMS, an ultrasmooth OTS monolayer was formed on elastic dielectric surface.

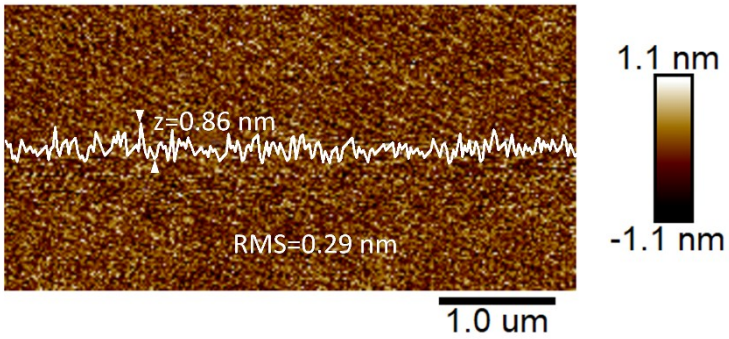

Fig. S5 AFM image of OTS-modified SiO_2 dielectric layer after the PDMS post-peeling treatment. The calculated surface roughness is ～0.29 nm, and the height of our OTS layer is ～0.89 nm, which is similar with previous report.[17]

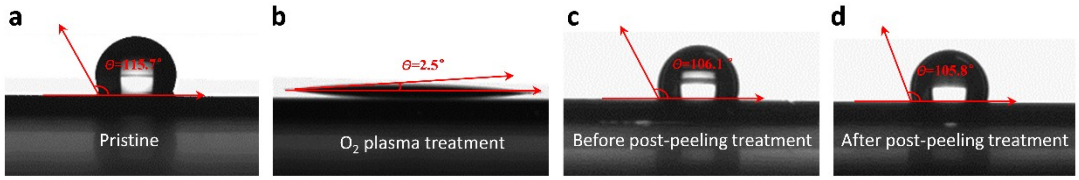

Fig. S6 Different contact angle of DC 1-2577 elastic dielectric. (a) Pristine. (b) After O_2 plasma treatment. (c,d) Before and after PDMS post-peeling treatment. The contact angle of pristine elastic dielectric layer was 115.7°. After O_2 plasma treatment, the contact angle changed to 2.5°. This result indicates that DC 1-2577 dielectric changes from hydrophobic interface to hydrophilic interface, because it produces a large number of hydroxyl groups on DC 1-2577 surface after O_2 plasma treatment. Fig. S6c and 6d show contact angle variation of OTS modified DC 1-2577 before and after PDMS post-peeling treatment. Their contact angles are almost the same (～106°), and obviously higher than the contact angle after O_2 plasma treatment. This result confirms the formation of OTS after modification and the existence of OTS after post-peeling treatment.

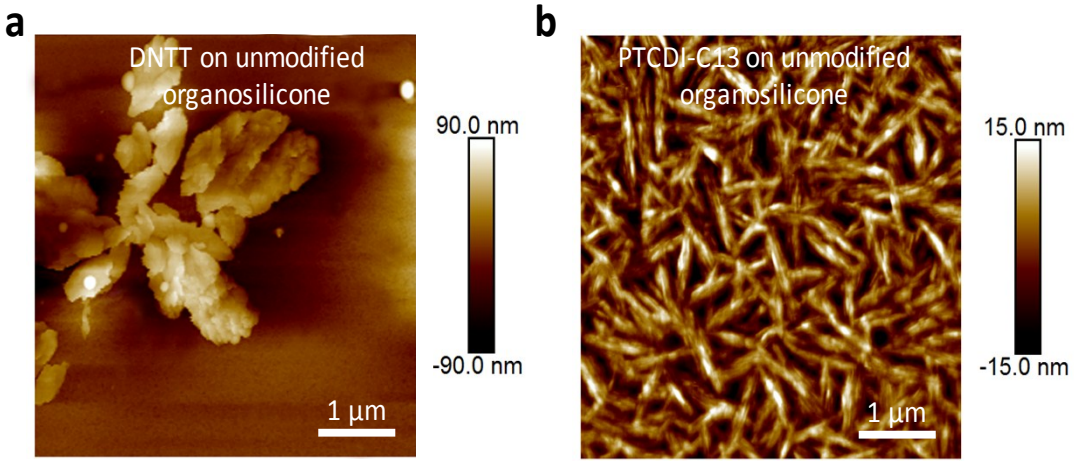

Fig. S7 AFM images of a) DNTT and b) PTCDI-C13 thin film on the pristine elastic dielectric (DC 1-2577) without OTS modification.

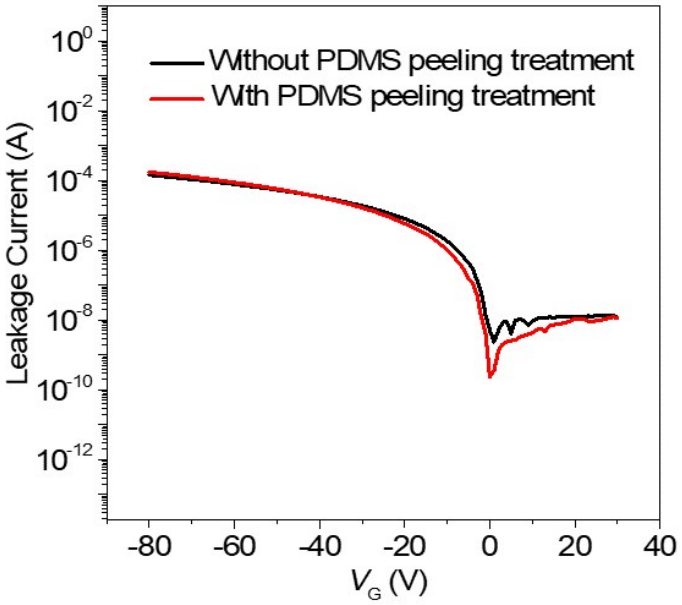

Fig. S8 Variation of gate leakage current before and after PDMS post-peeling treatment.

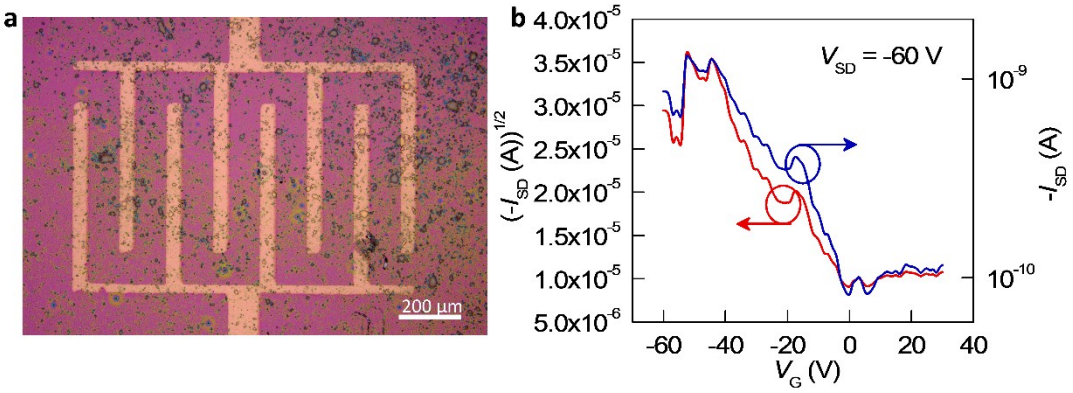

Fig. S9 DNTT OTFT based on solvent-treated OTS-modified DC 1-2577 elastic dielectric. (a) Optical image. (b) Electrical property.

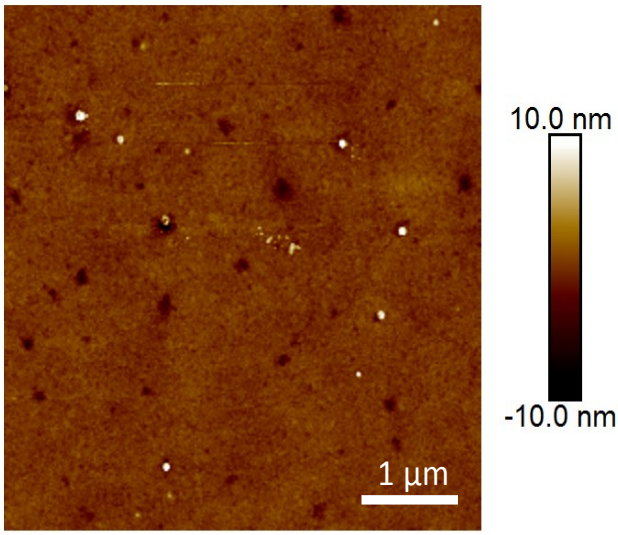

Fig. S10 AFM image of an elastic dielectric with OTS treatment time of 4h. After PDMS post-peeling treatment process, some residues of OTS aggregates randomly remain on the surface of the elastic dielectric.

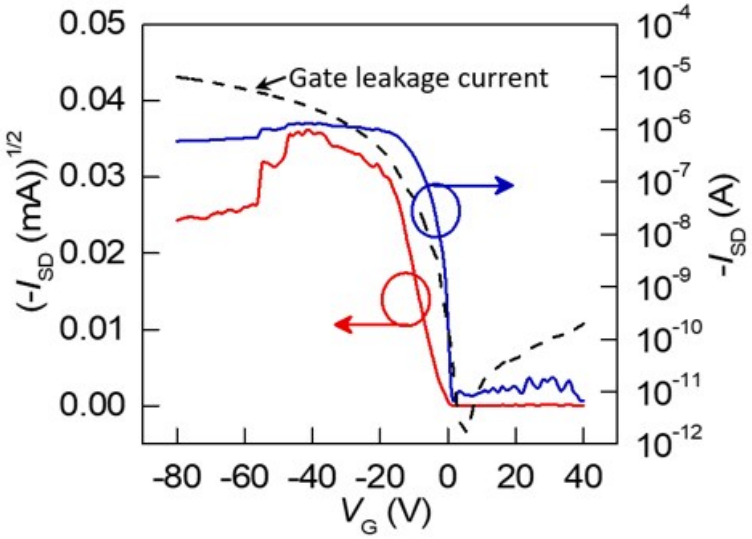

Fig. S11 Typical transfer characteristic of a DNTT OTFT with a thinner DC 1-2577 elastic dielectric after PDMS-peeling treatment.

References

[1] B. Kang, E. Song, S. B. Lee, B.-G. Chae, H. Ahn and K. Cho, *Chem. Mater.*, 2018, **30**, 6353.

[2] Z. Bao, V. Kuck, J. A. Rogers and M. A. Paczkowski, *Adv. Funct. Mater.*, 2002, **12**, 526.

[3] A. Chortos, J. Lim, J. W. F. To, M. Vosgueritchian, T. J. Dusseault, T.-H. Kim, S. Hwang and Z. Bao, *Adv. Mater.*, 2014, **26**, 4253.

[4] G. Schwartz, B. C. K. Tee, J. Mei, A. L. Appleton, D. H. Kim, H. Wang and Z. Bao, *Nat. Commun.*, 2013, **4**, 1859.

[5] A. Chortos, G. I. Koleilat, R. Pfattner, D. Kong, P. Lin, R. Nur, T. Lei, H. Wang, N. Liu, Y.-C. Lai, M.-G. Kim, J. W. Chung, S. Lee and Z. Bao, *Adv. Mater.*, 2016, **28**, 4441.

[6] C. Lu, W. Y. Lee, C. C. Shih, M. Y. Wen and W. C. Chen, *ACS Appl. Mater. Interfaces*, 2017, **9**, 25522.

[7] Y.-L. Rao, A. Chortos, R. Pfattner, F. Lissel, Y.-C. Chiu, V. Feig, J. Xu, T. Kurosawa, X. Gu, C. Wang, M. He, J. W. Chung and Z. Bao, *J. Am. Chem. Soc.*, 2016, **138**, 6020.

[8] D. Kong, R. Pfattner, A. Chortos, C. Lu, A. C. Hinckley, C. Wang, W.-Y. Lee, J. W. Chung and Z. Bao, *Adv. Funct. Mater.*, 2016, **26**, 4680.

[9] J. Mun, G.-J. N. Wang, J. Y. Oh, T. Katsumata, F. L. Lee, J. Kang, H.-C. Wu, F. Lissel, S. Rondeau-Gagné, J. B. H. Tok and Z. Bao, *Adv. Funct. Mater.*, 2018, **28**, 1804222.

[10] R. Pfattner, A. M. Foudeh, C. Liong, L. Bettinson, A. C. Hinckley, D. Kon and Z. Bao, *Adv. Electron. Mater.*, 2018, **4**, 1700326.

[11] Xu, S. Wang, G.-J. N. Wang, C. Zhu, S. Luo, L. Jin, X. Gu, S. Chen, V. R. Feig, J. W. F. To, Simon Rondeau-Gagné, J. Park, B. C. Schroeder, C. Lu, J. Y. Oh, Y. Wang, Y.-H. Kim, H. Yan, R. Sinclair, D. Zhou, G. Xue, B. Murmann, C. Linder, W. Cai, J. B.-H. Tok, J. W. Chung and Z. Bao, *Science*, 2017, **355**, 59.

[12] J. Y. Oh, S. Rondeau-Gagné, Y.-C. Chiu, A. Chortos, F. Lissel, G.-J. N. Wang, B. C. Schroeder, T. Kurosawa, J. Lopez, T. Katsumata, J. Xu, C. Zhu, X. Gu, W.-G. Bae, Y. Kim, L. Jin, J. W. Chung, J. B. H. Tok and Z. Bao, *Nature*, 2016, **539**, 411.

[13] S. Zhou, M. Li, Q. Tang, Z. Song, Y. Tong and Y. Liu, *IEEE Electron Device Lett.*, 2017, **38**, 1031.

[14] S. Wang, J. Xu, W. Wang, G.-J. N. Wang, R. Rastak, F. Molina-Lopez, J. W. Chung, S. Niu, V. R. Feig, J. Lopez, T. Lei, S.-K. Kwon, Y. Kim, A. M. Foudeh, A. Ehrlich, A. Gasperini, Y. Yun, B. Murmann, J. B. H. Tok and Z. Bao, *Nature*, 2018, **555**, 83.

[15] H. T. Kim and O. C. Jeong, *Microelectron. Eng.,* 2011*,* **88**, 2281.

[16] P. Zhao, R. Ni, K. Wang, X. Hong, Y. Ding, T. Cong, J. Liu and H. Zhao, *Anal. Bioanal. Chem., 2017,* **409**, 4207.

[17] Y. Ito, A. A. Virkar, S. Mannsfeld, J. H. Oh, M. Toney, J. Locklin and Z. Bao, *J. Am. Chem. Soc.*, 2009, **131**, 9396.