An Ultrastable Large-Area Atomically Flat 2D Polymer Dielectric for Low-Voltage Flexible Organic Field-Effect Transistors

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Electronic Supplemental Information

Table S1 Properties and applications of recently developed 2DP films.

Ref.	Synthesis method	Size	Thickness	RMS [nm]	Support	Reaction times	Application
1	direct synthesis	centimeter	25 nm	N/A	ITO/glass	3 days	resistive memory
2	air-liquid interface	4-inch	70 nm	N/A	N/A	5 days	N/A
3	air-liquid interface	centimeter	30 nm	1.5	N/A	48 h	memristor
4	air-liquid interface	wafer	25 nm	0.61	N/A	24 h	memristor
5	precursor deposition	N/A	N/A	N/A	Au (111)	N/A	catalysis
6	spin-coating	N/A	27 nm	4	N/A	N/A	solar cell
7	air-liquid interface	wafer	2.5-46 nm	N/A	N/A	24 h	OFET (active layer)
8	liquid-liquid interface	N/A	20 μm	N/A	N/A	3 days	dielectric
9	direct synthesis	wafer	20 nm	0.70	graphene / MoS_2	12 h	dielectric
this work	air-liquid interface	centimeter	15 nm	0.69	N/A	24 h	OFET (dielectric)

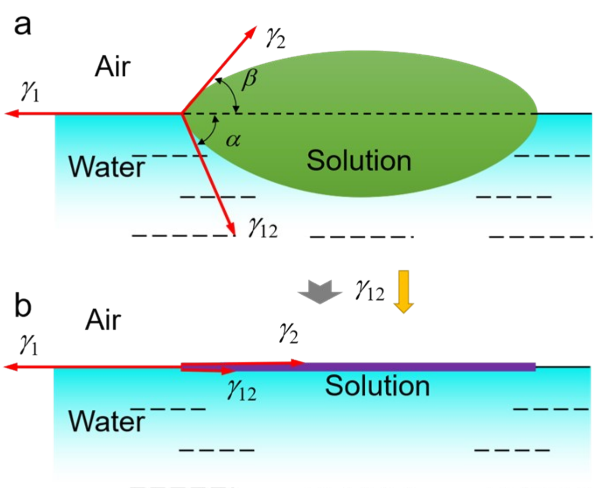

Fig. S1 Balancing of surface tension at air-liquid interface with (a) pure chloroform (b) mix solvents. γ_1, γ_2 are the surface tensions of the water and solution, respectively, and γ_{12} is the interfacial surface tension of the two phases.

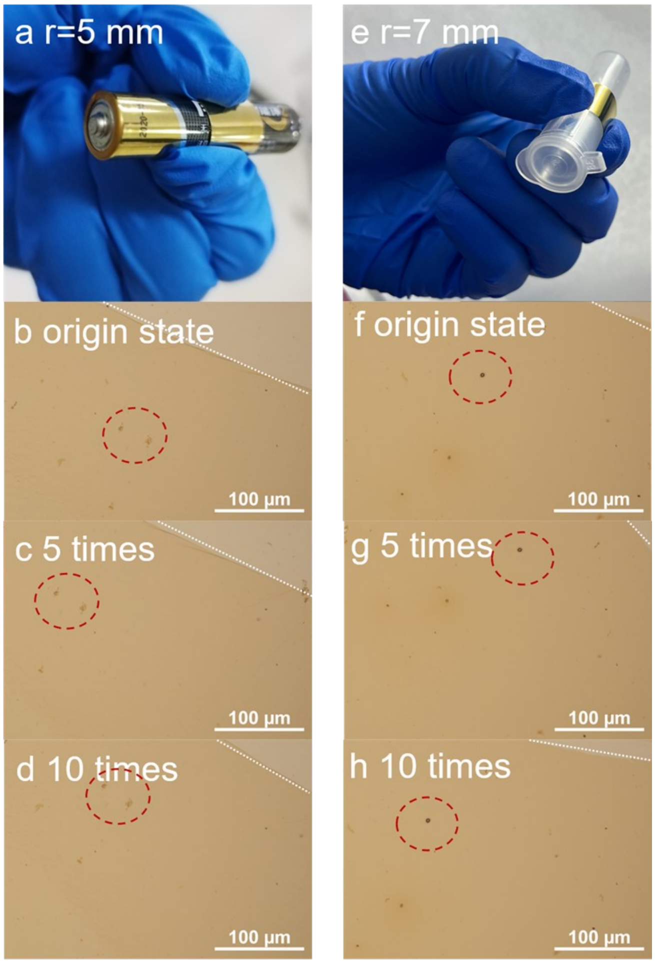

Fig. S2 Photographs of the poly-TT 2DP film being bent (a, e) and the optical microscopy after different bending radius of *r* = 5 mm (b, c, d) and *r* = 7 mm (f, g, h).

To measure the mechanical strength quantitative, we transferred the 2DP film (15 nm in thickness) to PEN substrate (125 μm in thickness). The substrate carrying the 2DP film was then bent to a radius of 5mm and 7mm. The strains ϵ were calculated according to the following equation:

$$\epsilon = \frac{h_s}{2r} \times 100$$

where *r* is the curvature radius, h_s is the substrate thickness (125 μm). Obviously, when the strain was 1.25% (*r* = 5 mm) and 0.89% (*r* = 7 mm), respectively, the film showed no cracks, which indicated the potential application as gate dielectrics in flexible electronics.

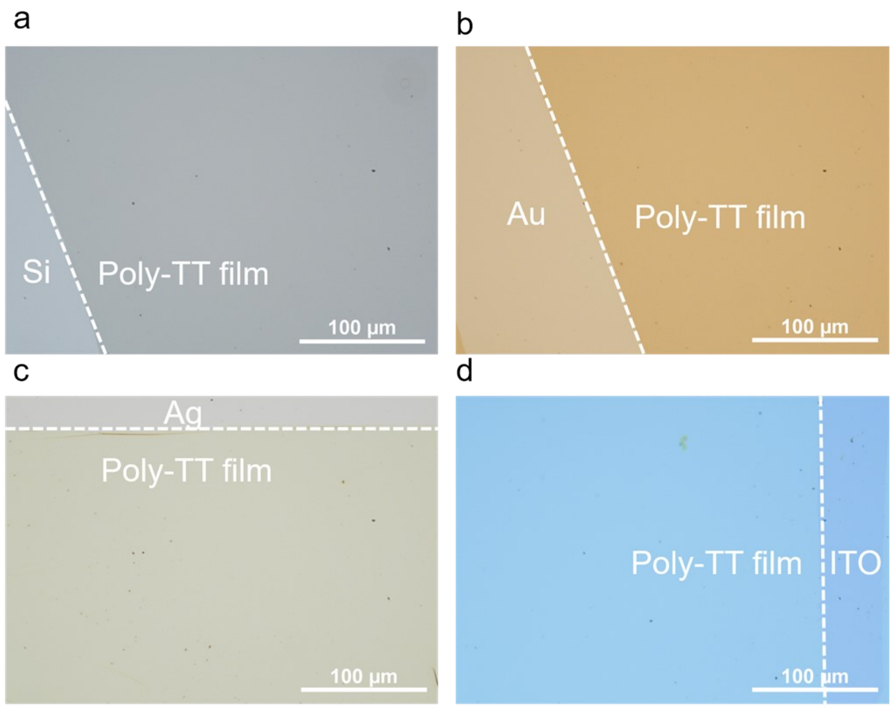

Fig. S3 poly-TT 2DP films transferred on various substrates. (a) Si (b) Au (c) Ag (d) ITO.

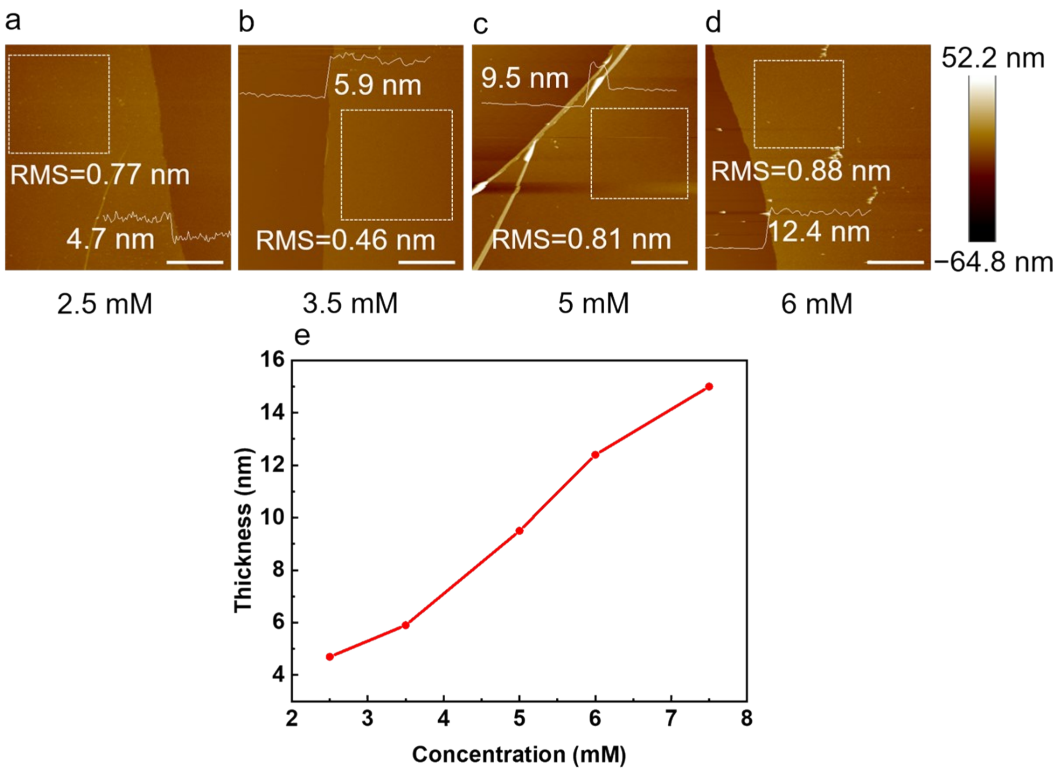

Fig. S4 (a-d) AFM images of poly-TT 2DP films with different TAPB monomer concentrations. (e) Graph of TAPB monomer concentration verse poly-TT 2DP film thickness. Scale bar, 5 μm.

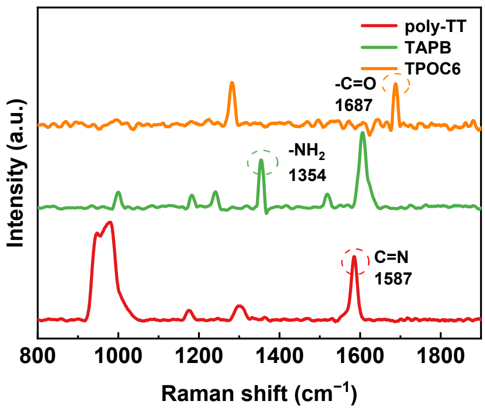

Fig. S5 Raman spectra of poly-TT 2DP film, TAPB and TPOC6.

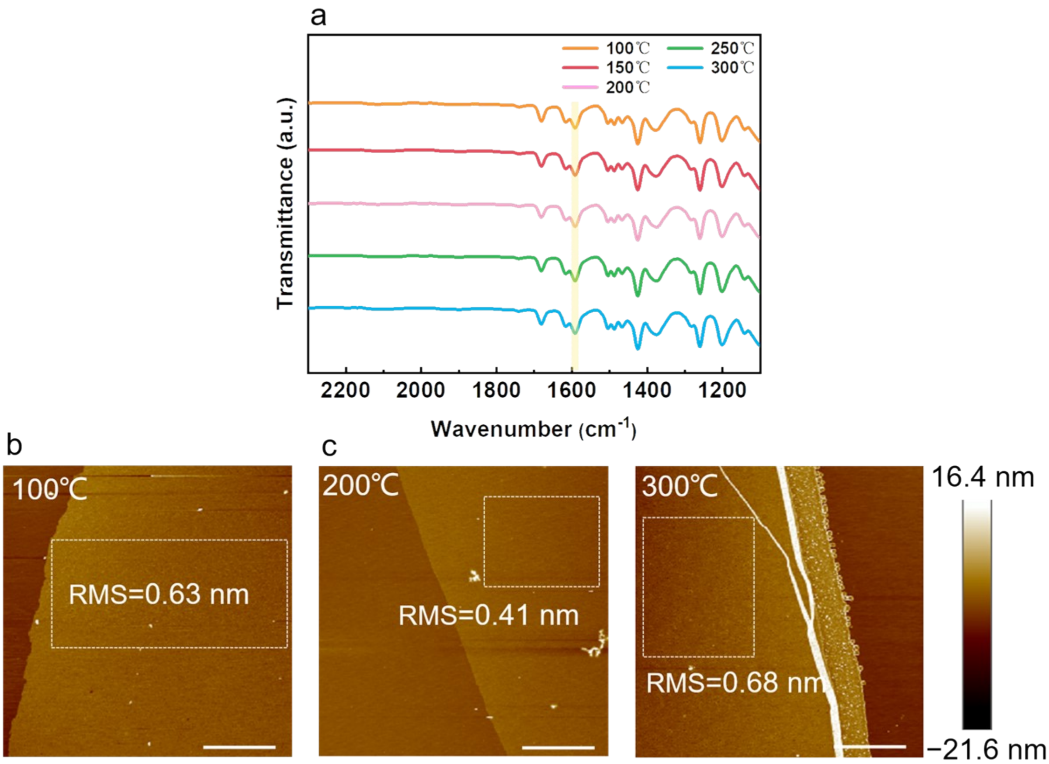

Fig. S6 (a) FTIR of poly-TT 2DP films, which were heated at various temperatures for 20 min, respectively. (b,c,d) AFM images of poly-TT 2DP films, which were heated at (b) 100 °C, (c) 200 °C, (d) 300 °C for 20 min. Scale bar, 5 μm.

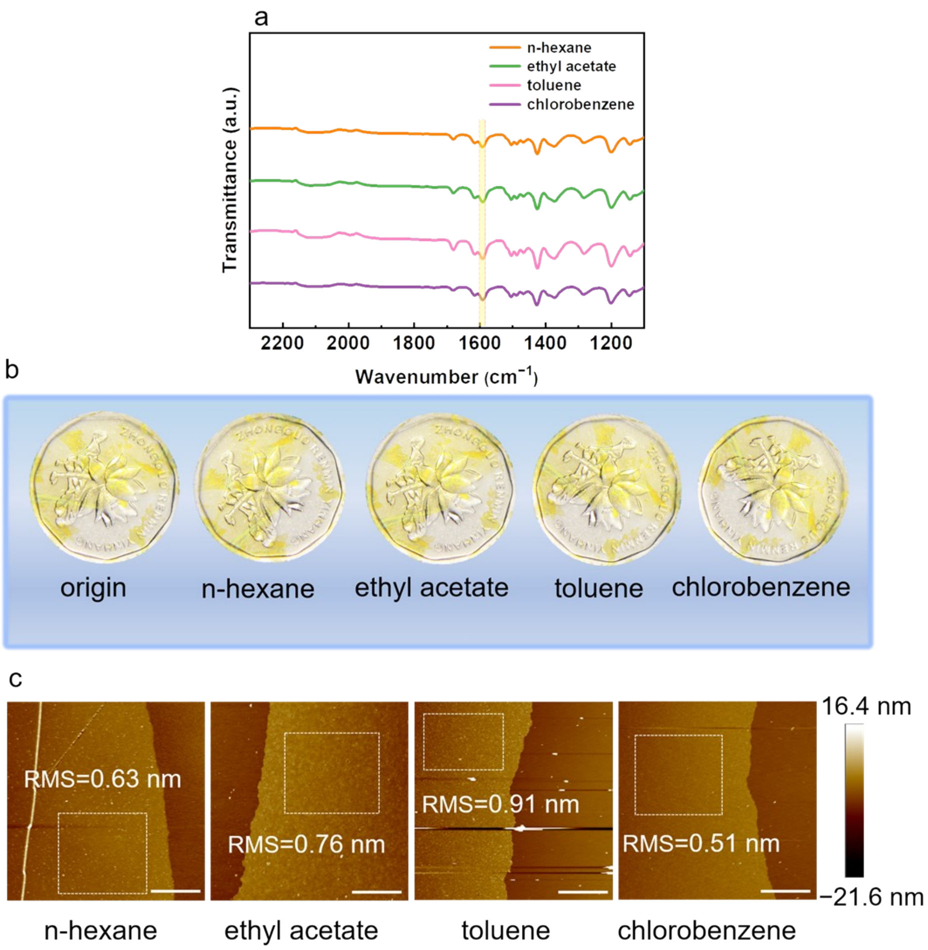

Fig. S7 (a) FTIR, (b) digital images and (c) AFM images of poly-TT 2DP films after immersing in n-hexane, ethyl acetate, toluene and chlorobenzene for 12 h, respectively. Scale bar, 5 μm.

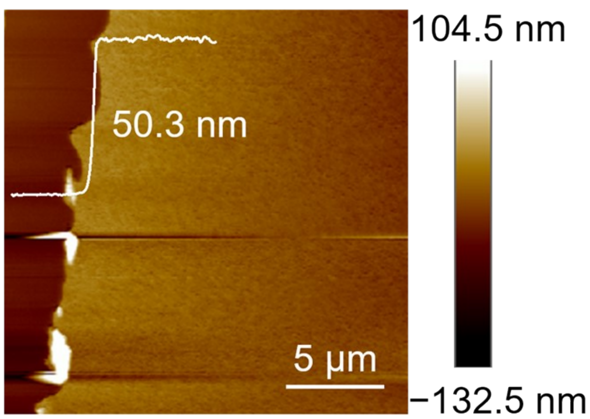

Fig. S8 AFM image of the poly-TT 2DP film as dielectric layer in OFETs. Thick film of 50 nm was made by 6 consecutive transfers of the 8~9 nm 2DP grown by the air-liquid interfacial polymerization method.

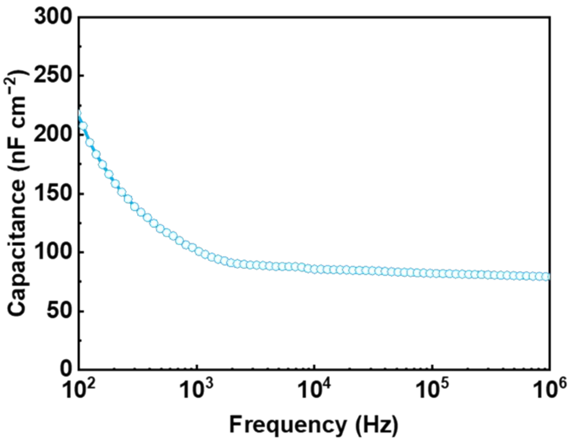

Fig. S9 Capacitance versus frequency (100 Hz~1 MHz) plot of poly-TT 2DP film.

Noting that the capacitance of MIMs at a frequency lower than 10^4 Hz tends to increase, which may arise from electric double-layer (EDL) polarization.[10] In Schiff base condensation reaction, CH_3COOH was added into the system as a catalyst, thus, traces of CH_3COOH might exist in the film. After applying electric field, the free ions moved toward the electrode, leading to the development of EDL polarization and the increase of the capacitance.

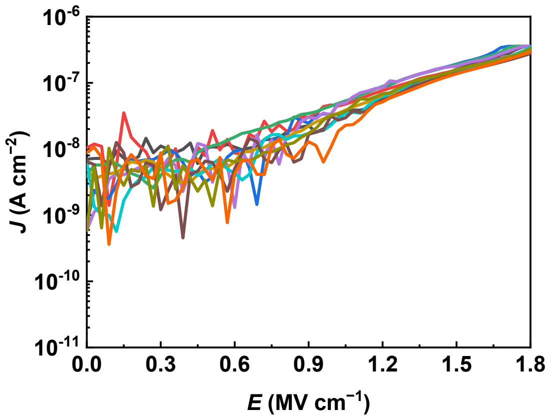

Fig. S10 Leakage current density of 10 MIM devices after heating at 300 °C for 20min.

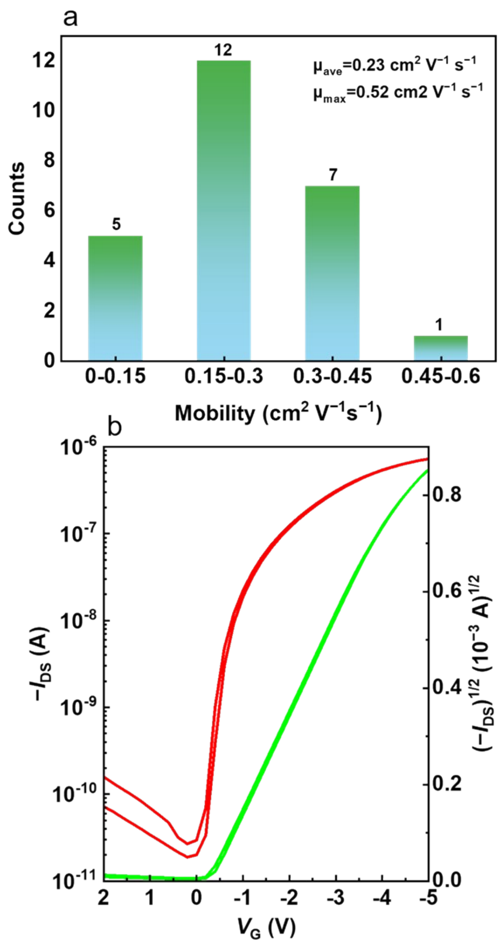

Fig. S11 (a) Histogram of hole mobilities measured from 25 flexible OFETs. (b) Hysteresis characteristics of flexible OFET.

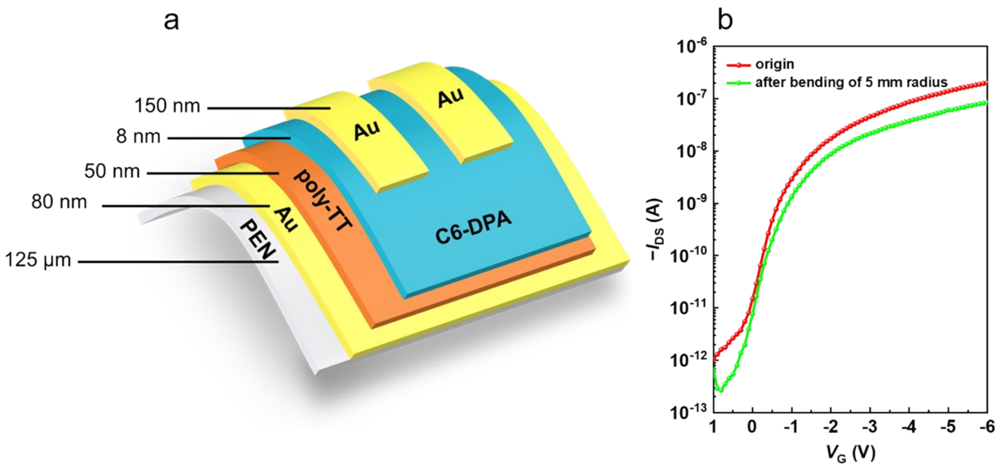

Fig. 12 (a) Scheme of one flexible OFET. (b) Transfer curves for the flexible OFET before and after bending test.

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