

## Supplementary Information

### **Interfacial Spreading for Rapid Formation of Uniform Gate Dielectric Layers on Flat and Curved Substrates for Organic Devices**

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Supplementary Video 1: Whole process for the formation of gate dielectric thin film.

### Supplementary Note 1:

A 5 wt% solution of polystyrene (PS) was prepared using a solvent mixture of chloroform and tetrahydrofuran (THF) in a 4:1 volume ratio. Following thorough stirring, the solution was filtered using a polytetrafluoroethylene (PTFE) syringe filter with a pore size of 0.22  $\mu\text{m}$  to remove any particulates. Subsequently, 10  $\mu\text{L}$  of the filtered solution was carefully dispensed onto the water surface using a micropipette. The dip-coating process was then carried out for the formation of PS thin film under the same conditions previously employed for CL-PVP. When chloroform was used as the sole solvent, the polymer solution demonstrated poor spreading behavior on the water surface. The addition of THF, which is miscible with both water and chloroform, effectively reduced the interfacial tension between the water and chloroform phases, thereby enhancing the spreading of the solution. Post-deposition, residual solvents and water were removed by placing the sample on a hot plate at 70  $^{\circ}\text{C}$  for 10 min, followed by thermal treatment in a vacuum oven at 120  $^{\circ}\text{C}$  for 1 h. Film thickness was measured at nine distinct positions, as illustrated in Figure 3b. While the thickness uniformity was somewhat lower than that observed for CL-PVP, a continuous and relatively uniform PS film was successfully obtained, with an average thickness of approximately 125 nm across all measured positions.

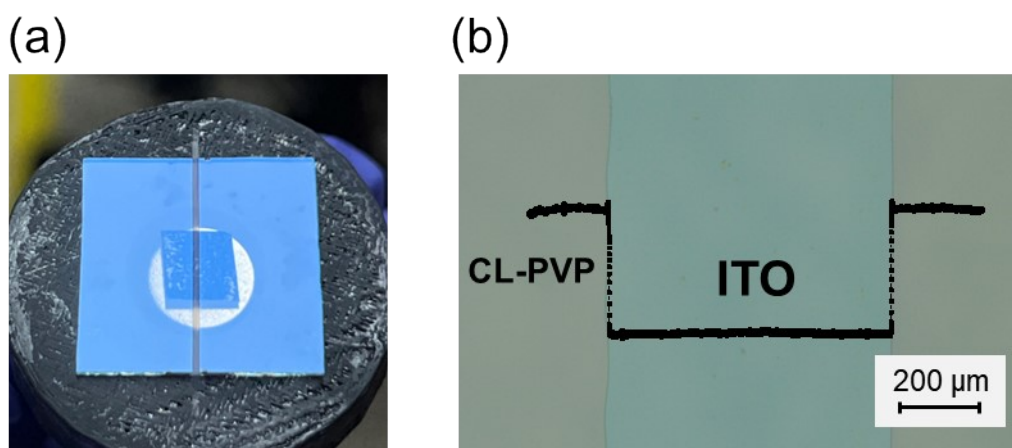


Figure S1. Simple patterning using Scotch Magic Tape: (a) An indium tin oxide (ITO)-coated glass substrate (20 mm  $\times$  20 mm), cleaned up to the isopropanol (IPA) stage, was partially masked with Scotch Magic Tape (500  $\mu\text{m}$  wide) prior to UV/ $\text{O}_3$  treatment. The tape was applied to the regions where patterning was desired. (b) Optical microscopy image and corresponding surface profile of the area where the CL-PVP film was selectively removed by peeling off the tape, which was removed immediately after film transfer, and the sample was subsequently subjected to thermal crosslinking.

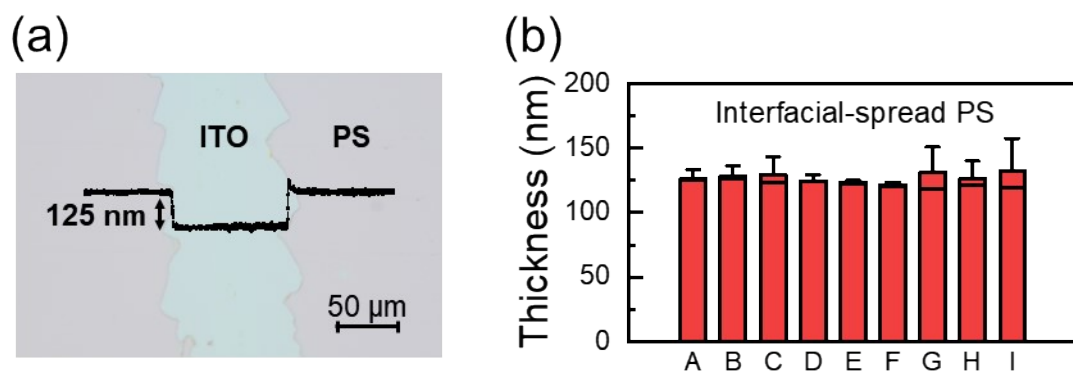


Figure S2. Thickness characterization of interfacial-spread PS film: (a) Optical microscopy image and corresponding surface profile of a scratched area of the substrate, (b) Average film thickness measured at each of the nine positions indicated in Figure 3b for the interfacial-spread PS films.

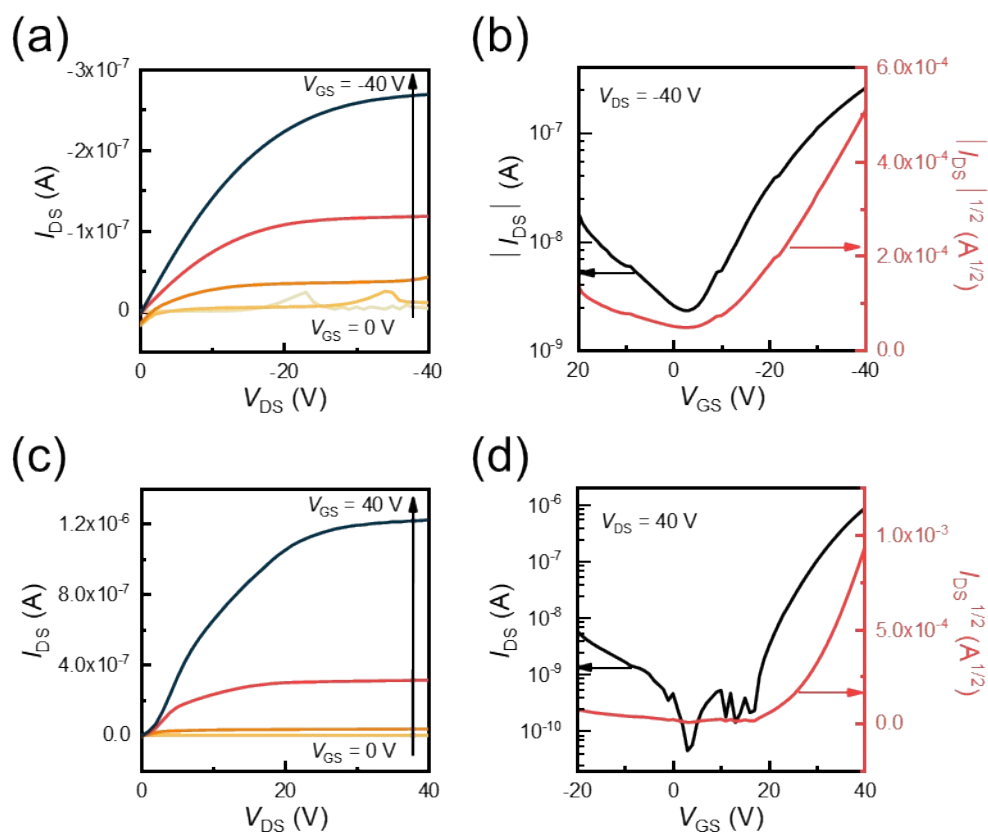


Figure S3. Electrical characteristics of individual OFETs in the inverter circuit: (a) Output characteristics and (b) transfer characteristics of the pentacene-based OFET employing interfacial-spread CL-PVP as the gate dielectric; (c) Output characteristics and (d) transfer characteristics of the PTCDI-C<sub>13</sub>-based OFET with interfacial-spread CL-PVP gate dielectric.