**Supporting Information**

# Inkjet Printing Patterns of Highly Conductive Pristine Graphene on Flexible Substrates

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## 1. Preparation of pristine graphene sheets and ink

Graphite powder (2.0 g) was put into a high-pressure stainless steel reactor installed with a probe sonication system (Bilon-1200Y, Shanghai Bilon Experiment Equipment Co, Ltd, China). The reactor was heated up to 40℃by an electric heating sleeve. $CO_2$was then pumped into the reactor by a manual pump. After the pressure and the temperature reached the presetting ones, the ultrasonic generator started to run for two hours under 120W. The graphite was exfoliated into mono- or few-layer graphene sheets under the coupled effect of the ultrasound and supercritical $CO_2$.

After exfoliation, the exfoliated pristine graphene sheets were mixed in the beaker with 30mL of 0.1wt/v % EC/cyclohexanone solution to make the ink. The resultant suspension was dispersed in ultrasonic bath (40 W) for 30 min to make the graphene be combined further with EC. The large flakes of the suspension was removed by centrifugation at 10000 rpm (～10250×g) for 30 min(TG16-II,Changsha Pingfan Instrument and Meter Co, Ltd, China). The graphene concentration was calculated through the Lambert-Beer law $A / l = \alpha_{660}\ C$ (l is the cell length) and the absorption coefficient$\alpha_{660}$= 2460 L $g^{-1}$ $m^{-1}$. The absorbance of the as-prepared graphene suspension was measured by a UV-Vis spectrophotometer (PC 756, Shanghai Spectrum Instruments Co., Ltd, China) at $\lambda$ = 660 nm. The graphene concentration of the ink was adjusted to be 1.0 mg/mL (0.72 wt % solids content).

## 2. Characterization of graphene sheets (main text Figure 2)

Several milliliters of the as-prepared graphene supernatant were deposited onto a $Si/SiO_2$ (300nm) wafer and annealed at 400℃ for 30 min to remove the solvent and

EC. Atomic force microscopy (AFM) images were obtained using an Environment Control AFM system in tappingmode with a SII Nanonavi E-Sweep Model.The image was collected with 350 nm×350 nm scans, and the thickness of all the pieces (234 flakes) in the AFM image was determined using Nanonavi Analysis software.Raman spectrum was obtained with a RenishawinVia Reflex Raman System equipped with a 532 nm laser source anda 100×objective, with an incident power 5 mW.

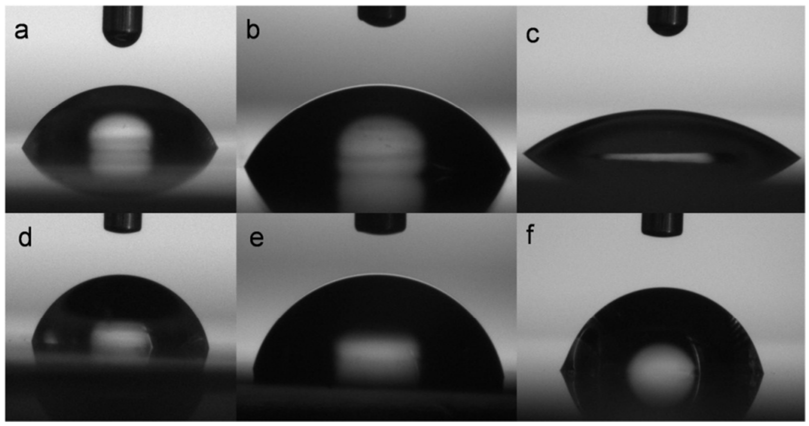
**3. The jetting performance of the graphene ink**

A video was recorded using the Drop Watcher system of the Dimatix (DMP 2800) printer. The video of jetted drops showed the jetting performance of the graphene inks. The as-formulated ink can provide well-directed and constant jetting out of all nozzles at an even velocity. The spherical drops form after ～ 300 μm.

Video1

**4.Surface modification of substrates**

Polyimide (PI, DuPont Kapton 125μm) and glass slide substrates were cleaned with acetone in an ultrasonic bath for 30 min, and rinsed with deionized water. Poly(ethylene terephthalate) (PET,150μm)substrates were cleaned similarly with ethanol. All the cleaned substrates were dried under a stream of $N_2$, and then submerged into HMDS solvent for one hour. The HMDS-coated substrates were rinsed first with2-propanol, and then dried with a stream of $N_2$. The contact angles of the HDMS-treated substrates were measured using an optical contact angle measurement system (DSA30, KRÜSS GmbH Co., Germany) to characterize the wetting property of the substrates. The contact angles of untreated PET, PI , and glass slide are ~68.2°, ~66.1°, and 38.4°, respectively. The contact angles of the HMDS-treated PI , PET, and glass slide are ~77.2°, ~77.9°, and 77.8°, respectively.




FigureS1.The contact angle images of water drops dispensed on before and a) PET (68.2° ) , b) PI (66.1° ), and c) glass slide (38.4° ) after HMDS- treated d) PET (77.2° ) , e)PI(77.9° ), and f) glass slide (77.8° ) substrates.

**5. Transmittance spectra of printed graphenepatterns**

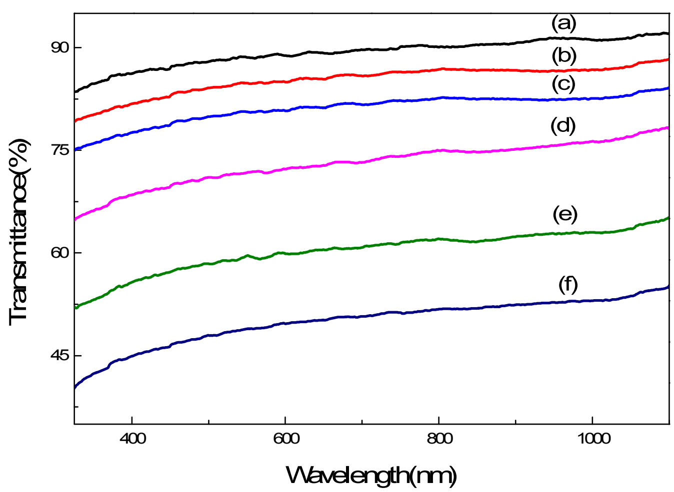



Figure S2. Transmittance spectra of the 6 printed graphene patterns on glass sildes after 3, 6, 10,20, 30 and 40 printing passes from (a) to (f).