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3-D Conformal Graphene for Stretchable and Bendable Transparent

Conductive Film

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1. Experimental section

1.1 Preparation of Graphene Samples.

First of all, the copper foils with periodic micro structures were prepared by lithography and wet etching, keeping the depth of the structure for ~2µm and the period of the structure for ~20µm, then cleaned for use. We selected a tube-style chemical vapor deposition(CVD) system to deposit graphene(Gr) films on cleanly structured copper foils (S-Cu) substrates (Gr/S-Cu) using mixture gas of hydrogen (purity:99.999%) and methane (purity:99.999%). Under atmospheric pressure, mixture gas of 25% methane in hydrogen was introduced into the chamber after substrate temperature (T) increased up to 1000 °C. The graphene growth maintained for 10-30 minutes, and then the samples were cooled to room temperature rapidly.

1.2 Preparation of Conformal Graphene.

The PDMS polymer was prepared by mixing a silicone elastomer base with a curing agent with a ratio of 10:1. Then the as-prepared PDMS was coated onto the surface of Gr/S-Cu, and was dried on the hot plate at 80 °C for 180min. A aqueous solution of ferric nitrate solution ($Fe(NO_3)_3$) was used to sufficiently dissolve S-Cu without leaving any residue overnight at room temperature. After cleaning, the perfect graphical surface structure was copied from micro-structured copper wafers, and presented in the inverted PDMS/Gr, which was named as confromal graphene (CGrs/PDMS).

1.3 Characterization.

The as-grown samples were directly characterized by a Renishaw in Via Manual WiRE3.4 Microscopes Raman Spectrometer with the laser excitation energy of 532nm. Atomic Force Microscopy (AFM) characterization of structured copper foils and CGrs were conducted under a Dimension Icon equipment of Bruker Corporation. Transmittance of CGrs, PDMS and CGR/PDMS were measured by Ultraviolet spectrophotometer. The sheet resistance of CGrs was tested under a self-assemble electrochemical workstation system at ambient temperature. The micro-photographs were got by an electron microscope.

2. Figures



Fig.S₁ The optical microscope images of large-scale graphene. (a) Conformal graphene film grown on structured copper wafers, (b) Conformal graphene film transferred on transparent and flexible PDMS. The both images are the magnifying LOGO in Fig.1b and 1c.

Fig.S₁ shows completely inverted LOGO patterns on Fig.S_{1a} and Fig.S_{1b}. The distinct patterns on Fig.S_{1b} shown on the surface of CGrs/PDMS suggest the integrality and advisability of this conformal transfer method.



Fig.S2 The SEM images of large-scale graphene (a) grown on structured copper wafers and (b) transferred on transparent flexible PDMS.

The consistent micro-patterns on Fig.S_{2a} and Fig.S_{2b} mean that it is a perfect and practical transfer method for the production of CGrs.



Fig.S3 Transparency of large-area CGrs layers transferred on PDMS, the blue, red and black lines represent the transmittance of CGrs/PDMS, structural PDMS substrate and relative CGrs, respectively.



Fig.S₄ Electrochemical workstation electrical measurement on conformal graphene films shows the line function relationship of current and potential (I-V).

The typical I-V characteristics plots are shown in Fig.S₄. It is found that the current increases linearly with voltage from 0 V to +5 V, which could be used to estimate the conductivity of graphene films.



Fig.S₅ Resistance (black line) and Gauge Factor (blue line) of a 1×1 cm² CGr film on PDMS substrate stretched along the x direction. (a) monolayer CGr, (b) 2 layers CGr, (c) 3 layers CGr and (d) few-layer CGr correspond to growth times of 2, 5, 10, and 20min, respectively.

Fig.S₅ shows the strain tension and strain sensitivity of the CGrs/PDMS with different numbers of stacked layers (1, 2, 3 and few) on longitudinal condition, using the instant resistance and Gauge factor, respectively. For the stretching test on longitudinal condition, with the increasing numbers of stacked layers, CCrs/PDMS show clearly increased strain tension on the black lines. The instant resistance shows a straight line-like type on the front half part of the strain tension for 0-10%, 0-20%, 0-20% and 0-40% of 1, 2, 3, and few layers CGrs/PDMS, respectively. And the remaining instant resistances of four samples on the latter part of their corresponding strain tension show zooming trends with increscent resistance interval. However, strain sensitivity (blue lines) of the thickening of CGrs express inverse rules. The opposite trend of resistance and Gauge factor on Fig.S₅ reveal that these stacked layers of graphene films were another crucial factor for the stretching capabilities of 3D conformal samples. The reason for this case: compared with monolayer sample, on the same tensile tests, the thicker graphene films exhibit greater extensibility but weaker contact response on tensile direction.