Supplementary Information

Methane as an effective hydrogen source for single-layer graphene synthesis on Cu foil by plasma enhanced chemical vapor deposition

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1. ICP-CVD system

1.1 Detailed geometry

Fig. S1a shows a schematic diagram of the inductively coupled plasma chemical vapor deposition (ICP-CVD) system. At the top of the chamber, RF coil is mounted below the inlet ports which allow gas flow into the chamber. The diameter of RF coil is about 25 cm and the center of the coil is located 35 cm above the top surface of the graphite substrate holder. Plasma is generated by applying 13.56 MHz RF bias power to the RF coil. No bias voltage is applied to the substrate stage, because it is known that the electric field aligned perpendicular to the surface prevents planar growth of graphene and results in vertically standing graphene sheets.^{1,2}

1.2 Substrate heating by plasma

To investigate the heating effect of substrate by RF plasma, the substrate temperature is monitored under different plasma powers of 50 and 400 W. It is conducted without filament heating of substrate and the temperature is measured by a K-type thermocouple which is attached to substrate. When the plasma power is 400 W, the temperature increases only

about 3 °C after 12 minutes (Fig. S1b). When the plasma power is reduced to 50 W, the temperature does not change even after 14 minutes. Considering the plasma powers (< 200 W) and the growth times (\leq 3 minutes) used in the graphene growth, we conclude that the substrate heating effect by RF plasma is negligible in our system.



Fig. S1 (a) Schematic diagram of ICP-CVD system. Remote plasma is generated above the substrate. (b) Substrate heating effect by plasma under different plasma powers of 50 and 400 W.

2. Differential-pumping technique for mass spectra measurement

A residual gas analyzer (RGA 100, Stanford Research Systems) is attached at the main chamber to investigate the mass spectra of discharged species. The high pressure of $\sim 10^{-2}$ Torr during the graphene growth is not suitable for the measurement because the RGA system requires a high vacuum ($<10^{-4}$ Torr) environment. While the pressure could be lowered to the desired level by reducing the gas flow, plasma could not be generated in this case; the pressure should be higher than 10^{-3} Torr to generate plasma. To overcome these two conflicting issues, we employed a differential pumping technique³ in our ICP-CVD system by mounting an aluminum aperture (inner hole diameter ~5 mm) underneath the plasma reactor as shown in Fig. S2. Due to the highly reduced pumping conductance between the plasma reactor and the main chamber, we are able to keep ~ 10^{-2} Torr in the plasma reactor and ~ 10^{-4} Torr in the main chamber under proper gas flows (Ar/CH₄ = 5:5 sccm). We investigated the mass spectra of discharged species in this configuration.



Fig. S2 Illustration of ICP-CVD system with an aluminum aperture mounted underneath the plasma reactor for differential pumping.

3. Transport properties of a single-domain graphene device

A single-domain graphene device is fabricated using mechanically exfoliated graphene from HOPG. The channel length and width of this device are 1 and 3 μ m, respectively. The dependence of sheet conductance on the applied gate voltages is shown in Fig. S3. We calculated an electron mobility of ~3,640 cm²V⁻¹s⁻¹ at $n = -1 \times 10^{12}$ cm⁻².



Fig. S3 Gate voltage dependent conductance of exfoliated graphene films from HOPG.

References

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