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Supporting Information

Three Step Fabrication of Graphene at Low Temperature by Remote Plasma Enhanced Chemical Vapor Deposition

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Due to the thermal instability of cyclobenzene structure, we achieved graphene films at low temperatures (300 ^{o}C-600 ^{o}C) by remote plasma enhanced chemical vapor deposition (RPECVD). The sketch map of CVD growth equipment for synthesis of graphene by naphthalene is shown in Fig. S1.

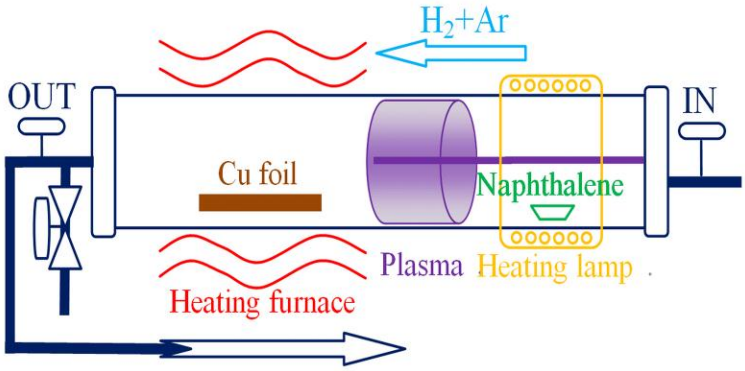

Fig. S1 The modified RPECVD growth equipment for synthesis of graphene by naphthalene.

For graphene RPECVD process, defects in graphene gradually increase with the decrease of growth temperature. The defects are partly attributed to the insufficient catalysis of Cu at very low growth temperature, which causes the incomplete dehydrogenation of carbon related radicals. The representative C1s spectra of XPS for graphene samples are shown in Fig. S2. The C 1s peak was shown at 284.7 eV,[1] which could be divided into two sub peaks. The main peak of graphite-like sp^2 C is located at 284.4 eV. The other small peak at 285.2eV is clearly visualized, which correspond to C-H bonds, respectively, originating from the C-H sp^3 structure and amorphous phase.[2] As shown in Fig. S2a, no obvious C-H peaks are observed

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for graphene synthesized at 500 °C. The C-H peak increased obviously with the decrease of growth temperature. It is also in agreement with increased structural disorder due to the disruptionsin the sp^2 carbon framework. Such incomplete dehydrogenation would decorate the structure of graphene sheet and introduce defects in the graphene films.

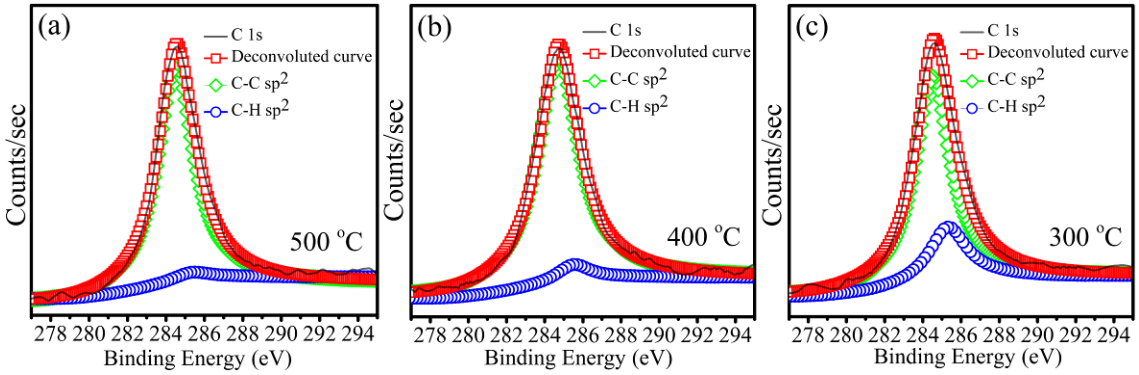

Fig. S2 C1s XPS spectra at different growth temperatures. (a) 500 °C; (b) 400 °C; (c) 300 °C.

High-resolution optical images of continuous graphene films synthesized at 300-600 °C were introduced to further characterize the surface topography of graphene transferred on SiO_2/Si substrate. It could be clearly observed that most positions of graphene synthesized at low temperatures are monolayer grapheme (see in Fig. S3). Meanwhile, it was also found that the percentage of monolayer reduced obviously by the decrease of growth temperature, which resulted in the local aggregation of carbon species and induced the nucleation of multilayer domains and amorphous phase.

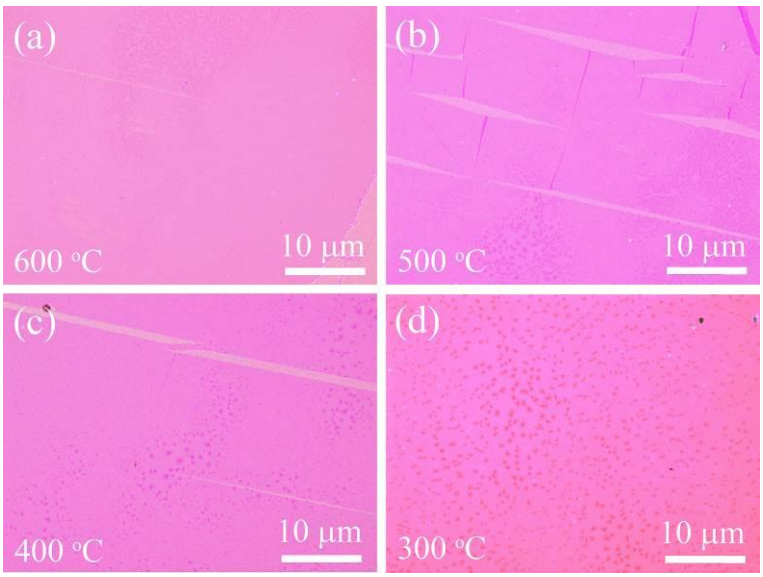

Fig. S3 High-resolution optical images of transferred graphene films grown at 300-600 °C.

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Six carbon sources (benzene, naphthalene, phenanthrene, chrysene, perylene and coronene) which contain 1-6 benzene rings were introduced as the appropriate precursors to grow high quality graphene at low temperature 600 ^{o}C. As shown in Fig. S3a, Raman spectra indicate the good growth control and relative high quality of the obtained continuous graphene films. Light transmittance and optical micrographs of the transferred graphene films on quartz substrates are shown in Fig. S3b. The 96.5-96.7% transmittance ratio and the continuous optical images confirm the synthesis of uniform single-layer graphene films (shown in Fig. S3b), which is consistent with the Raman results.

It was found that The H-C_α and H-C_β bonds in naphthalene molecular are more instable than the H-C bonds in benzene,[3] which is helpful for promoting the formation of graphene at low temperatures. On the other hand, phenanthrene, chrysene, perylene and coronene are high-cost and not easy to be obtained. Due to relative high sublimation temperature of these four carbon sources, they must be heated to higher temperature (even higher than graphene growth temperature) to obtain enough gaseous precursors, which induce the nucleation and growth of graphene domains. Base on the consideration for low-cost fabrication and application of graphene, we determined naphthalene as the most suitable carbon source.

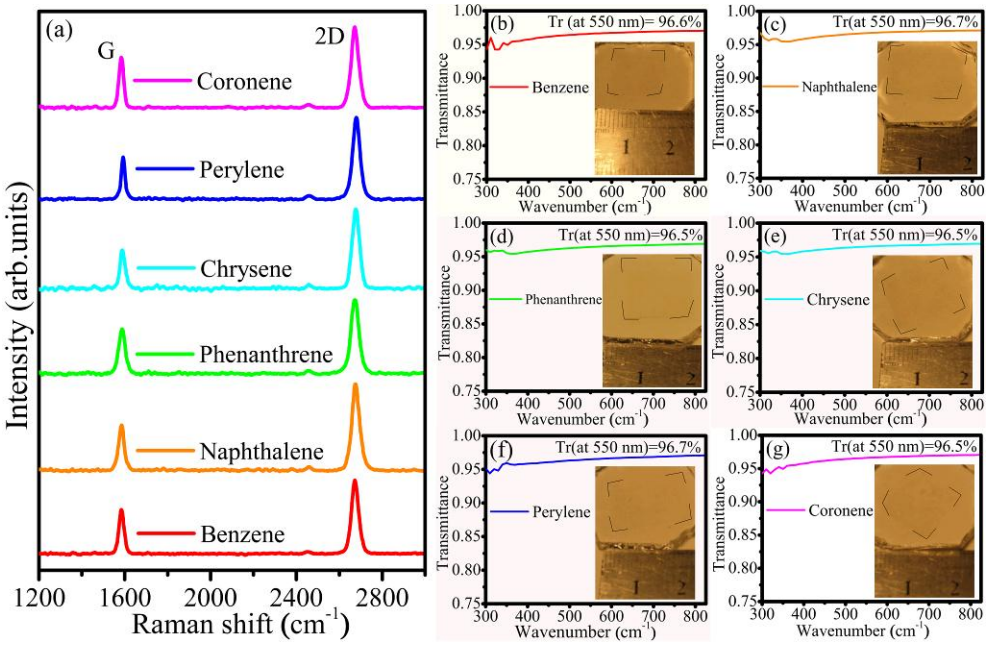

Fig. S4 (a) Raman spectra of graphene synthesized at 600 ^{o}C using benzene, naphthalene, phenanthrene, chrysene, perylene and coronene as precursors; (b) Light transmittance and

optical micrographs of the transferred graphene films on quartz substrates.

The synthesis parameters during the RPECVD growth play a key role for high quality continuous films. Fig. 4a shows the Optical image of transferred continuous graphene films on SiO_2/Si substrates synthesized at 600 ^oC. It was found that graphene growth at low temperature is not self-limiting under high concentration of carbon reactants (C_r). The growth rate could be accelerated by increasing C_r and obtained continuous graphene sheets. But it also promotes growth of the multilayers. Typical Raman spectrum of the resulted sample at different site verified the non-uniform of graphene layers (see in Fig. S4b). Additionally, D-band (~1350 cm^{-1}) intensities in the Raman spectra were higher at multilayer sites. It is possibly due to the local aggregation of carbon reactions, resulting in the formation of amorphousphase on the surface.

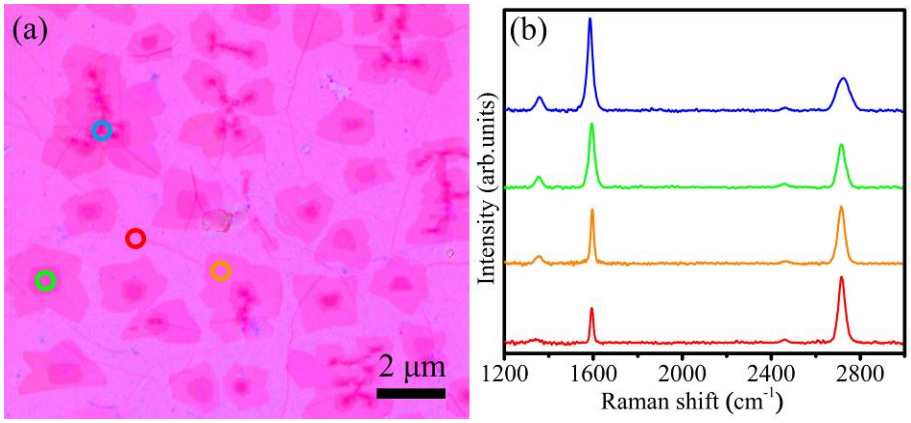

Fig. S5 (a) Optical image of the graphene transferred to Si/SiO_2 substrate synthesized at 600 ^oC; (b) Raman spectra of RPECVD graphene samples at different sites.

References:

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