Supporting Information

Graphene crystals grown on the SiO_2/Si substrate at low temperature by controlling the initial nucleation and suppressing the subsequent multiple nucleation

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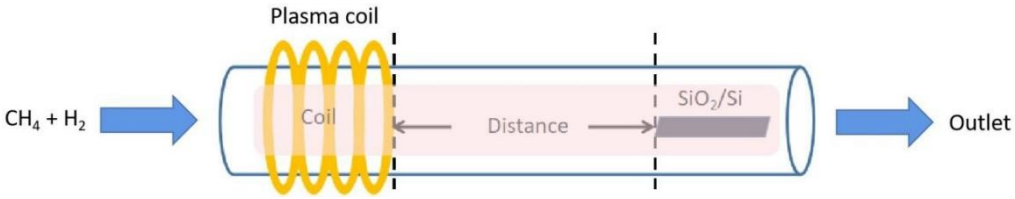

Fig. S1 Schematic illustration showing the PECVD setup to grow large graphene crystals on the SiO_2 substrate.

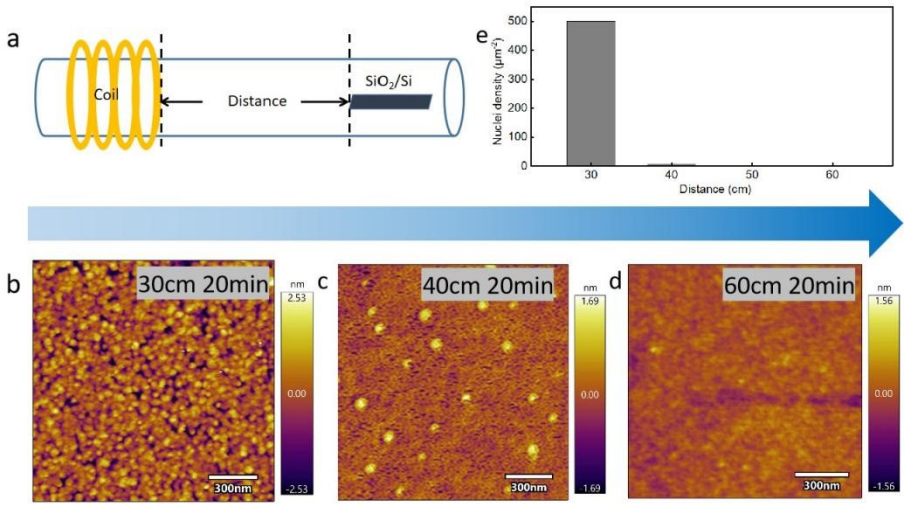

Fig. S2 (a) Schematic diagram showing the distance between the coil and SiO_2/Si substrate. (b-d) AFM height images of graphene nucleation at a distance of 30 cm, 40 cm and 60 cm, respectively. (e) Histogram showing the relationship between the distance and the nucleation

density. Other parameters were kept the same: 3.5 sccm CH_4 and 1 sccm H_2, 20 W, 30 Pa, 20 min. With various distance, the concentrations of active carbon-containing species around the substrate can be very different, which will greatly affect the growth behavior of graphene. At a distance of 30 cm, the shortest distance allowed in our PECVD setup, graphene seeds with a very high density of ~ 500 μm^{-2} were formed on SiO_2 surface, which almost cover the whole SiO_2 surface (Fig. S2a). This high density shuts down the door of growing large graphene crystals and instead nanographene would be probably achieved. At a longer distance of 40 cm, the density is dramatically decreased to ~ 12 μm^{-2} (Fig. S2c) due to the decreased concentrations of active carbon-containing species near SiO_2 surface. At a further longer distance of 60 cm, no graphene seeds is even formed after growth (Fig. S2d) because of the insufficient supply of active carbon-containing species for graphene to nucleate. Empirically, 40 cm is adopted as the optimized working distance in this work.

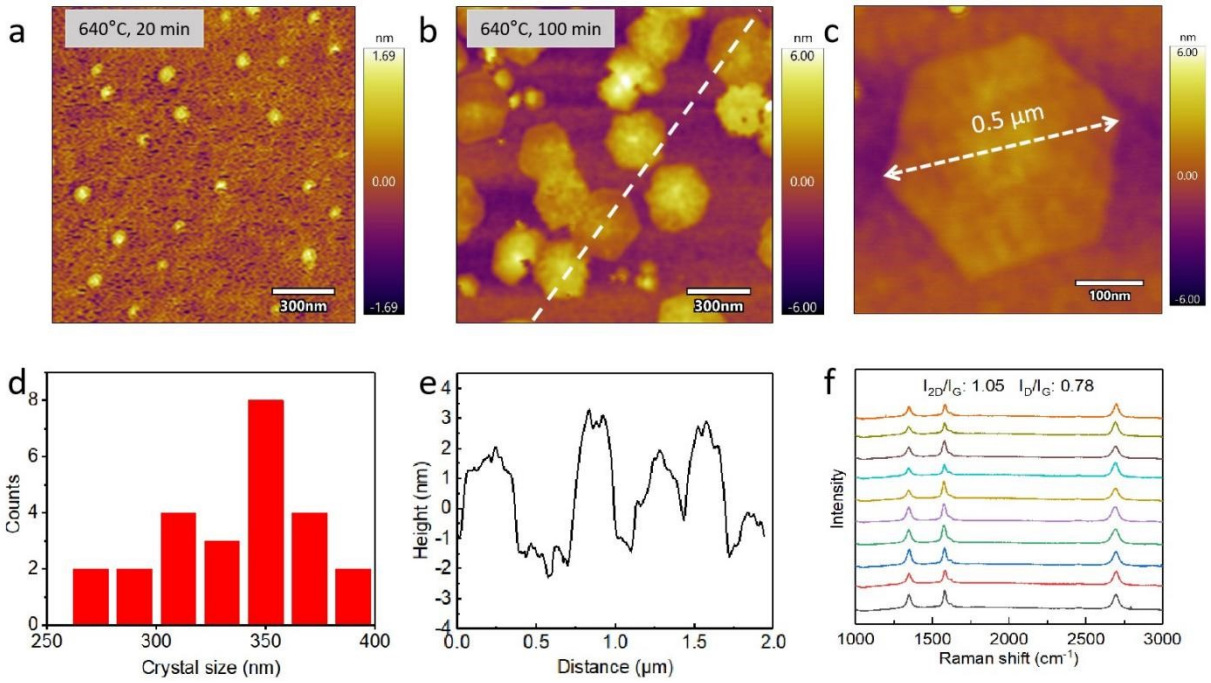

Fig. S3 AFM height images of SiO_2 surface after growth at 640°C for 20 min (a) and 100 min (b,c). (d) The histogram of statistical distribution of graphene crystal size from sample in (b). (e) The height profile along the white dashed line in (b). (f) Raman spectra of sample from 10 random points in (b). After growth at 640°C for 20 min, small graphene nuclei are formed on the SiO_2 surface with a density of ~ 12 μm^{-2} and an average size of about 70 nm (Fig. S3a). As the

growth time is further extended to 100 min, these nuclei grow into larger crystals without increasing nucleation density (Fig. S3b). It indicates that the subsequent repeated nucleation during long-time growth is also effectively suppressed under this growth condition. These crystals have an average size of 345 nm (Fig. S3d) and the largest size can reach 0.5 μm (Fig. S3c). However, the nonuniform color contrast in Fig. S3b reflects the giant variations of layer thickness of graphene, as shown by the height profile in Fig. S3e with a large height fluctuation of 3~5 nm, much thicker than the graphene crystals grown at 620°C (Fig. 6). Raman data in Fig. S3f also confirms that thicker graphene is achieved in this case, based on a smaller average I_{2D}/I_G of ~ 1.05 than that of 1.58 at 620°C.

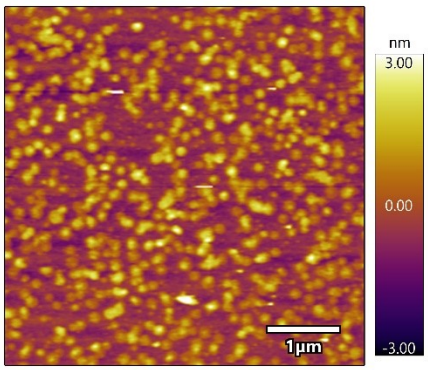

Fig. S4 AFM height image of SiO_2 surface after growth at 660°C and under 20 W. At higher temperature above 640°C, even the plasma power is low, the nucleation density increases fast with time, due to the enhanced nucleation rate at higher temperature

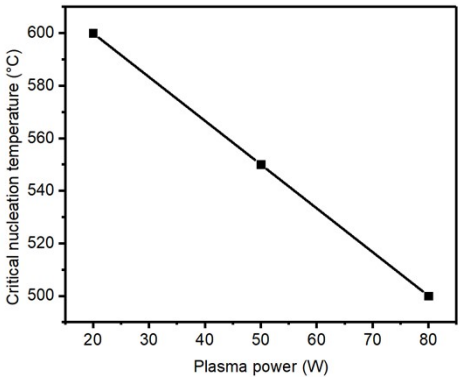

Fig. S5 The relationship between plasma power and critical nucleation temperature. It seems that the critical nucleation temperature decreases with the increase of plasma power.