

## *Supporting information*

# Synthesis and morphology transformation of single-crystal graphene domains based on activated carbon dioxide by chemical vapor deposition

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## 1 Main physical and chemical properties of catalyst for CO<sub>2</sub>:

**Table S1.** The main physical and chemical properties of the Ni/Al<sub>2</sub>O<sub>3</sub> catalyst for CO<sub>2</sub>.

item	content
Chemical component	NiO: 18±1.0%; TiO <sub>2</sub> : 6-8%; Al <sub>2</sub> O <sub>3</sub> :75±2%
Specification	Φ4.5~5.5 mm
Bulk density	0.9-1.0 g/cm <sup>3</sup>
Chemical property	CO <sub>2</sub> +H <sub>2</sub> $\xrightarrow{\text{catalyst}}$ CH <sub>4</sub> +H <sub>2</sub> O

## 2 Experiment parameters of graphene synthesis:

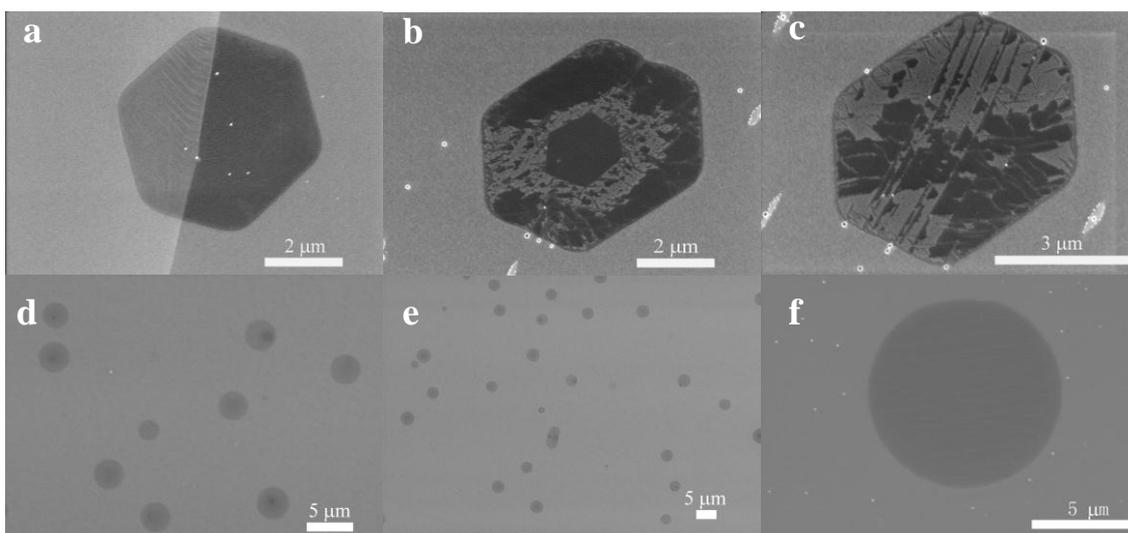
**Table S2.** The experiment parameters of the graphene growth based on activated CO<sub>2</sub> at ambient pressure.

Carbon source	Experiment number (#)	The flow rate ratio of CO <sub>2</sub> to H <sub>2</sub> (sccm)	Growth temperature (°C)	Growth time (min)
CO <sub>2</sub>	1	1:200	1000	30
	2	3:200		
	3	5:200		
	4	10:200		
	5	15:200		
	6	20:200		
	7	30:200		

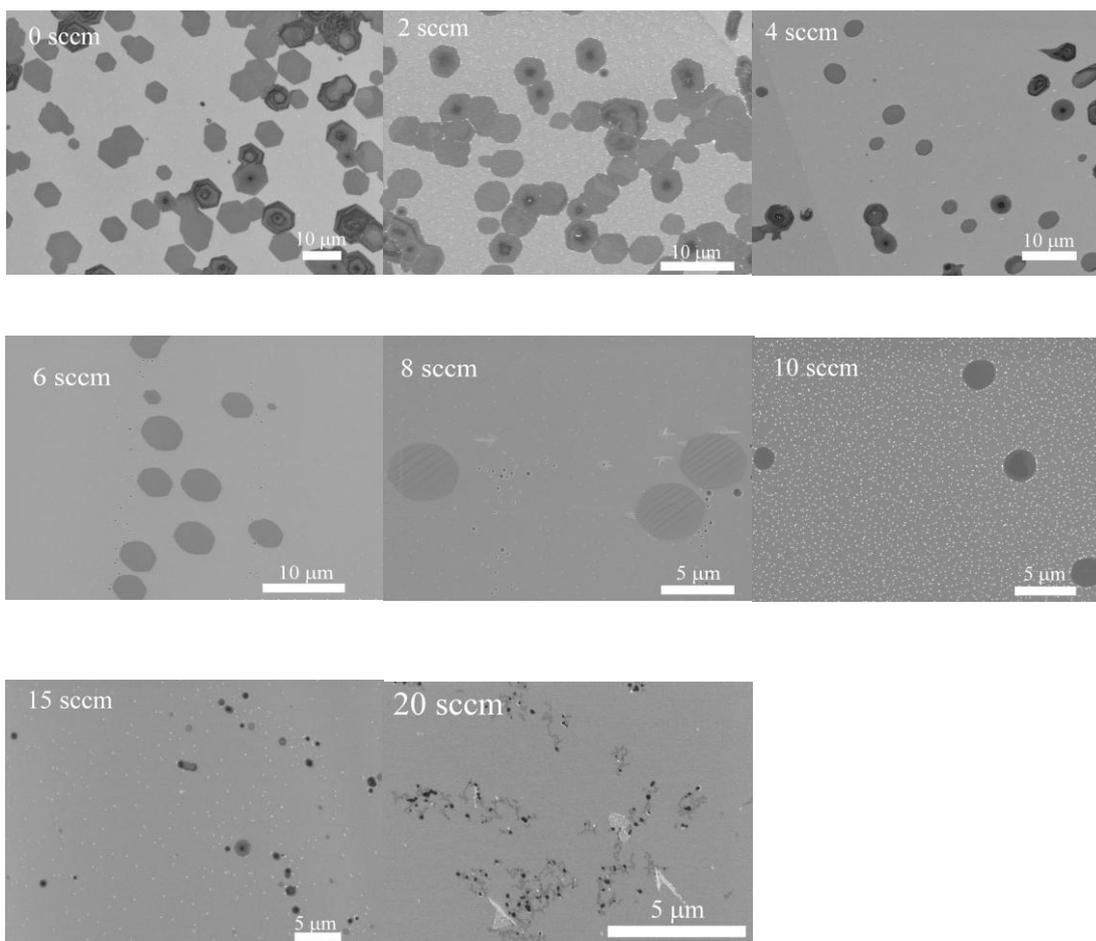
**Table S3.** The experiment parameters of the graphene growth based on a mixture of CH<sub>4</sub> and H<sub>2</sub> diluted by CO<sub>2</sub> at ambient pressure.

Carbon source	Experiment number (#)	The flow rate ratio of CH <sub>4</sub> to H <sub>2</sub> to CO <sub>2</sub> (sccm)	Growth temperature (°C)	Growth time (min)
CH <sub>4</sub>	1	5:200:0	1000	30
	2	5:200:2		
	3	5:200:4		
	4	5:200:6		
	5	5:200:8		
	6	5:200:10		
	7	5:200:15		
	8	5:200:20		

### 3 Supplementary SEM characterizations

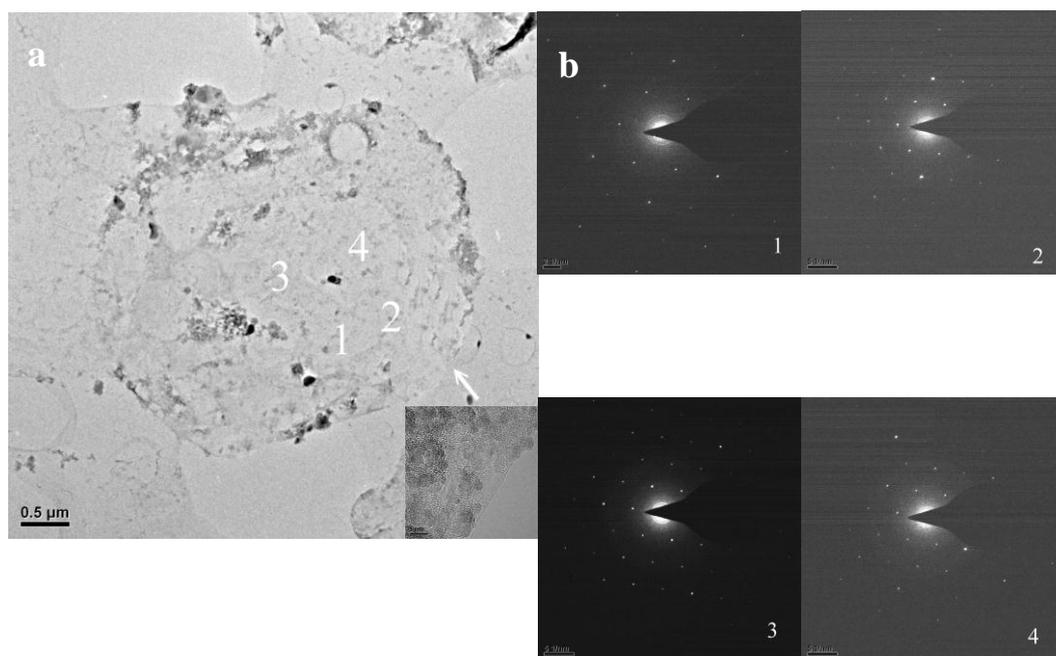


**Figure S1.** (a-c) High-magnified SEM images of graphene domains grown on Cu surfaces using a mixture of 5 sccm CO<sub>2</sub> and 200 sccm H<sub>2</sub>. (d-f) Low and high-magnified SEM images of graphene domains grown on Cu surfaces using a mixture of 20 sccm CO<sub>2</sub> and 200 sccm H<sub>2</sub>.



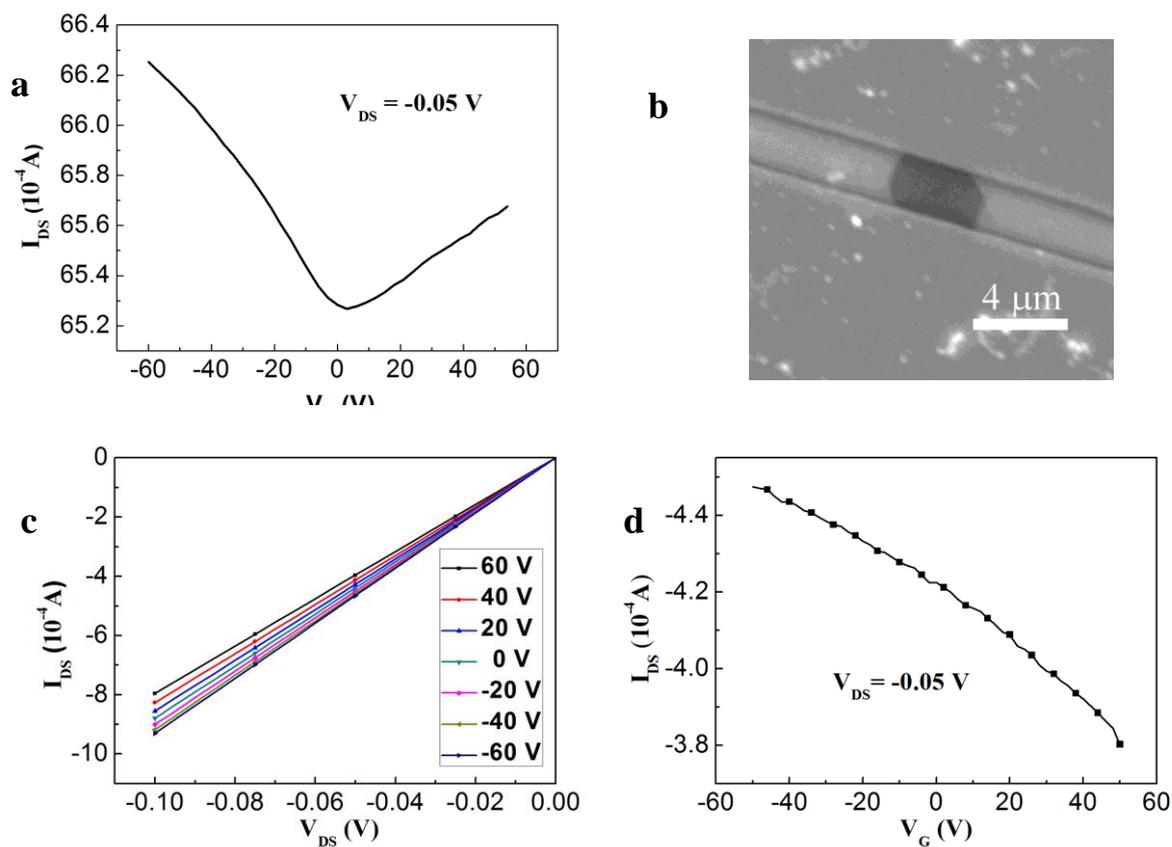
**Figure S2.** SEM images of graphene domains on copper foils using a mixture of 5 sccm  $\text{CH}_4$  and 200 sccm  $\text{H}_2$  diluted by different flow rates of  $\text{CO}_2$  (also see Table S2). The growth time was 30 mins.

#### 4 TEM characterizations of round graphene domain.

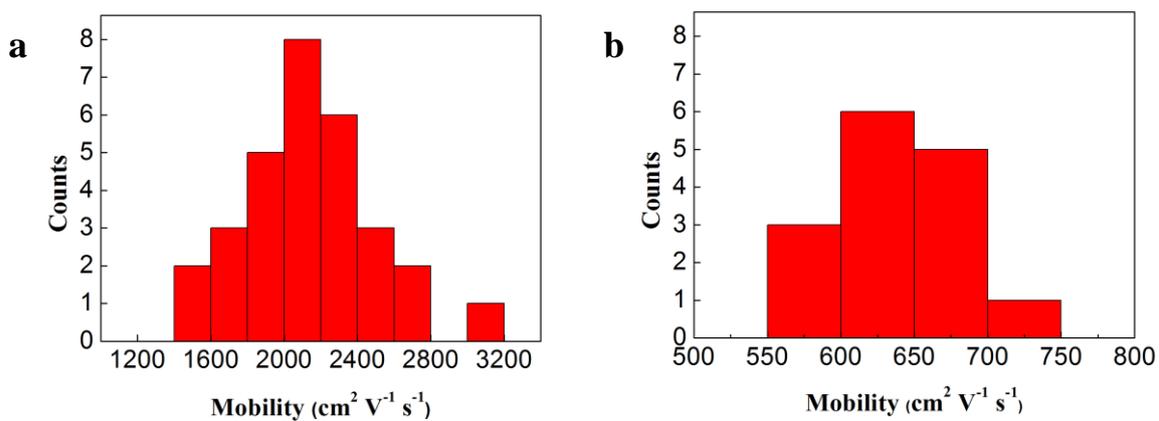


**Figure S3.** TEM characterization of round-shaped graphene. (a) Low-magnification TEM image showing an individual round graphene domain. The inset shows the high resolution TEM image corresponding to the region marked by white arrow in (a) showing single layer. (b) The corresponding SAED on the regions marked by numbers in (a) respectively.

## 5 Details of hexagon graphene devices performance



**Figure S4.** Electrical properties of hexagon graphene at room temperature. (a) Transfer characteristics of a device at  $V_{DS} = -0.05$  V in  $N_2$  and the neutral (Dirac) point is at  $\approx 3$  V. (b) A top view SEM image of the hexagon graphene device with a channel length of  $\sim 2.2 \mu\text{m}$  and channel width of  $\sim 4 \mu\text{m}$ . (c)  $I_{DS}$ - $V_{DS}$  characteristics for the hexagon graphene device in air at varied  $V_G$ . (d) Transfer characteristics of the hexagon graphene device at  $V_{DS} = -0.05$  V in air.



**Figure S5.** Histogram of mobility distribution from 15 CO<sub>2</sub>-derived graphene devices (8 hexagon graphene devices and 7 round graphene devices). (a) Histogram of hole mobility distribution from 15 devices in air and N<sub>2</sub>. (b) Histogram of electron mobility distribution from 15 devices in N<sub>2</sub>.