Supporting Information

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Nucleation density calculation

N/S is defined as the nucleation density, where ***N*** is the number of the graphene grains on copper foil after growth and ***S*** is the area of the copper foil.

Average growth rate calculation

The average growth rate ***L/2t*** was used as the growth rate, where ***L/2*** is half of the largest graphene grain size and ***t*** is the total growth time.

Table S1. Cu hill height and graphene grain size as a function of growth time.

Figure	Temperature(°C)	CH_4 (sccm)	H2 (sccm)	Growth time (h)	Cu hill height (μm)	Graphene grain size (μm)
3a	1077	0.1	100	0.5	1.405	68.934
3b	1077	0.1	100	1	3.992	248.793
3c	1077	0.1	100	1	9.550	692.551
		0.15		1.5		
3d	1077	0.1	100	1	11.990	1088.434
		0.15		2		
3e	1077	0.1	100	1	0	1478.555
		0.15		3		

Table S2. Growth rate of each growth stage with different CH_4 concentration.

Figure	Temperature(°C)	CH_4 (sccm)	H2 (sccm)	Growth time (h)
4a	1077	0.1	100	0.5
4b	1077	0.1	100	1
4c	1077	0.1	100	1
		0.15		1.5

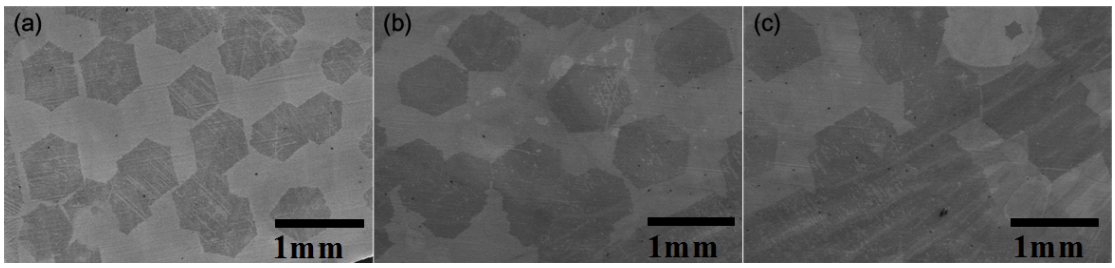

Fig. S1- (a-c) SEM images of as-grown millimeter scale graphene grains grown on Cu foils.

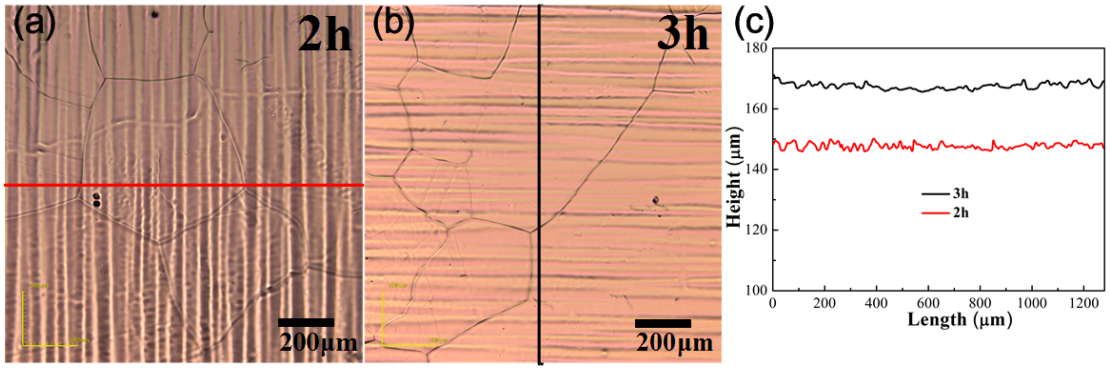

Fig. S2- (a-b) 3D laser microscopy image of Cu surface after running the reactors without carbon feedstock for 2h and 3h, respectively. (c) The profile line of Cu surface morphology denoted with red line in (a) and dark line in (b). The morphology's amplitude is within 2μm.

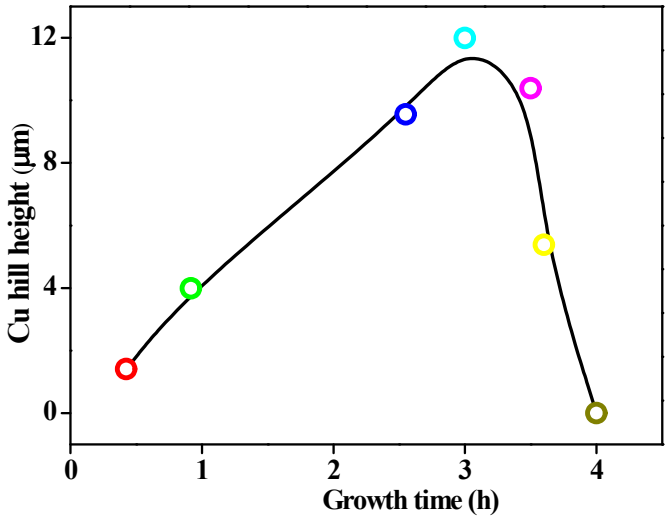

Fig. S3 Cu hill height as a function of growth time. Cu hill height increases linearly before the grains emerge due to the severe Cu evaporation. Graphene grains emerge at the growth time of 3h. Consequently, the Cu hill turns flat owing to the thermal motion.

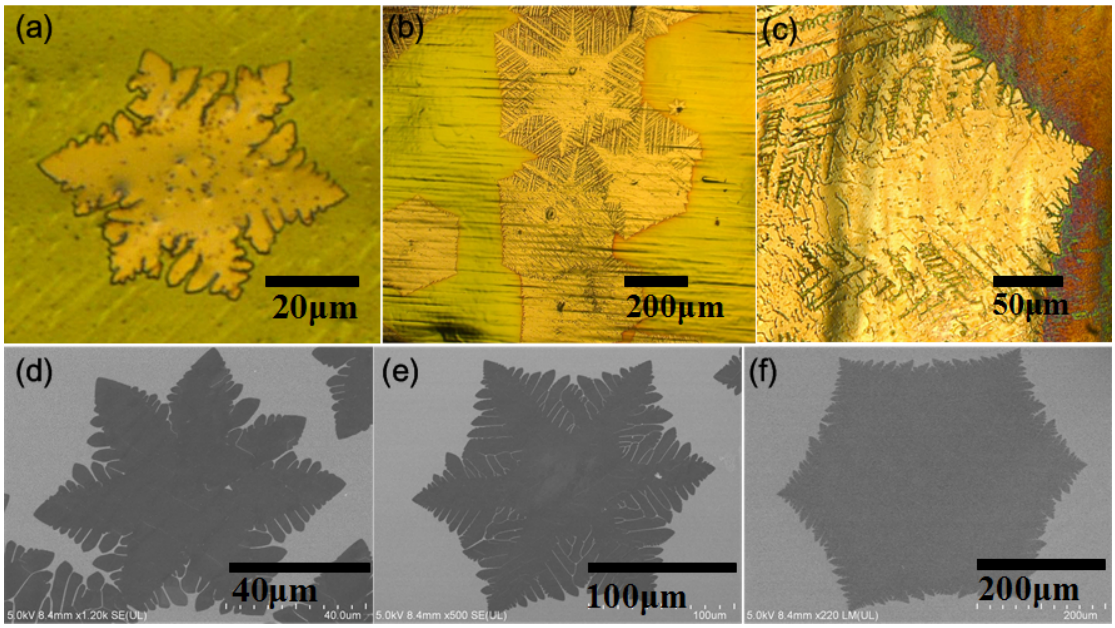

Fig. S4- (a-c) Optical images of graphene grains grown on Cu foils after oxidation treatment. (d-f) SEM images of graphene grains grown on Cu foils. The dendritic structures at the edge can be clearly identified, but the inter-branch boundaries are healed partially.