

Electronic Supplementary Information

Epitaxial nucleation of CVD bilayer graphene on copper

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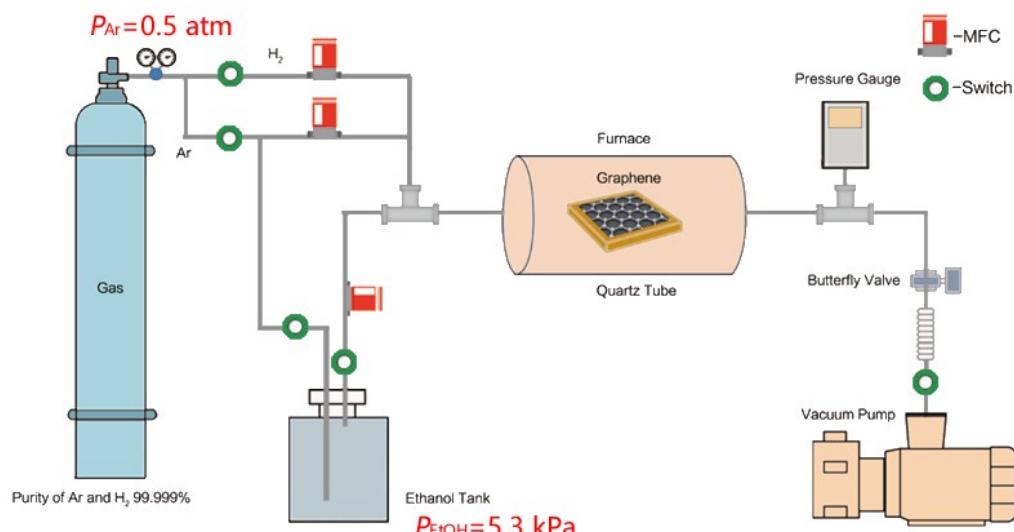


Figure S1. An ethanol-based epitaxial CVD system for BLG growth. The pressure of Ar flow was controlled by the release valve to be 0.5 atm, and the ethanol temperature was maintained at 19 °C, whose saturated vapor pressure is 5.3 kPa. Therefore, the flow rate ratio between Ar and ethanol in a bubbled ethanol flow is approximately 10:1.

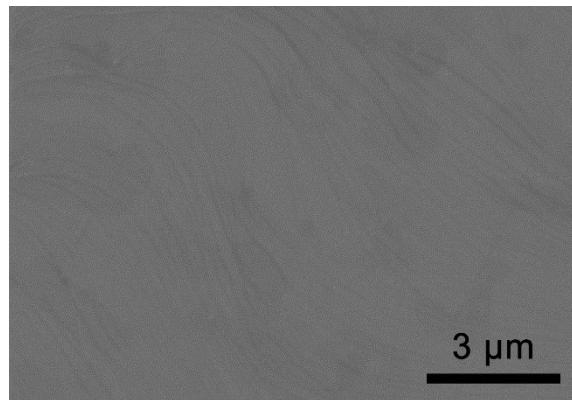


Figure S2. CVD growth after 5 hours still obtains a uniform BLG film, which indicates an equilibrium state for BLG growth using ethanol as the precursor.

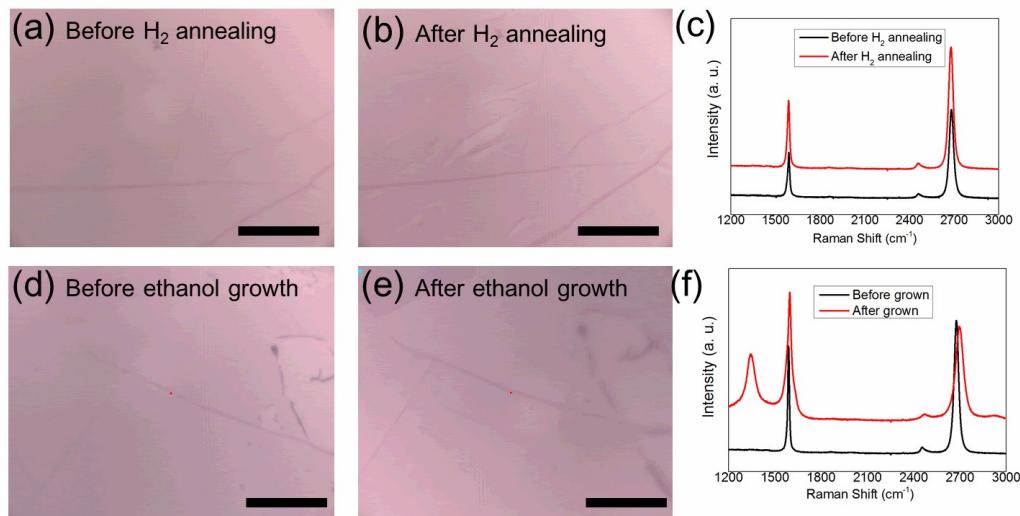


Figure S3. CVD growth at an equivalent condition using ethanol as the precursor but graphene transferred onto a Si/SiO₂ as the substrate instead of copper. No new graphene layer was obtained even consider the etching effect from H₂.

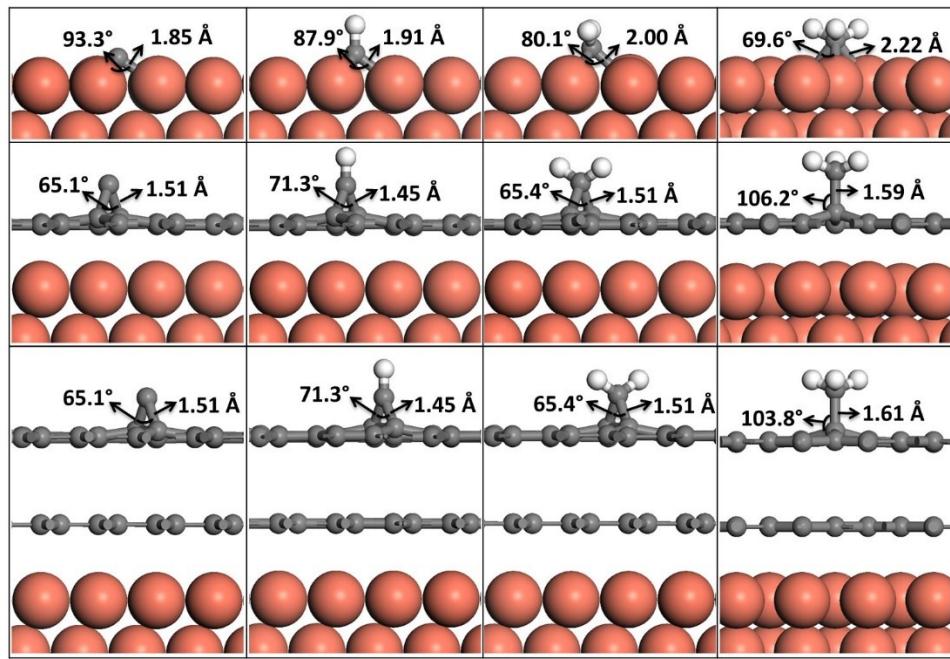


Figure S4. Geometries of different CH_i ($i=0, 1, 2, 3$) radicals on a Cu(111)/graphene substrate.

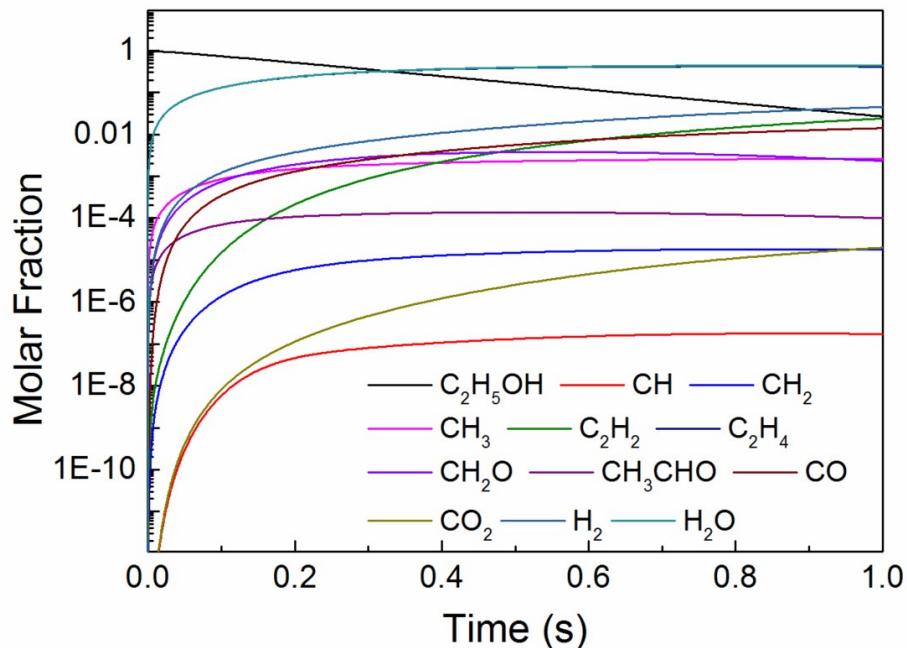


Figure S5. Decomposition products of $\text{C}_2\text{H}_5\text{OH}$ at high temperature calculated by CHEMKIN-II.

Table S1. Total energy data calculated for the radical/graphene/Cu(111) system.

Radical	C	CH	CH2	CH3
Total energy (eV)	-1.23966895	-6.05787351	-12.11872387	-18.21134477
Substrate	Cu(111)		Cu(111)/MLG	Cu(111)/BLG
Total energy (eV)	-140.81099300		-308.66524756	-475.86328245
	C	CH	CH2	CH3
Cu(111)	-147.39233313	-152.24136847	-156.34241493	-160.82963338
Cu(111)/MLG	-311.70228781	-316.63018221	-322.55594031	-328.01285288
Cu(111)/BLG	-479.17043701	-484.47056304	-490.43820732	-494.42125405

Table S2. Binding energy data for the CH_i radical on a graphene/Cu(111) substrate.

	C	CH	CH_2	CH_3
Cu	5.3417	5.3725	3.4127	1.8073
1LG/Cu	1.7303	1.8400	1.7049	1.0692
2LG/Cu	2.0637	2.5456	2.4524	0.3429