Supporting Information

Cooperative Island Growth of Large Area Single-Crystal Graphene on Cu Using Chemical Vapor Deposition

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Figure S1: Optical images of commercial Cu foils after 15 min oxidation in air at the given temperatures.
Figure S2: 3D optical micrographs of Cu foils after 3h annealing/reduction in H₂. f<sub>RT</sub> is the as received foil after annealing, f<sub>200</sub>, f<sub>300</sub>, and f<sub>400</sub> are the Cu foils which were oxidized in air respectively at 200, 300 and 400°C for 15 minutes before the annealing/reduction process. The rolling produced features are almost gone from f<sub>300</sub> and new microstructure emerged on f<sub>400</sub>. The same scale bar applies for all micrographs.

Figure S3: RMS roughness extracted from AFM images as a function of the oxidation and annealing treatment. f<sub>AR</sub> designates as received foils with no further treatment, f<sub>RT</sub> designates foils that had only a native room temperature oxide layer but, received the same H₂ annealing treatment as all the oxidized foils.
**Figure S4:** (a), XRD θ-2θ scans taken from three different Cu foils $f_{300}$, $f_{400}$ and $f_{500}$. Foil $f_{300}$ shows mixture of (100), (110), (111) and (311) orientations. Scans similar to that of $f_{300}$ were observed from $f_{RT}$ and $f_{200}$ as well. Foils $f_{400}$ and $f_{500}$ show (100) orientations only. (b), (111) pole figures taken from foil $f_{400}$. Foil $f_{500}$ also showed similar pole figure as of $f_{400}$.

**Figure S5:** Terminated size of graphene grain after 2 hours of continuous growth on $f_{300}$ (Cu foil oxidized at 300°C followed by H$_2$ reduction) at 10sccm H$_2$, 0.5sccm CH$_4$, 150mTorr, 1040°C.
Figure S6: Large single crystal graphene grains (SCGG) grown on $f_{300}$ and $f_{400}$ as indicated.
Figure S7: Graphene grains showing multilayer graphene patches nucleated at the nucleation centers or around impurity particles.

Figure S8: Cu foils $f_{300}$ and $f_{400}$ fully covered by single layer graphene after 2 hours of continuous growth at 10 sccm $H_2$, 1.5 sccm $CH_4$, 150 mTorr total pressure and 1040°C.
Figure S9: Optical micrographs from Cu foil at room temperature: (a) after 3 hours of annealing/reduction in H\textsubscript{2} before graphene nucleation, and (b) after graphene nucleation. Graphene preferentially nucleates on the rolling features of the Cu foil. Note that the graphene nucleation density is higher on the high rolling features, white arrows, and lower on flat regions, black arrow. (c) SEM image taken from the same Cu foil as in (b). White lines highlight the rolling features. Graphene islands can be imaged using optical microscope because of Cu mound formation below the graphene islands, as the Cu continuously evaporates from the region outside of the graphene islands. (Wofford, 2010 ref #66) Scale bar is 20 µm for all the images.
Figure S10: SEM images taken from typical graphene nucleation sites on Cu foils. Graphene nucleation preferentially takes place on defects or impurity particles on the Cu foil surface. Same scale bar applies for all the images.
Figure S11: SEM Images from Cu foils f_{300} and f_{400} after 0.5 hours of graphene growth at 40sccm Ar, 10sccm H$_2$, 1.5sccm CH$_4$, 500mTorr and 1040°C. Note that graphene grains show hexagonal morphology on f_{300} and rectangular morphology on f_{400}. Higher nucleation density on f_{400} is attributed to the higher density of step edges because of the higher roughness of this surface.
Figure S12: SEM images of graphene grown in the low nucleation regime. The plot of coverage as a function of time used for determining the characteristic time constant $\tau$ of the sigmoidal function from the equation $\theta(t) = 1/[1 + \exp(-t/\tau)]$. The value of the time constant obtained from the fit is $\tau=9.3$ min.