Supporting Information

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

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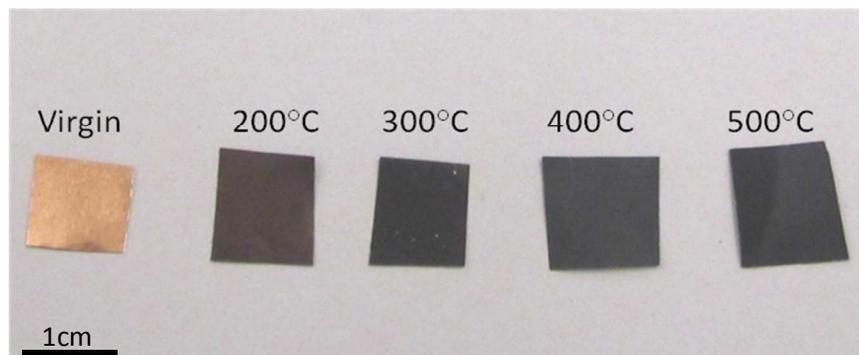
Figure S1: Optical images of commercial Cu foils after 15 min oxidation in air at the given temperatures.
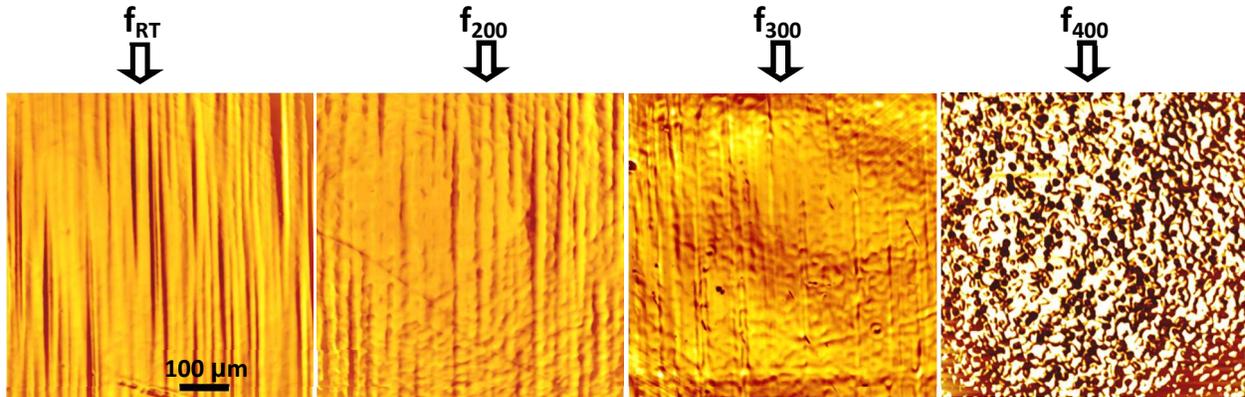
Figure S2: 3D optical micrographs of Cu foils after 3h annealing/reduction in H_2. f_{RT} is the as received foil after annealing, f_{200}, f_{300}, and f_{400} are the Cu foils which were oxidized in air respectively at 200, 300 and 400˚C for 15 minutes before the annealing/reduction process. The rolling produced features are almost gone from f_{300} and new microstructure emerged on f_{400}. The same scale bar applies for all micrographs.
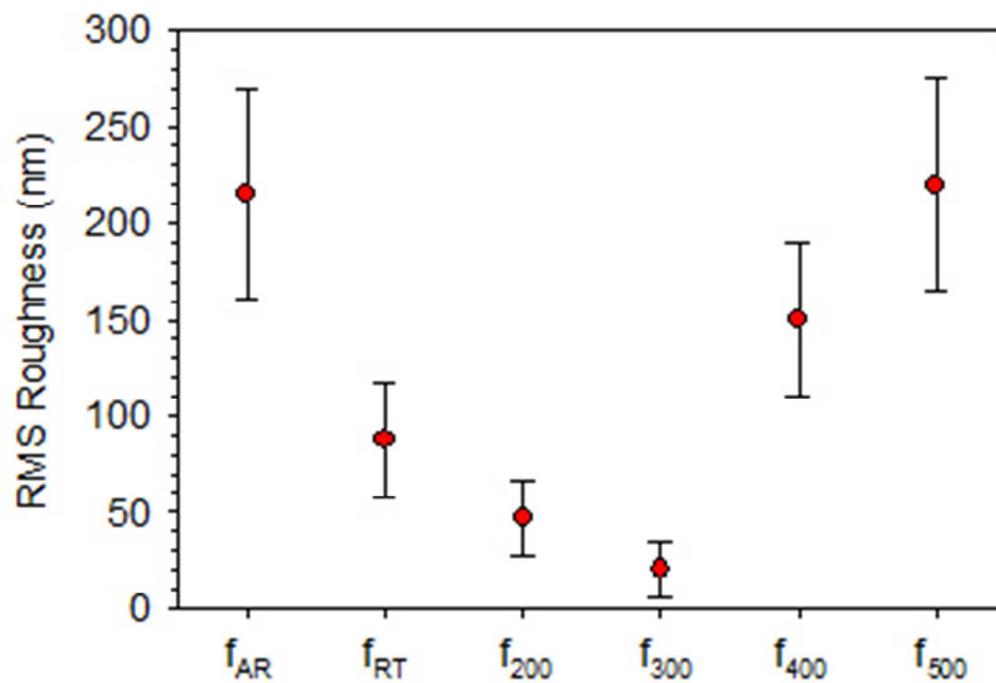

Figure S3: RMS roughness extracted from AFM images as a function of the oxidation and annealing treatment. f_{AR} designates as received foils with no further treatment, f_{RT} designates foils that had only a native room temperature oxide layer but, received the same H_2 annealing treatment as all the oxidized foils.
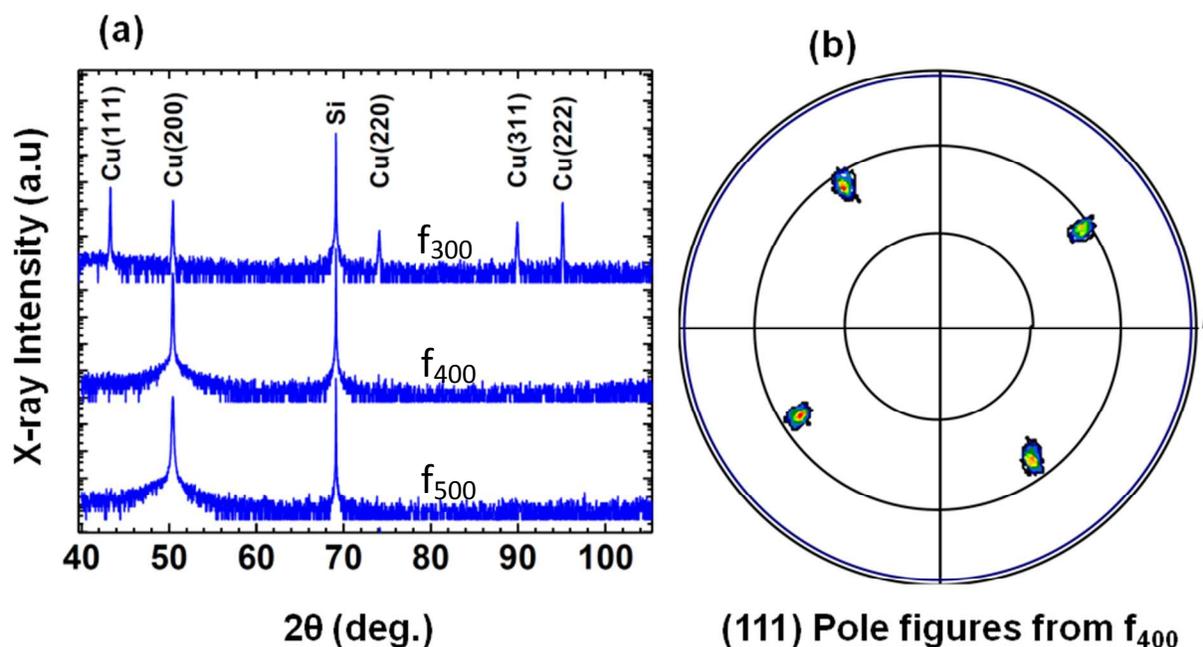
Figure S4: (a), XRD θ-2θ scans taken from three different Cu foils f_{300}, f_{400} and f_{500}. Foil f_{300} shows mixture of (100), (110), (111) and (311) orientations. Scans similar to that of f_{300} were observed from f_{RT} and f_{200} as well. Foils f_{400} and f_{500} show (100) orientations only. (b), (111) pole figures taken from foil f_{400}. Foil f_{500} also showed similar pole figure as of f_{400}.
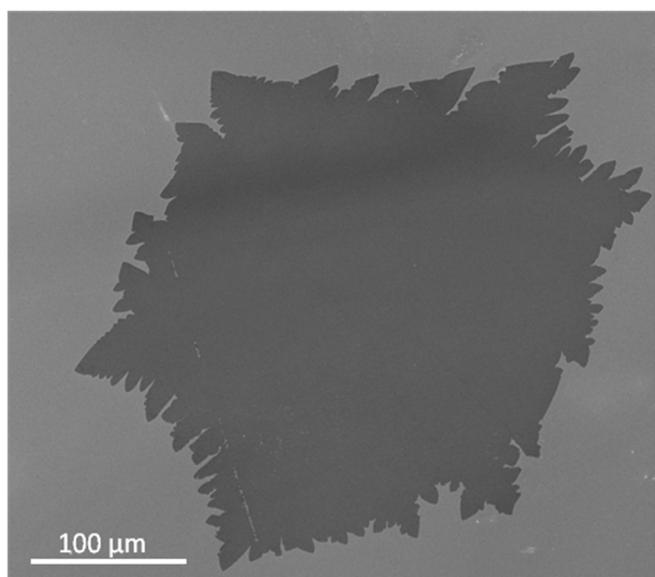

Figure S5: Terminated size of graphene grain after 2 hours of continuous growth on f_{300} (Cu foil oxidized at 300°C followed by H_2 reduction) at 10sccm H_2, 0.5sccm CH_4, 150mTorr, 1040°C.
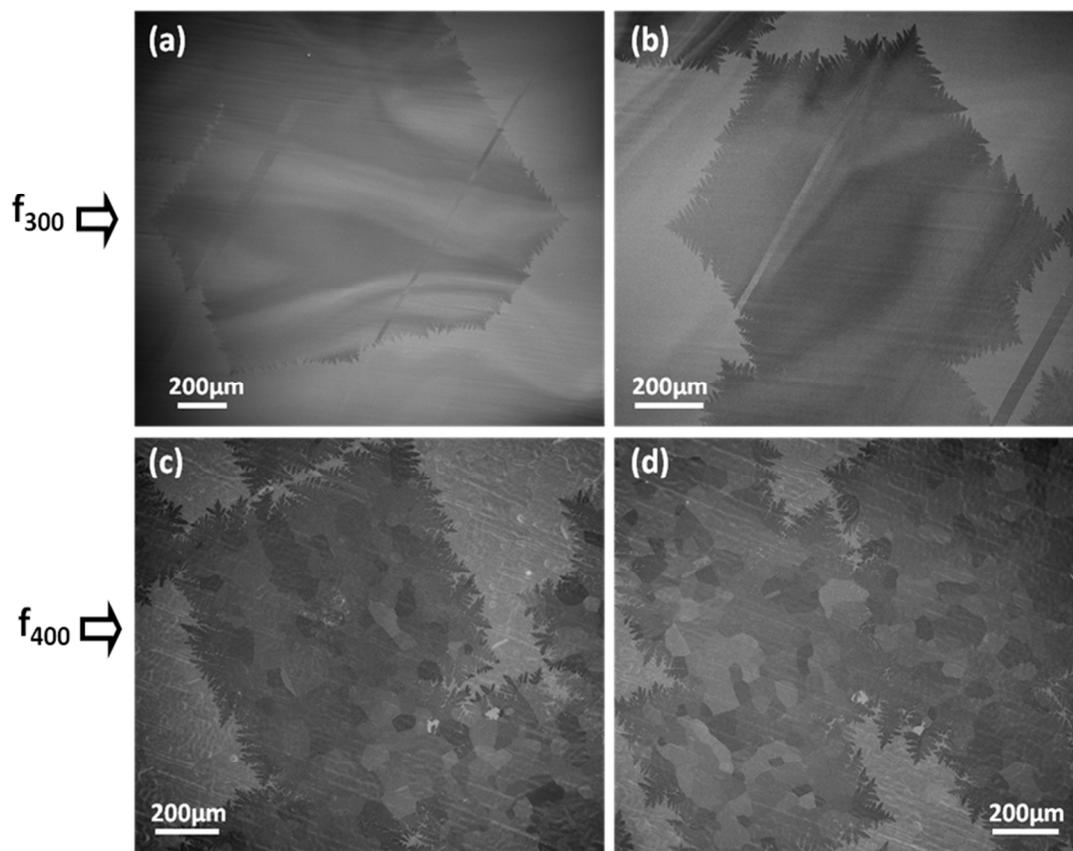
Figure S6: Large single crystal graphene grains (SCGG) gown on f_{300} and f_{400} as indicated.
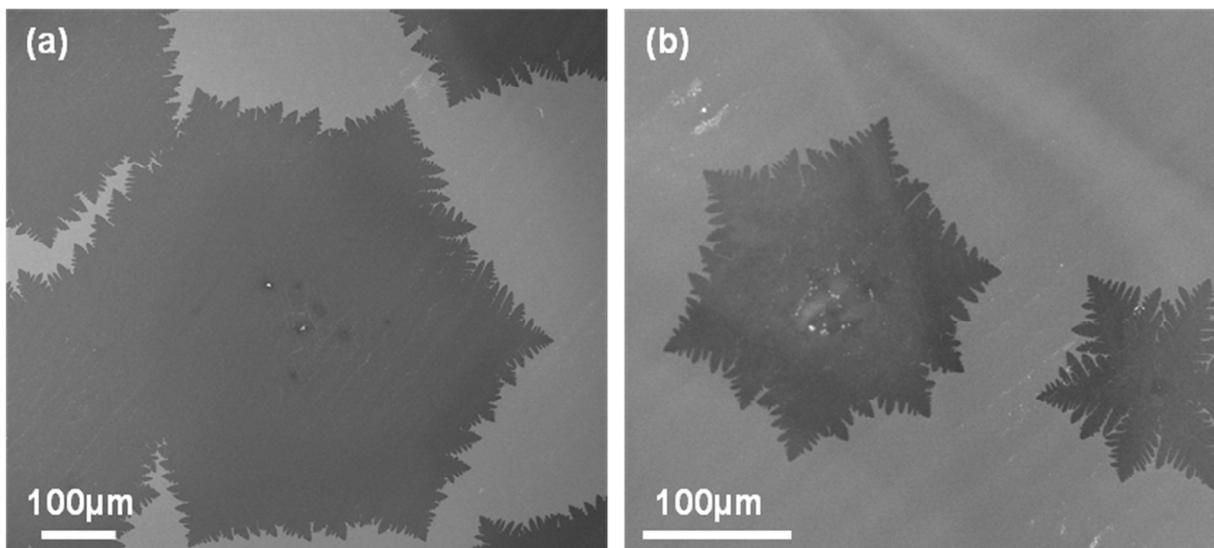
Figure S7: Graphene grains showing multilayer graphene patches nucleated at the nucleation centers or around impurity particles.
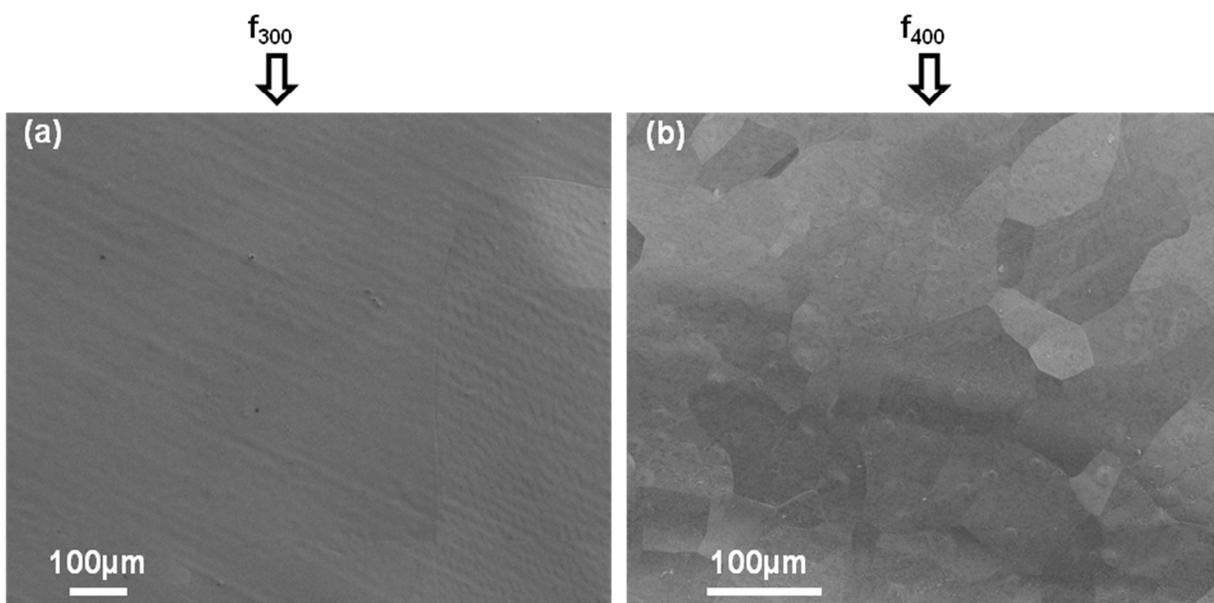

Figure S8: Cu foils f_{300} and f_{400} fully covered by single layer graphene after 2 hours of continuous growth at 10 sccm H_2, 1.5 sccm CH_4, 150 mTorr total pressure and 1040°C.
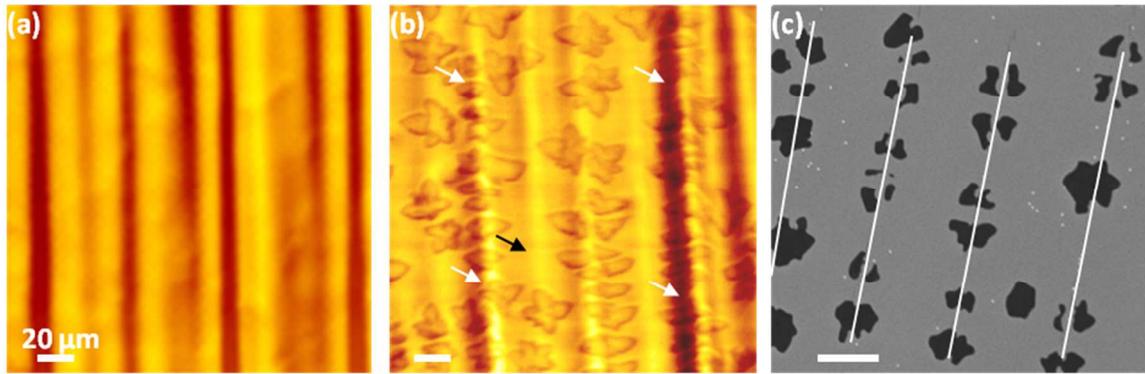
Figure S9: Optical micrographs from Cu foil f_{RT}: (a) after 3 hours of annealing/reduction in H_2 before graphene nucleation, and (b) after graphene nucleation. Graphene preferentially nucleates on the rolling features of the Cu foil. Note that the graphene nucleation density is higher on the high 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.
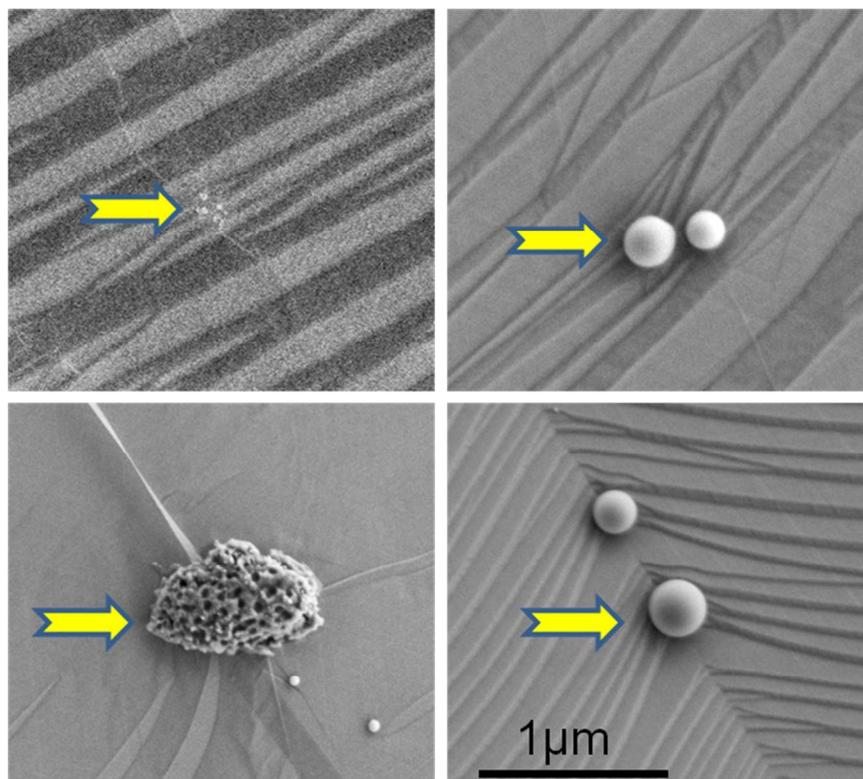
Figure S10: SEM images taken from typical graphene nucleation sites on Cu foils. Graphene nucleation preferentially takes place on defects or impurity particles on the Cu foil surface. Same scale bar applies for all the images.
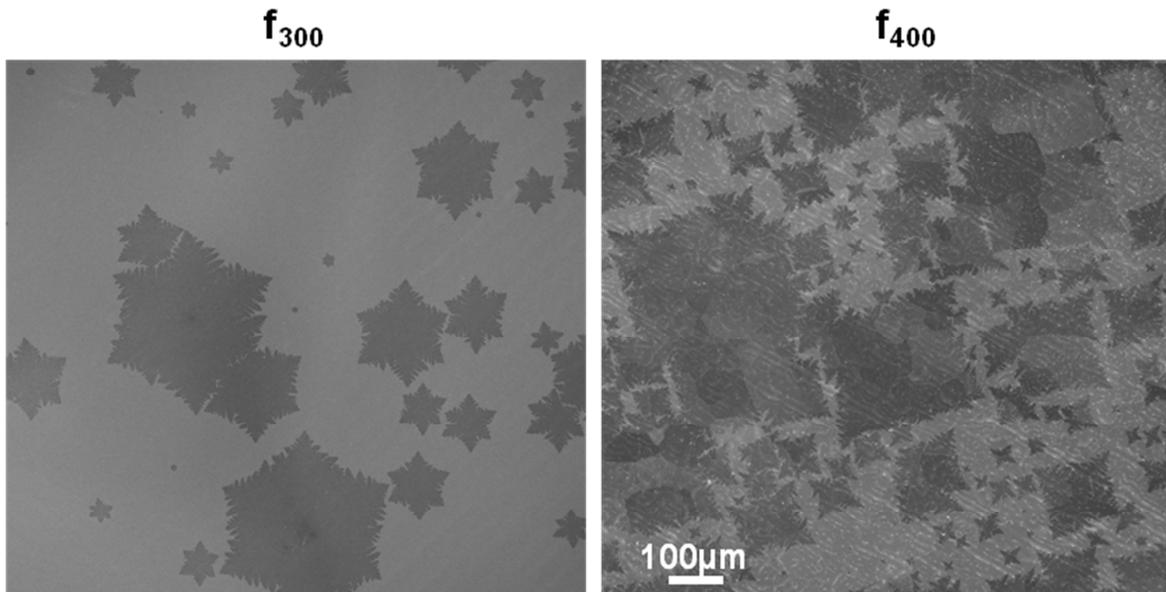
Figure S11: SEM Images from Cu foils f_{300} and f_{400} after 0.5 hours of graphene growth at 40sccm Ar, 10sccm H_2, 1.5sccm CH_4, 500mTorr and 1040°C. Note that graphene grains show hexagonal morphology on f_{300} and rectangular morphology on f_{400}. Higher nucleation density on f_{400} is attributed to the higher density of step edges because of the higher roughness of this surface.
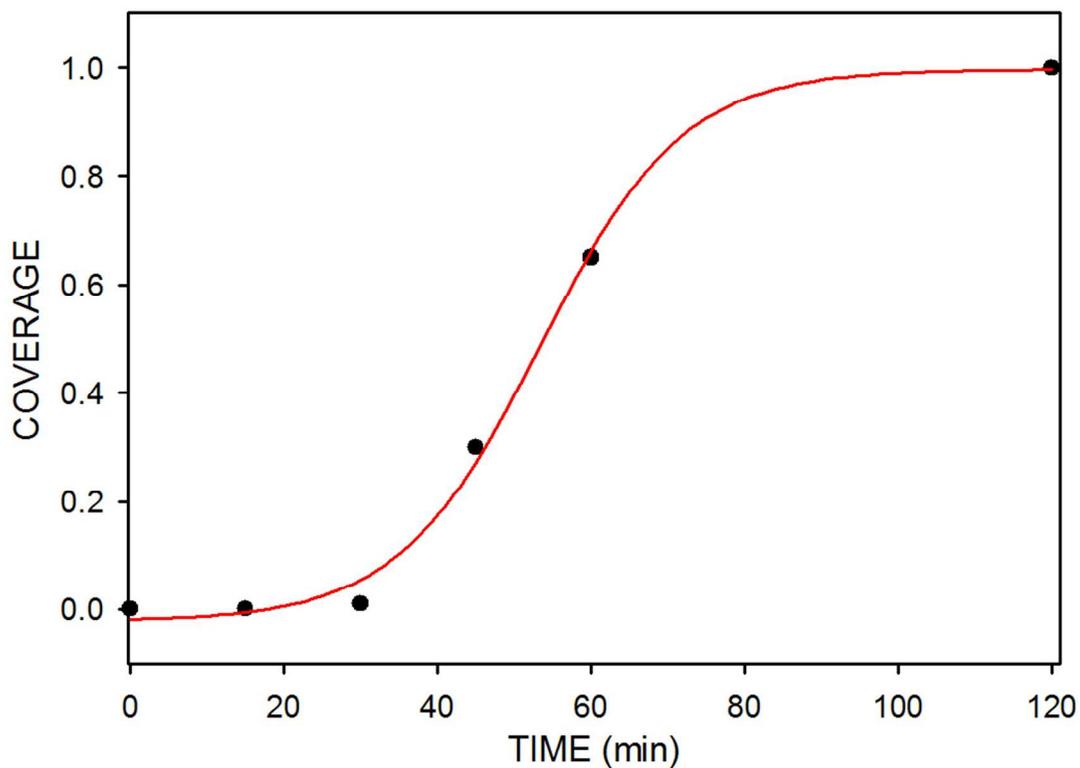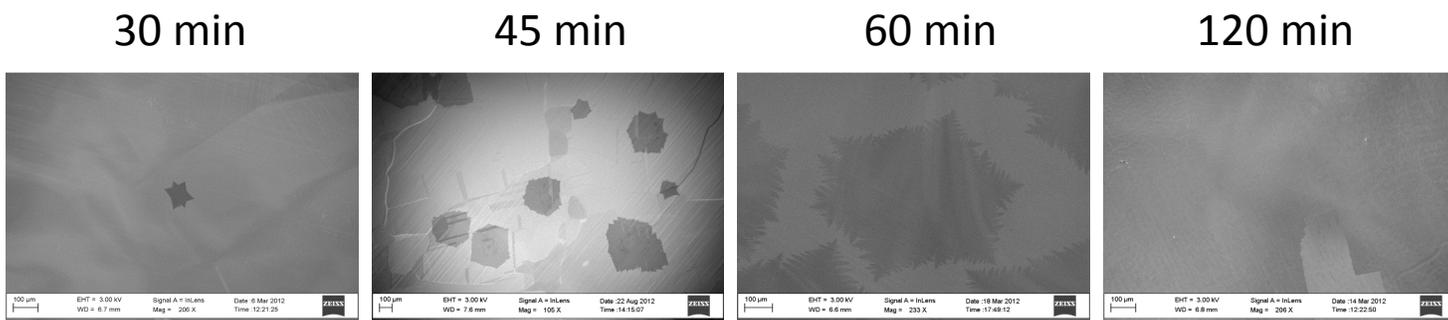
Figure S12: SEM images of graphene grown in the low nucleation regime. The plot of coverage as a function of time used for determining the characteristic time constant τ of the sigmoidal function from the equation $\theta(t) = 1/[1 + \exp(-t/\tau)]$. The value of the time constant obtained from the fit is τ=9.3 min.