Supplementary Information

Direct growth of self-crystallized graphene and graphite nanoballs with Ni vapor-assisted growth: From controllable growth to material characterization

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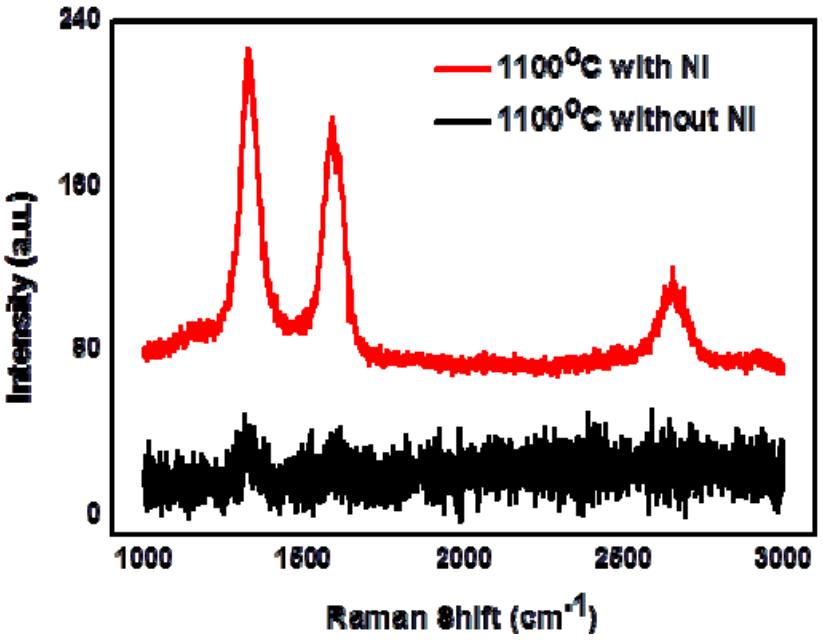


Figure S1 Raman spectrum for the graphene growth with and without Ni ingot-assisted approach.

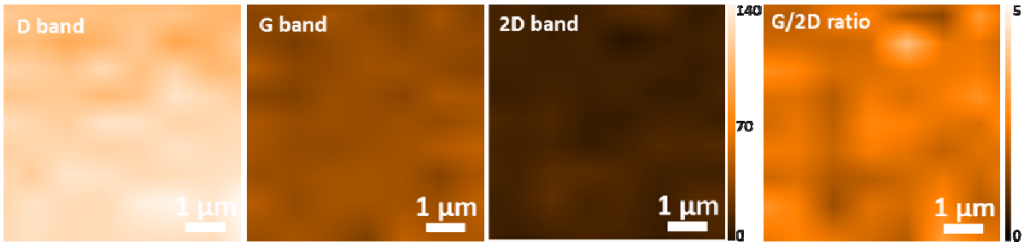


Figure S2 Raman mapping images of *D*, *G*, *2D* bands and ratio of I_G/I_{2D} for the graphene with 10 μm^2 size.

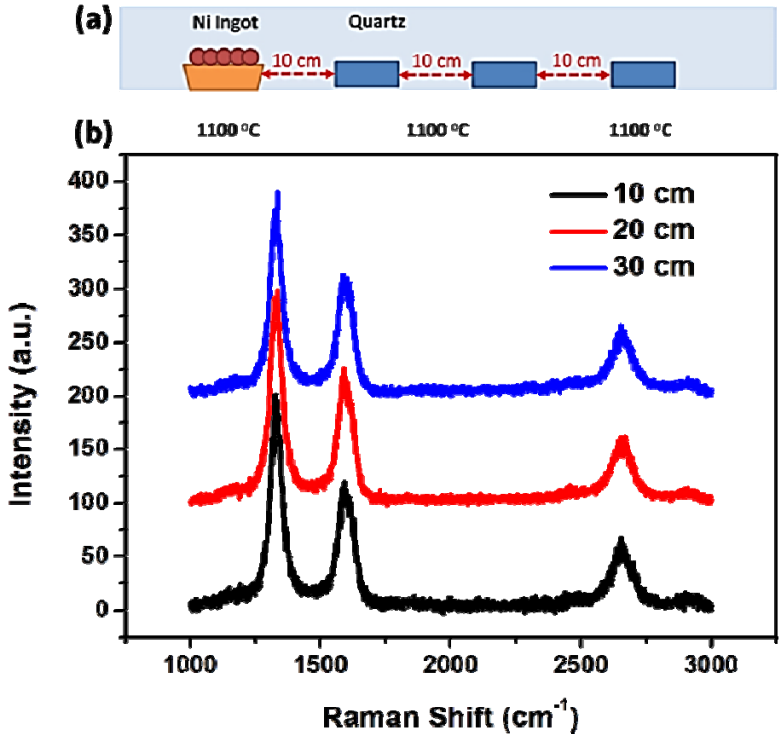


Figure S3 (a) Schematic of graphene growth at different distances between Ni ingots and quartz substrate. (b) The corresponding Raman spectra at different positions.

The direct growth of the graphene from the self-crystallization process without the transfer process can be also applied on an Al_2O_3 substrate

We demonstrated growth of graphene on alumina oxide as a substrate. The corresponding Raman spectra were shown in Fig. S4 where the growth temperature is 1000 ºC for 5 minutes at 60 torr with Ar 100/H_2 20/CH_4 50 sccm. Clearly, distinct graphene signals, including *D*, *G*, and *2D* peaks can be observed,

indicating the growth of the graphene on arbitrary insulator substrate. We have added this figure into the supplementary information.

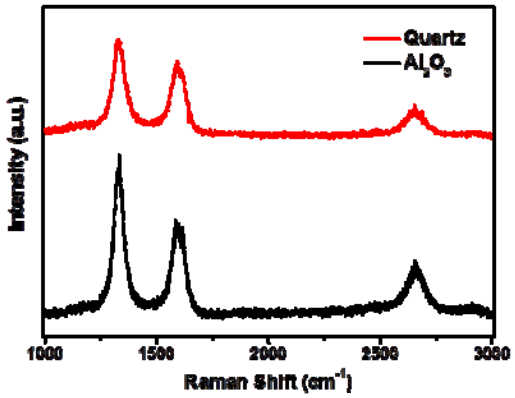


Fig S4 Raman specta for the growth of the graphene on alumina oxide and quartz as substrates.

HRTEM image of graphen deposited at 1100 °C with a pressures of 60 torr for 5 minutes

Graphene was deposited at 1100 °C with a pressure of 60 torr for 5 minutes on quartz substrate. The thickness of multi-layer graphene is measured to be ~38.4 nm, corresponding to 110 layers with a distance between layer to layer of ~ 0.35 nm.

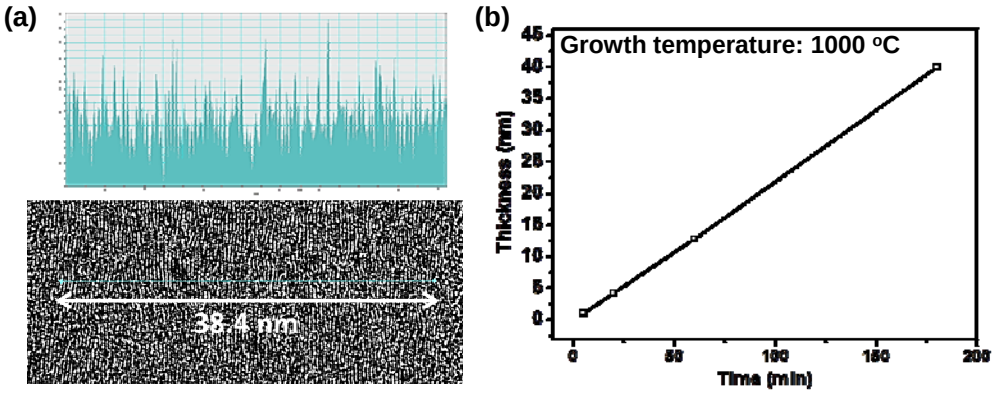


Figure S5 (a) A HRTEM image of graphene deposited at 1100 °C and 60 torr for 5 minutes. (b) A plot of thicknesses as a function of the growth time at the fixed growth temperature of 1000 °C

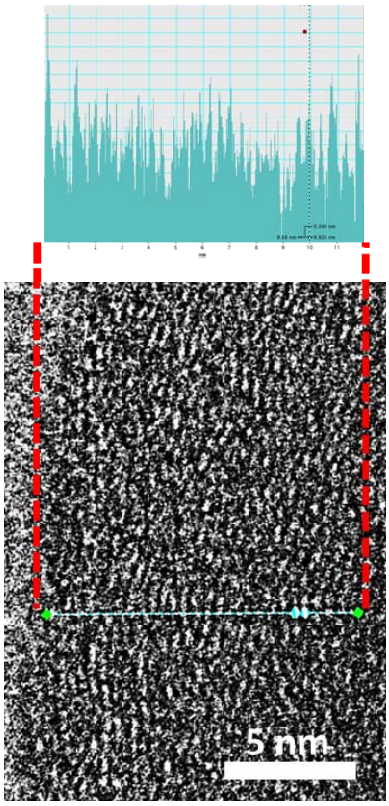


Figure S6 A high resolution TEM image of GNBs.

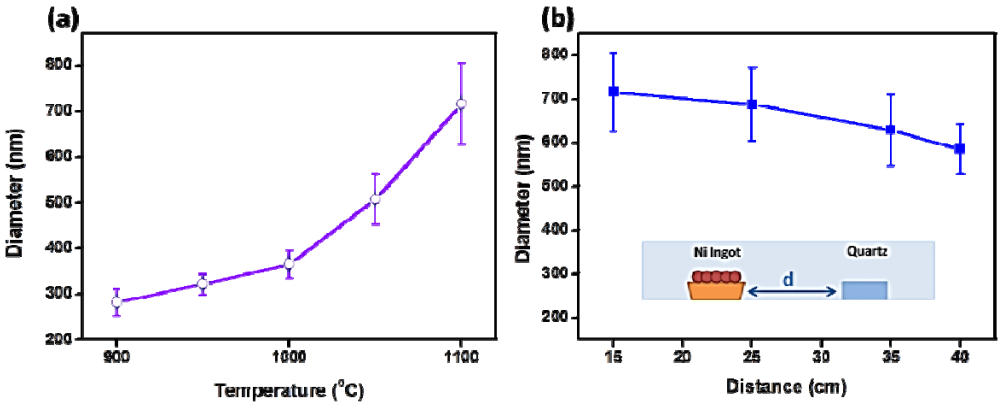


Figure S7 Controlled diameters of GNBs at (a) different growth temperatures and (b) distances between Ni ingots and quartz substrate.

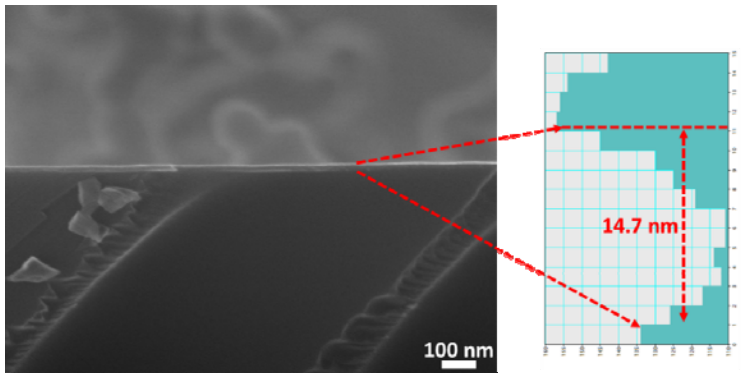


Figure S8 A SEM image of graphene layer before growth of the GNBs

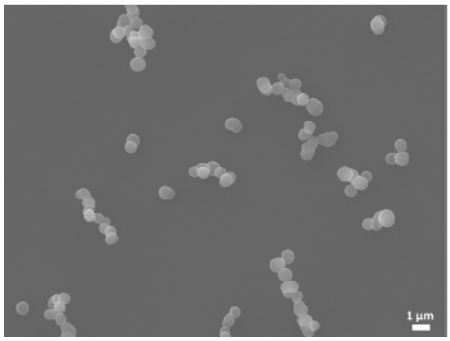


Figure S9. A SEM image for GNBs at a low cooling rate (~ 0.3 °C /s)

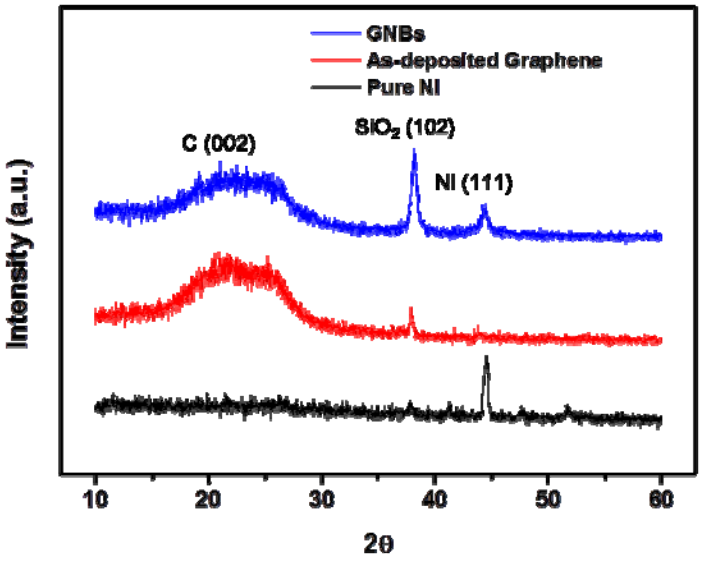
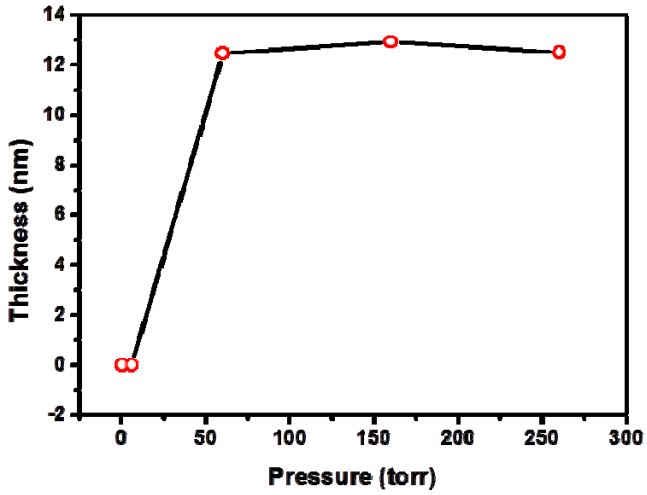


Figure S10. XRD results with grazing angle for GNBs, graphene, and Ni ingots.



Figrue S11. Thickness of grapheen growth at growth temperature and time of 1000 °C and 5 min with different growth pressure.

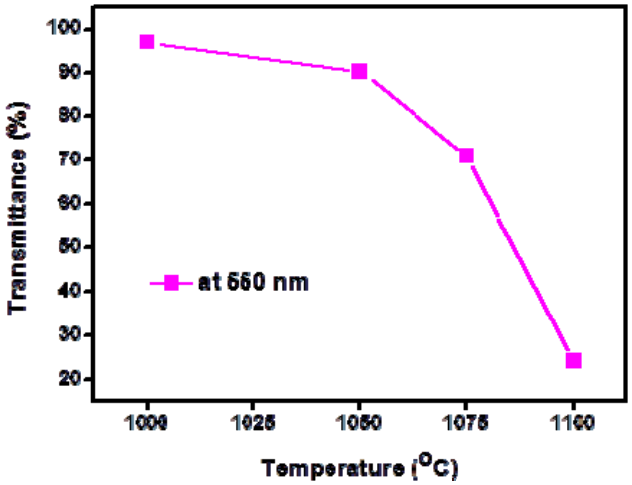


Figure S12. Transmittance spectra at 550 nm of graphene grown at different temperatures.

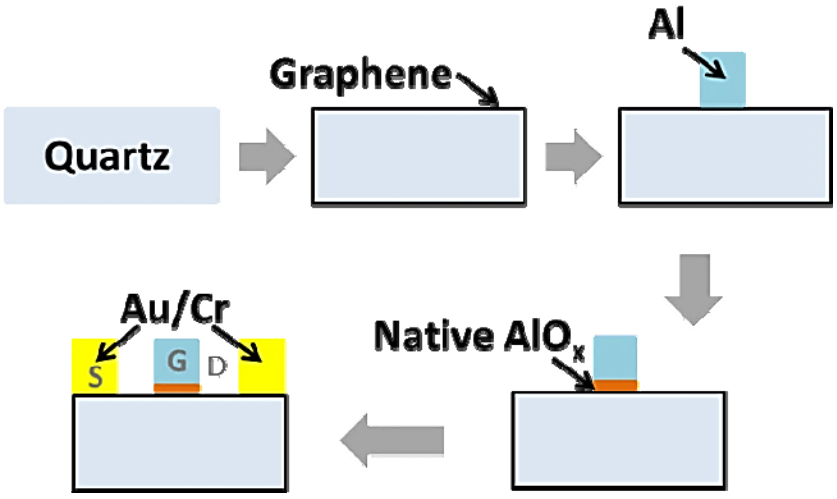


Figure S13. Fabrication processes of self-oxidation process for formation of top-gate device on graphene layer. After depositing graphene on the quartz, a top gate metal Al 350 nm was deposited by e-gun deposition. Subsequently the device was loaded into a sealed chamber with extremely pure O_2 ambiance upto 1kg/cm^2 for 48 hour, resulting in formation of AlO_x layer. Finally, Au 50 nm/ Cr 3 nm was deposited by e-gun deposition as source and drain.