

## Supplementary Information

### Large Scale Direct Synthesis of Graphene on Sapphire and Transfer-free Device Fabrication

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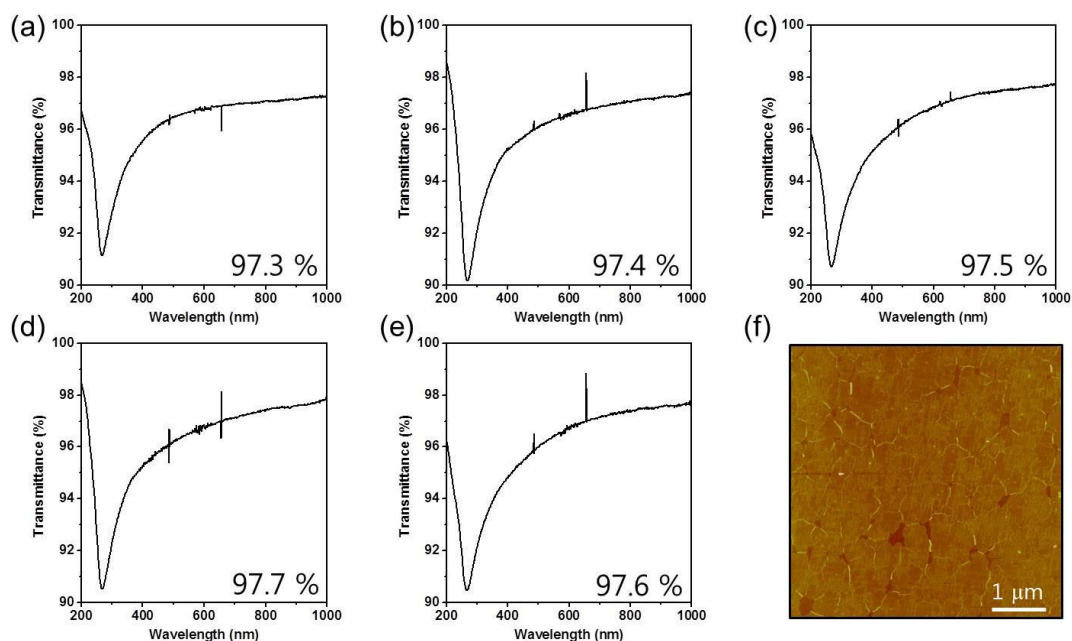
E-mail: [choihc@postech.edu](mailto:choihc@postech.edu)

### Experimental details

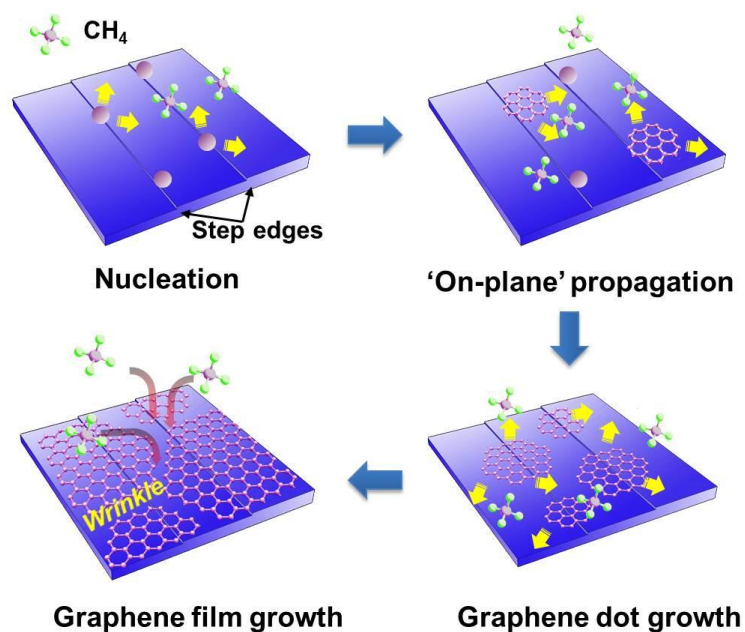
*Transfer of graphene:* To obtain electron diffraction pattern of graphene, as-grown graphene was transferred from  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (0001) substrate to holey silicon nitride TEM grids (PELCO Holey Silicon Nitride Support Films) with 2.5  $\mu$ m diameter holes. The transfer method is similar to the one used in ref. 1. Initially, 15 nm of gold was thermally evaporated on as-grown graphene sample, on top of which 4 % of poly(methyl methacrylate) (PMMA) dissolved in chloroform was drop-casted. After the PMMA was hardened, PMMA/gold/graphene layer was mechanically isolated from the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (0001) substrate. PMMA was then removed by soaking in methylene chloride and the gold/graphene layer was scooped by TEM grid. In the last step, suspended graphene on TEM grid was obtained by etching the gold using commercial gold etchant (Transcene, Type TFA).

*Characterization:* Atomic force microscope (AFM, Nanoscope IIIa, Digital Instrument Inc.) was used to investigate the morphology and height information. Raman spectra and images were acquired using a Raman spectroscope (Alpha 300R, WITEC) equipped with 532 nm of a diode laser. The spatial resolution of the spectrometer is  $\sim 250$  nm. The excitation laser power was optimized at below 5 mW to avoid potential thermal damages by laser sources. Electron diffraction pattern was obtained using a JEOL JEM-2100 operated at 200 kV. Note that there was no apparent damage to graphene samples while electron diffraction experiment. To measure the optical transmittance, double side-polished  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (0001) was used to grow graphene. For accurate measurements, one side of sapphire was etched by oxygen-plasma. Note that the complete removal of graphene was confirmed by Raman spectroscopy, and that there was no damage to the other side of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (0001) during the etching process.(Fig. S1f) Transmittance was measured using a UV-Vis spectrometer (Agilent 8453 spectrophotometer), and a bare  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (0001) was used as a reference blank. The measurements were made for five samples of graphene films which are grown for 120 min.

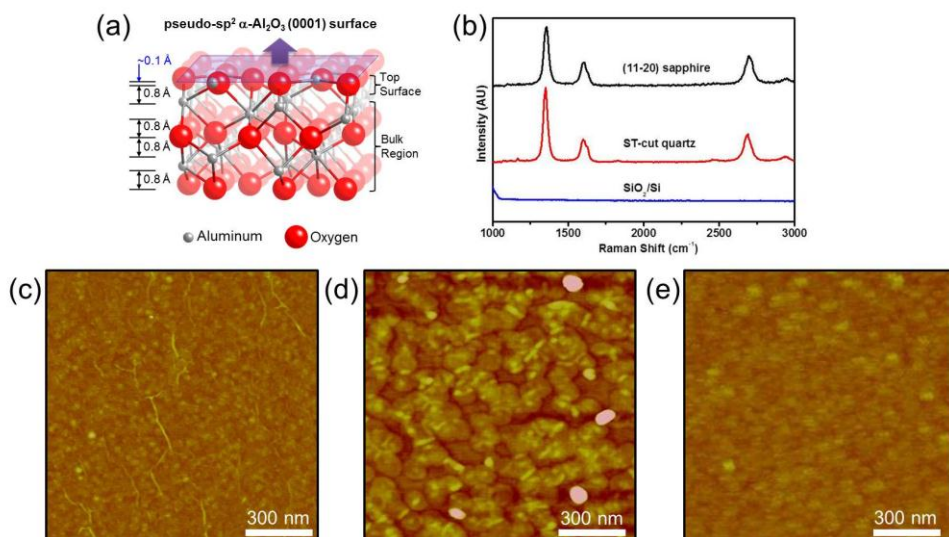
*Fabrication of field effect transistor device:* The top-gated field effect transistor was fabricated by standard photolithography technique. Negative type photoresist was spun on as-grown graphene and it was patterned to behave as a protecting layer against graphene etching. The sample was then exposed to oxygen plasma for 40s to remove the graphene sitting on unwanted areas. After fabricating source/drain electrode patterns with Cr (5 nm) / Au (35 nm) layer, the sample was thermally annealed at 300 °C at H<sub>2</sub> / Ar atmosphere for 3 hours. 100 nm thick film of SiO<sub>2</sub> was deposited on the whole wafer by electron beam evaporation to form a gate oxide layer, on which gate electrodes were patterned with another Cr (5 nm) / Au (35 nm) layer.



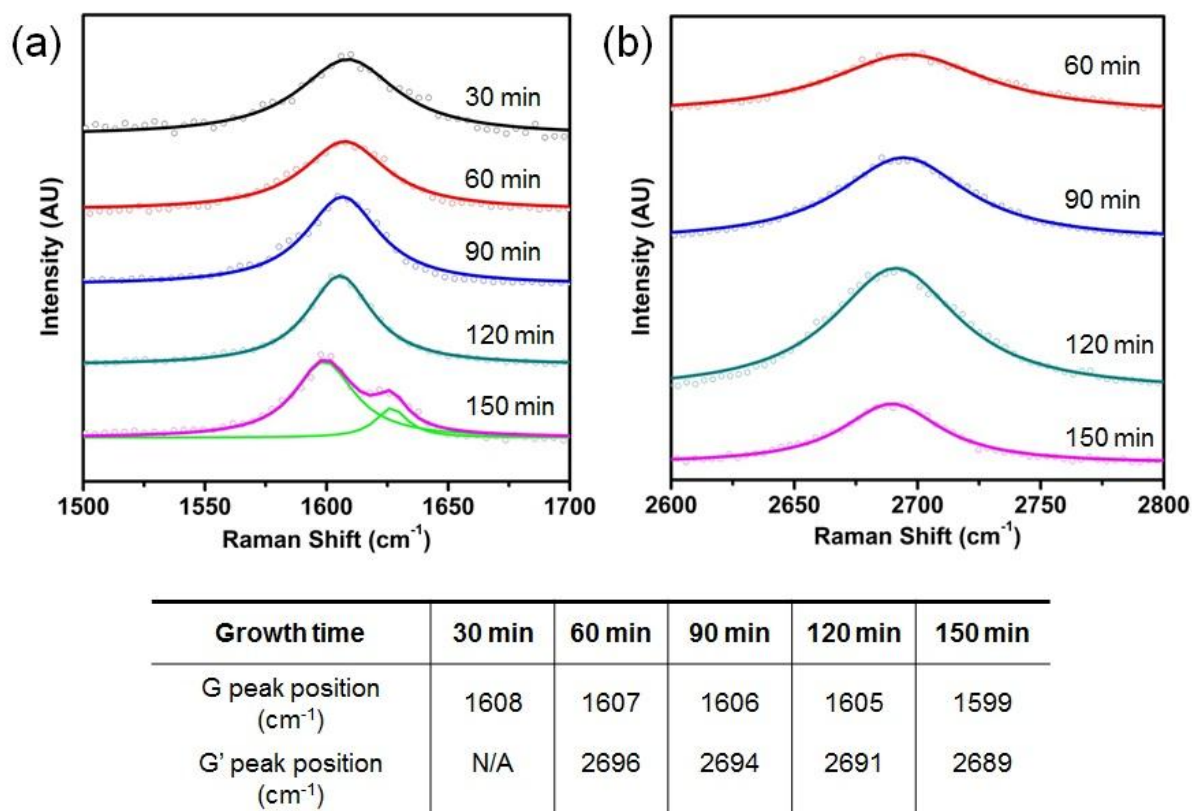
**Figure S1.** (a) – (e) Transmittance of graphene film grown on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (0001). The saturated transmittance values are shown on right bottom of the each spectrum. (f) A representative AFM image of graphene film on a front side after the opposite side of graphene was removed by oxygen-plasma.



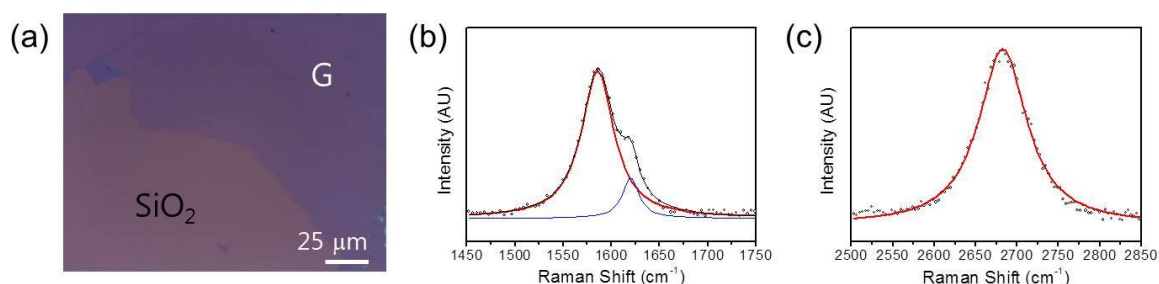
**Figure S2.** Schematic illustration of metal catalyst-free growth of graphene on a  $\alpha$ - $\text{Al}_2\text{O}_3$  dielectric substrate via chemical vapor deposition (CVD) process. The nucleation preferentially occurs at the step edges, followed by ‘on-plane’ propagation to yield graphene dots. Further growth increases the size of graphene dots, and finally forms graphene film with wrinkles at each boundary where graphene dots are merged.



**Figure S3.** (a) Schematic of  $\alpha$ - $\text{Al}_2\text{O}_3$  substrate showing relaxed (0001) top surface (b) Raman spectra measured from attempts of graphene growth of 120 min on various dielectric substrates. The top two spectra are normalized with G peak intensities. AFM images of graphene grown on (c)  $\alpha$ - $\text{Al}_2\text{O}_3$  (11-20) substrate and (d) ST-cut quartz substrate. (e)  $\text{SiO}_2/\text{Si}$  substrate on which graphene growth was attempted. All the images were taken after 120 min of reaction under the same condition used in **Fig. 1**.



**Figure S4.** Magnified view of Raman (a) G peak, and (b) G' peak regions from **Fig. 3f**. Each peak position is summarized in the table. Note that all the spectra are fitted with Lorentzian, and the fitted G' peak for the 30 min reaction sample is not available due to high level noise.



**Figure S5.** Raman spectra of graphene transferred to SiO<sub>2</sub> (300 nm)/Si (100) substrate. (a) Optical microscopy image of transferred graphene. Letter 'G' represents graphene region. Raman spectra of (b) G peak (red curve) and (c) G' peak. Each spectrum is fitted with Lorentzian. Note that weak D' peak (blue curve in (b)) is observed mainly due to the damage caused during the transfer process.

## References

- 1 M. Huang, H. Yan, C. Chen, D. Song, T. F. Heinz, J. Hone, *Proc. Nat. Acad. Sci. USA* 2009, **106**, 7304.