Supporting Information for

Pulsed-Grown Graphene for Flexible Transparent Conductors

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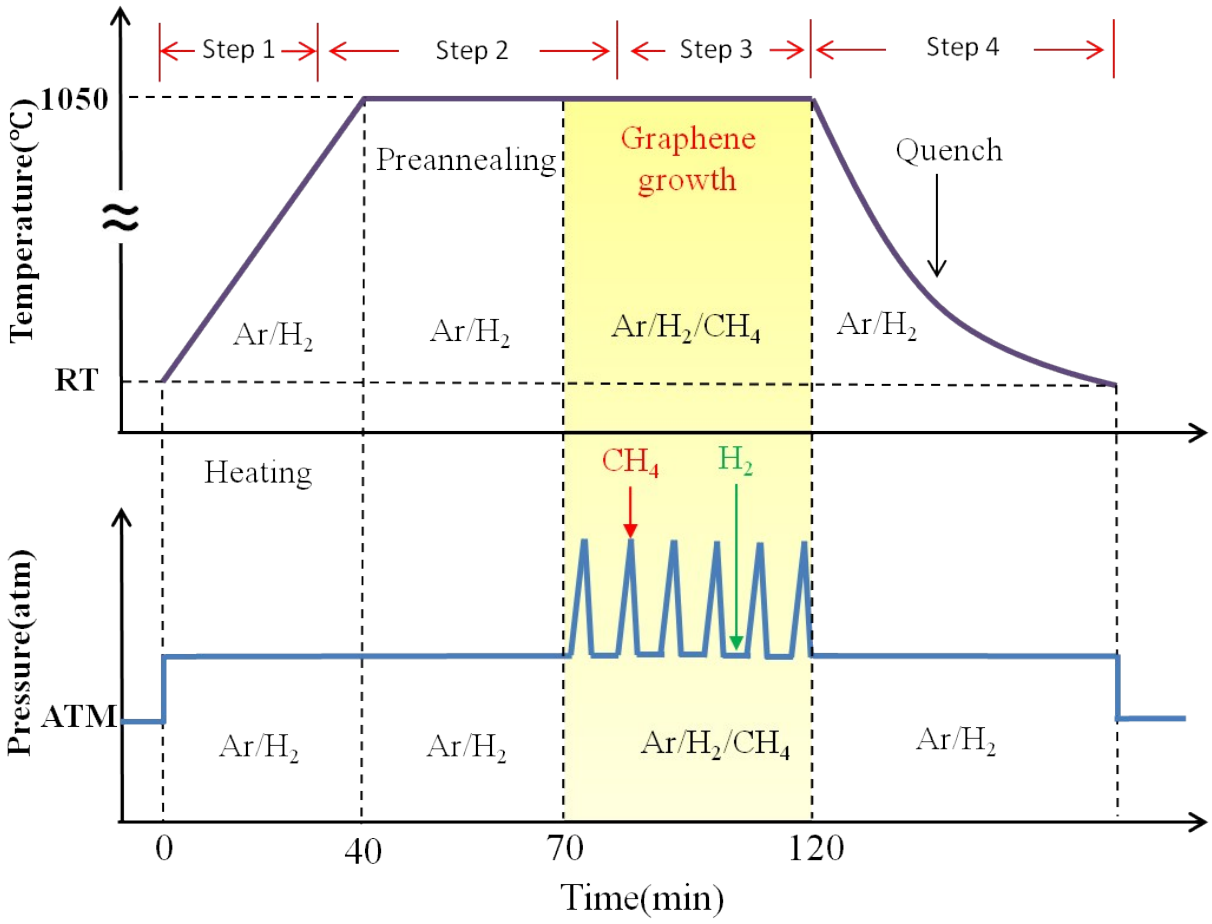


Fig. S1 Experimental details of pulse growth graphene. Temperature and pressure vs. time for modified AP-CVD growth of graphene on polycrystalline Cu foil. In step 1, the furnace was ramped up to 1050°C during 40 min. In step 2, pre-annealing of Cu foil was carried out at 1050°C for 30 min to enhance the grain size and smoothen the Cu surface. In step 3, methane with a flow rate of < 1 sccm was fed into the reaction chamber for 50 min (15-25 cycles) during which graphene growth occurs. In each cycle, a CH_4 pulse was active for 30s and then released for 1 min. Finally, in step 4, the Cu foil was moved to the cooling zone where a cooling system was equipped. Steady flow of 10-20 sccm hydrogen and 300 sccm of Ar was maintained throughout all the steps.

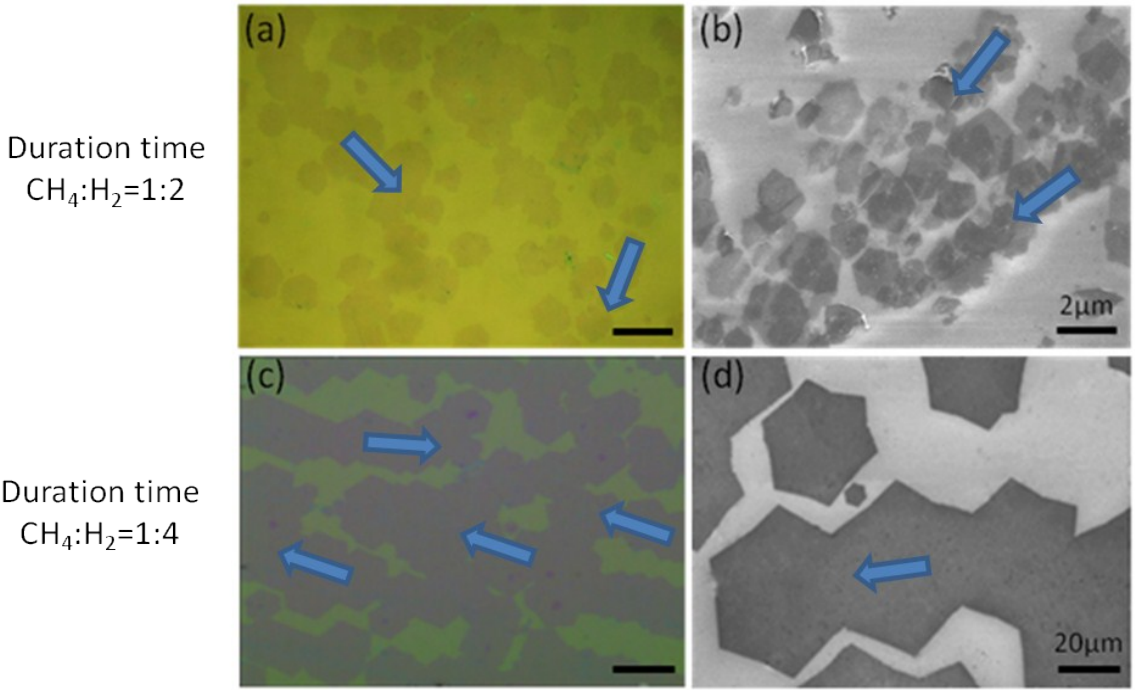


Fig. S2 Effect of dwell time of CH_4 pulses on graphene growth. (a-b) OM and SEM images of graphene grown for short duration time ratio of CH_4:H_2=1:2 and (c-d) that of long duration time ratio of CH_4:H_2=1:4. The average graphene domain size is small for longer dwell time of CH_4 pulses (a, b) and is large for shorter dwell time of CH_4 pulses (c, d). Scales bar is 2 μm for (a, b) and 20 μm for (c, d). The arrows show the preferred orientation of the graphene domains.

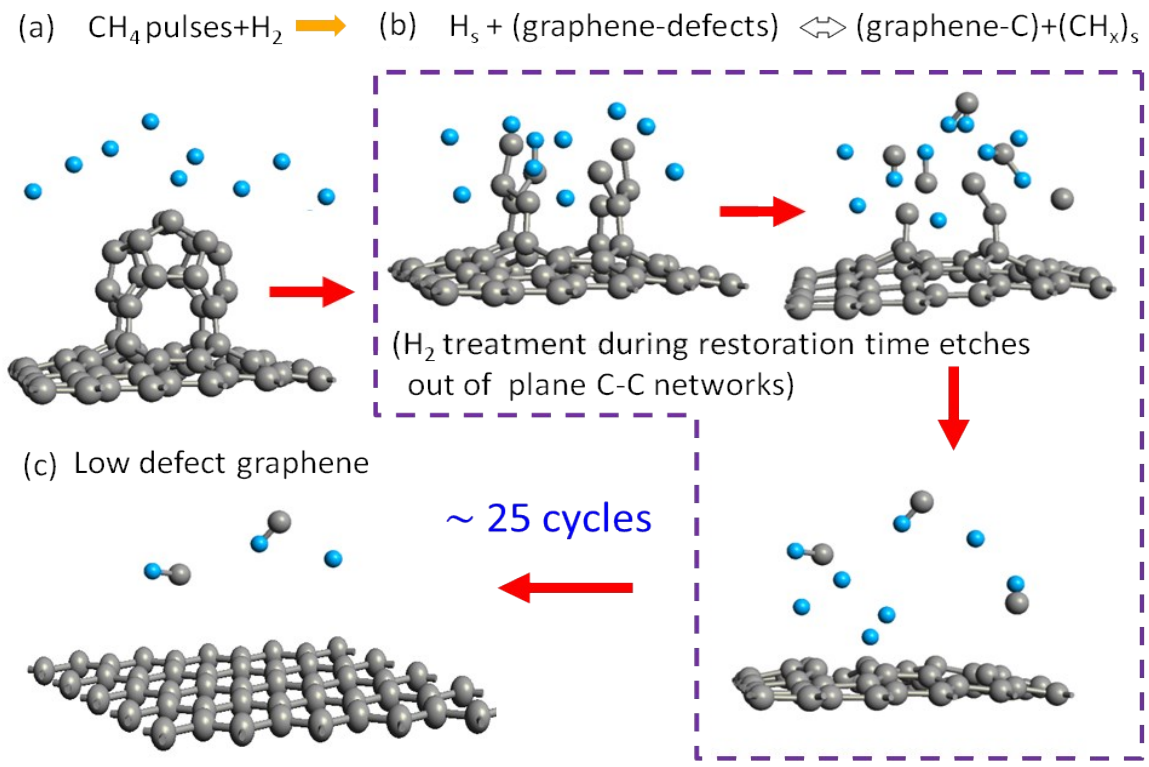


Fig. S3 Schematics of graphene growth mechanism using CH_4 pulses. (a) Evolution of surface defects emerging from unexpected bumps, protrusions, and grooves on the surface. (b) The H_2 treatment during restoration time after each CH_4 pulse etches out irregular carbon formations and leaves only sp^2 carbon networks. (c) Alternating 25 cycles of CH_4 promote the complete transformation of carbon into low defect sp^2 C-C networks.

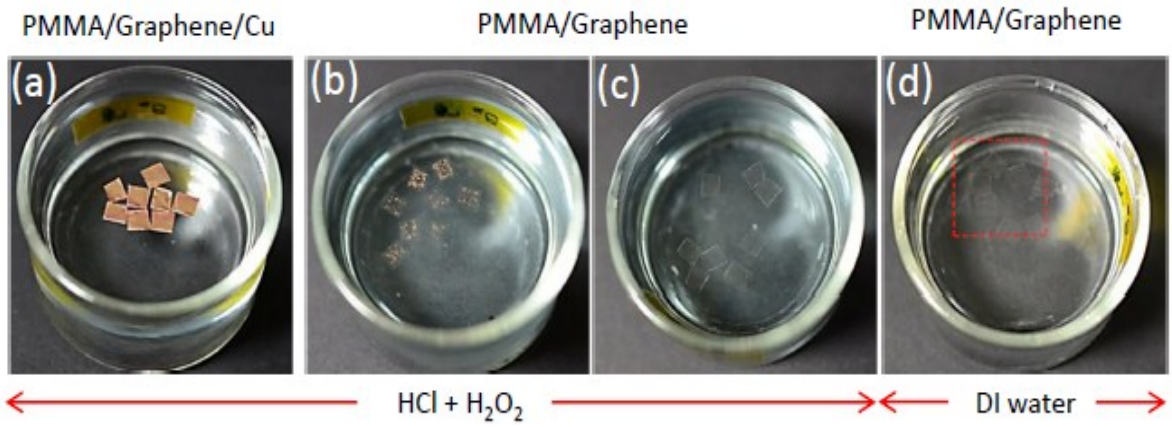


Fig S4. Optical photographs of graphene transfer process.

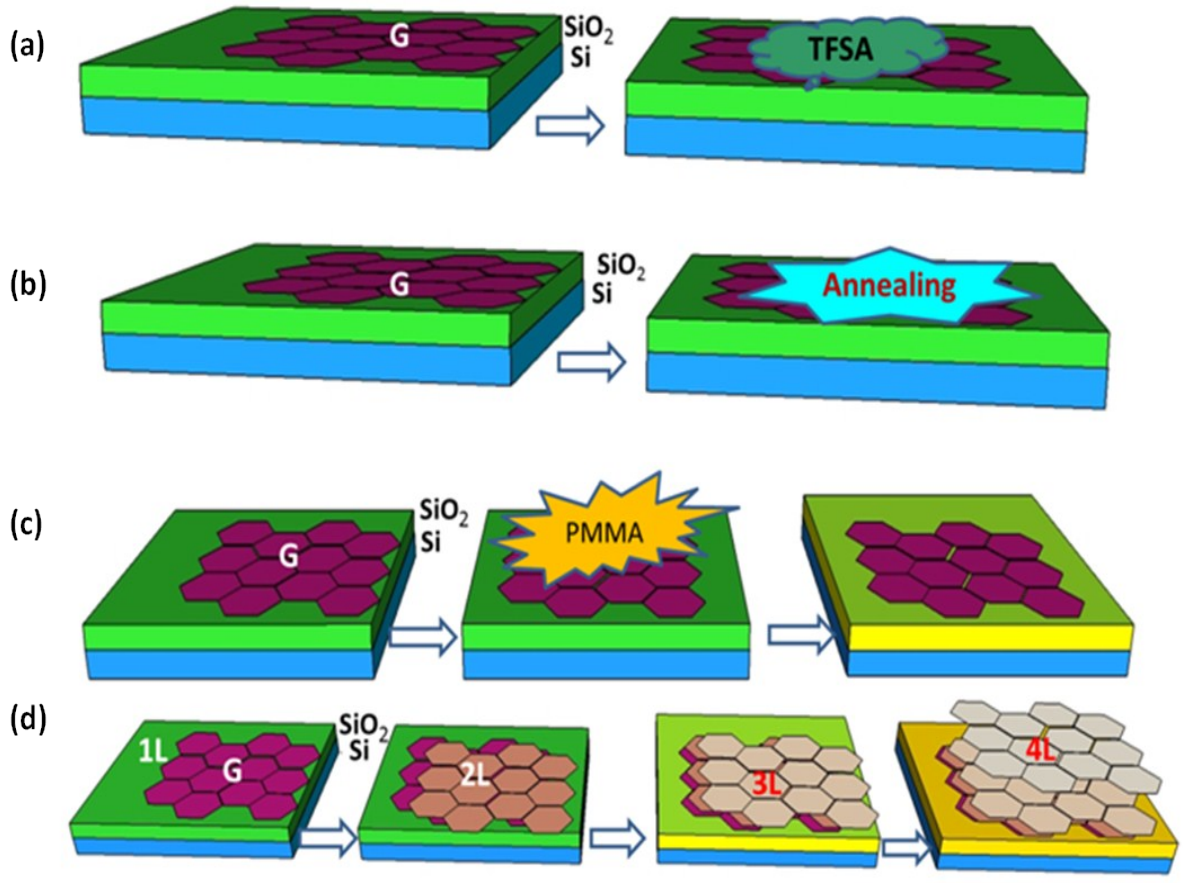


Fig. S5 Schematics showing (a) TFSA doping, (b) post annealing, (c) flattening using PMMA and (d) stacking of graphene films prepared on SiO_2 substrate.

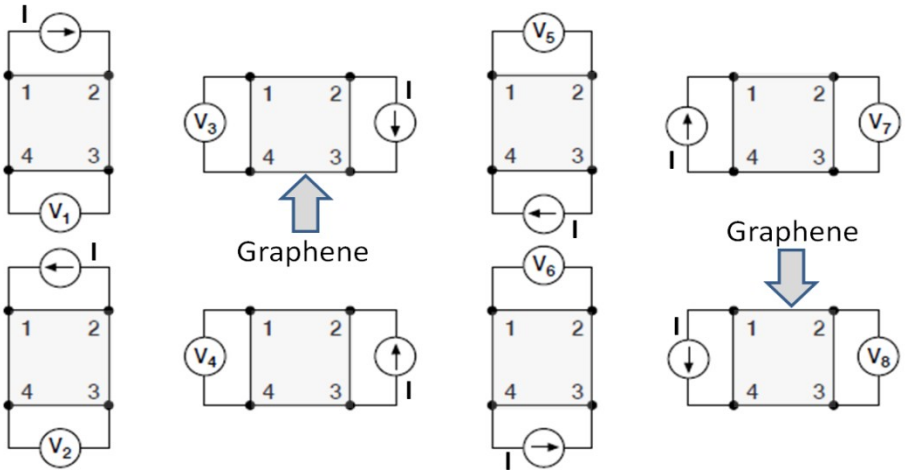


Fig. S6 Schematics of sheet resistance measurement using van der Pauw method.

The sheet resistances of prepared graphene films were measured in ambient conditions using van der Pauw method as shown in the Fig.S6. The samples were placed on a probe station with tungsten microprobes (10 μm in radius). Four probes were made on the samples using silver paste. The current was supplied using current source (Keithley 2400) and voltages were measured using voltage meter (Keithley 2000). Once all the eight voltage measurements were taken, two values of resistivity such as vertical resistivity (ρ_A) and horizontal resistivity (ρ_B) were derived as using the following formulas;

$$\rho_A=(\pi/\ln 2)\, f_A t_S\, (V_1-V_2+V_3-V_4)/4I \quad (1),$$

$$\rho_B=(\pi/\ln 2)\, f_B t_S\, (V_5-V_6+V_7-V_8)/4I \quad (2),$$

Where, t_s is the sample thickness; f_A and f_B are geometrical factors based on sample symmetry. They are related to the two resistance ratios Q_A and Q_B as shown in the following equations.

$$Q_A= (V_1-V_2)/(V_3-V_4) \quad (3),$$

$$Q_B= (V_5-V_6)/(V_7-V_8) \quad (4),$$

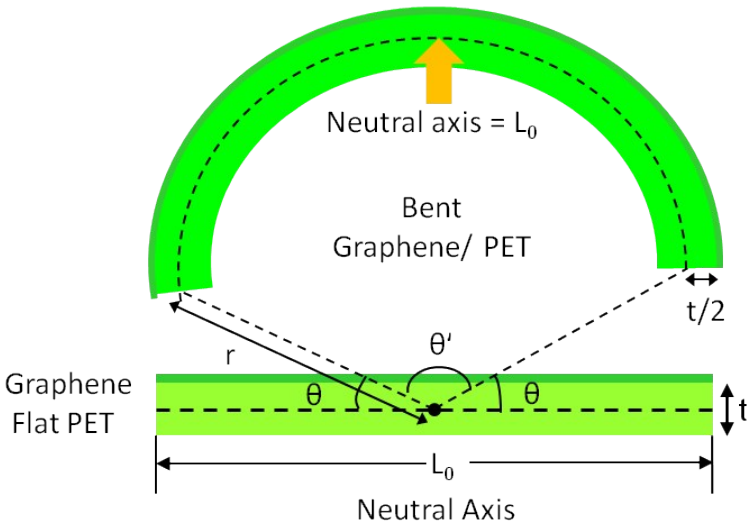
Finally, the average resistivity (ρ_{AVG}) was calculated using the formula below;

$$\rho_{AVG} = (\rho_A + \rho_B)/2 \quad (5)$$

Table S1: Reduction in sheet resistance (ΔR_s %) of graphene prepared by continuous flow method using various procedures adopted in literatures.

Methods	References	ΔR_s %
TFSA doping	Lai *et al. IEEE conf.,* 2013, 2436-2438.[32]	62-75
	Miao *et al. Nano Lett.,* 2012, 12, 2745–2750.[19]	30
	Tongay *et al. Nanotechnology,* 2011, 22, 425701.[33]	70
Annealing	Tolochko *et al. J. Phys.: Conf. Ser.* , 2017, 816, 012012.[35]	07
	Choi *et al. IEEE T. Nanotechnol.,* 2015, 14, 70-74.[36]	20-30
Stacking		
3 layer graphene	Xu *et al. Nanoscale Res. Lett.,* 2017, 12, 254.[23]	58
5 to 6 layer graphene	Choi *et al. Sci. Rep.,* 2016, 6, 24525. [24]	50
8 layer graphene	Karsy *et al. ACS Nano,* 2010, 4, 3839–3844.[7]	50

(a)



(b)

Bending angle (Degree)	Bending radius (mm)	Tensile strain (%)
18	28.66	0.02
36	12.73	0.05
54	7.43	0.08
72	4.77	0.13
90	3.18	0.19
108	2.12	0.29
126	1.36	0.45
144	0.79	0.78
162	0.35	1.77

Fig. S7 (a) Schematics of bending angle calculation. (b) Bending angle, bending radius and tensile strain values of present graphene film during flexibility test.