Supplementary Information

Time-dependent evolution of the nitrogen configuration in N-doped graphene films

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Synthesis and transfer of pristine and N-doped graphene films

All films were grown on a Cu foil using the atmospheric pressure CVD (APCVD). Initially, the Cu foil was cleaned by using an electrochemical cell with an electrolyte solution made from 300 mL distilled water, 150 mL ethanol, 150 mL ortho-phosphoric acid, 30 mL isopropanol and 3.0 g urea[38]. After cleaning, the Cu foil was placed at the centre of the cylindrical quartz tube inside a horizontal furnace. The furnace was heated to 1000 ^{o}C and then held at the designated temperature for 30 minutes. Growth of the N-doped graphene films was carried out at 2, 5, 10, and 20 min under 10 sccm flow rate of CH_4 and 5 sccm of NH_3. Similarly, pristine graphene films were grown using 10 sccm CH_4 at 2, 5, 10, 20 min. Finally the reactor was *rapidly* cooled to room temperature by pushing the quartz tube outside the reactor. The as-grown pristine and N-doped graphene films were then transferred onto a 300 nm SiO_2/Si substrate using the PMMA-assisted electrochemical delamination method [38].

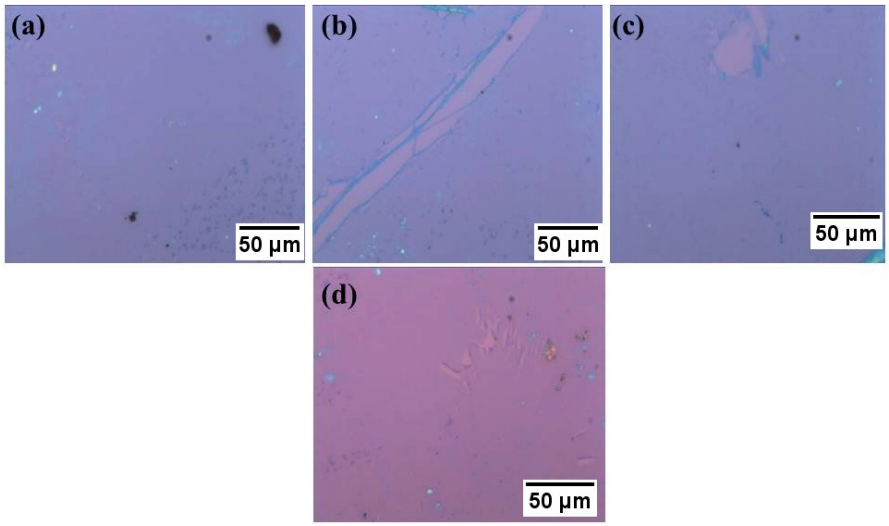
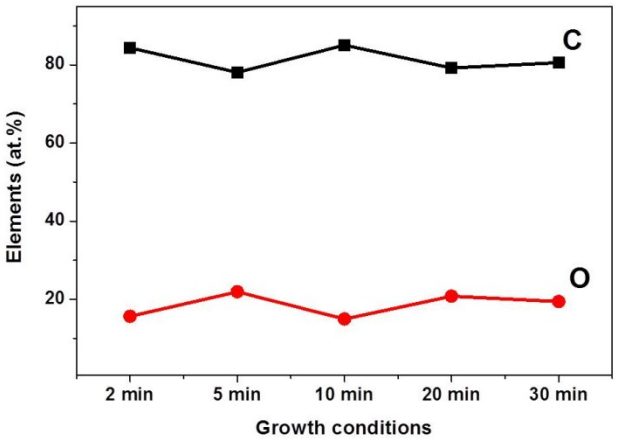


Fig S1: Optical images of pristine graphene films grown (2 min, (b) 5 min, (c) 10 min, and (d) 20 min growth times.



Sample	Elemental compositions (at. %)	
	C	O
2 min	84.4	15.6
5 min	78.1	21.9
10 min	85.1	14.9
20 min	79.2	20.8

Fig S2: Elemental plot and table of atomic compositions of pristine graphene films.

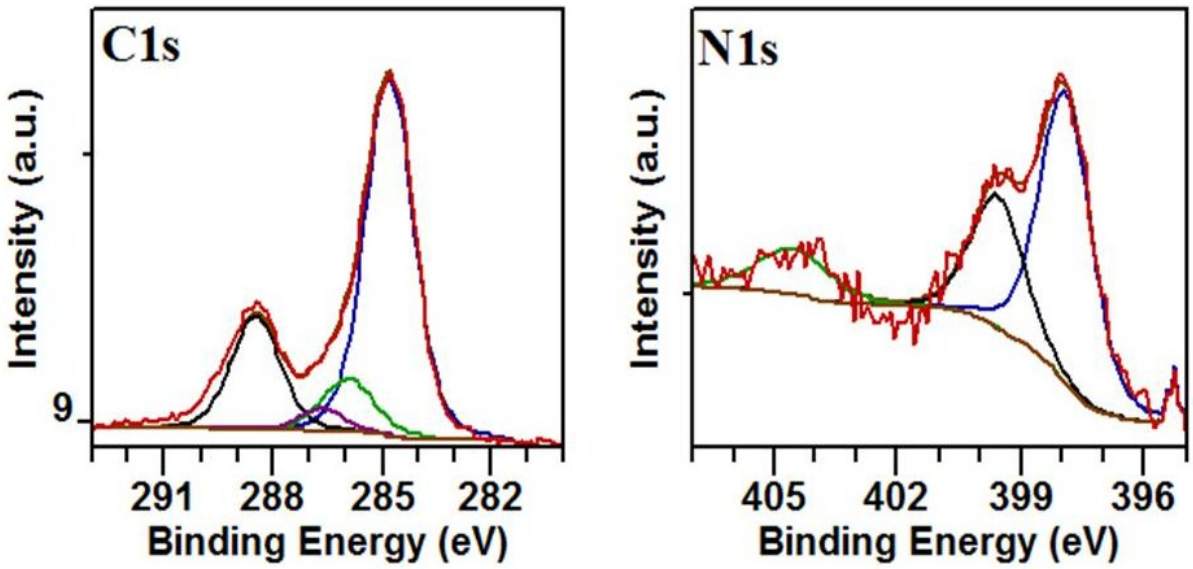


Fig S3: Core-shell XPS spectra of N-doped graphene films grown at 2 min with 10 sccm CH_4 and 5 sccm NH_3.

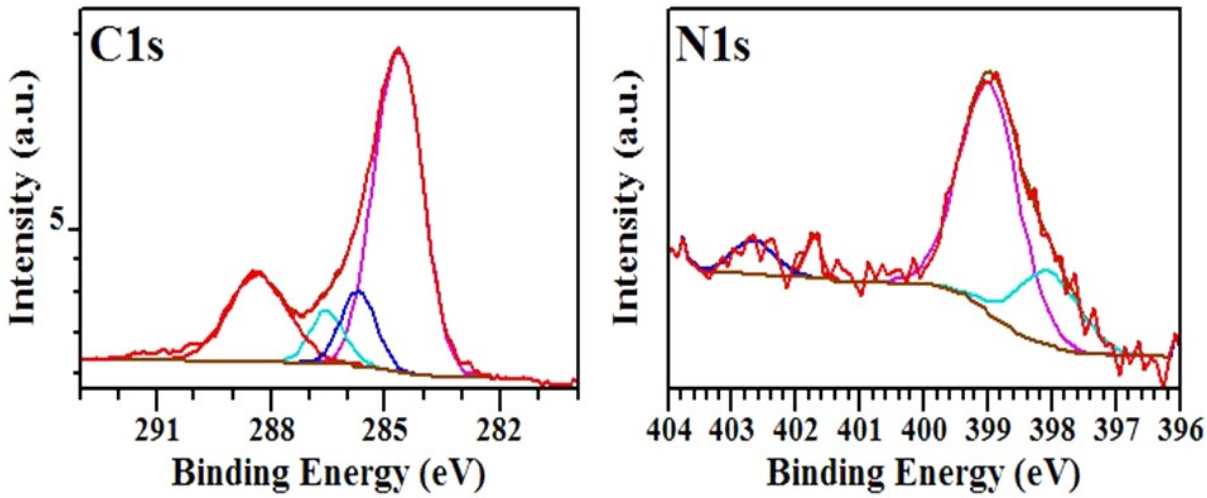


Fig S4: Core-shell XPS spectra of N-doped graphene films grown at 5 min with 10 sccm CH_4 and 5 sccm NH_3.

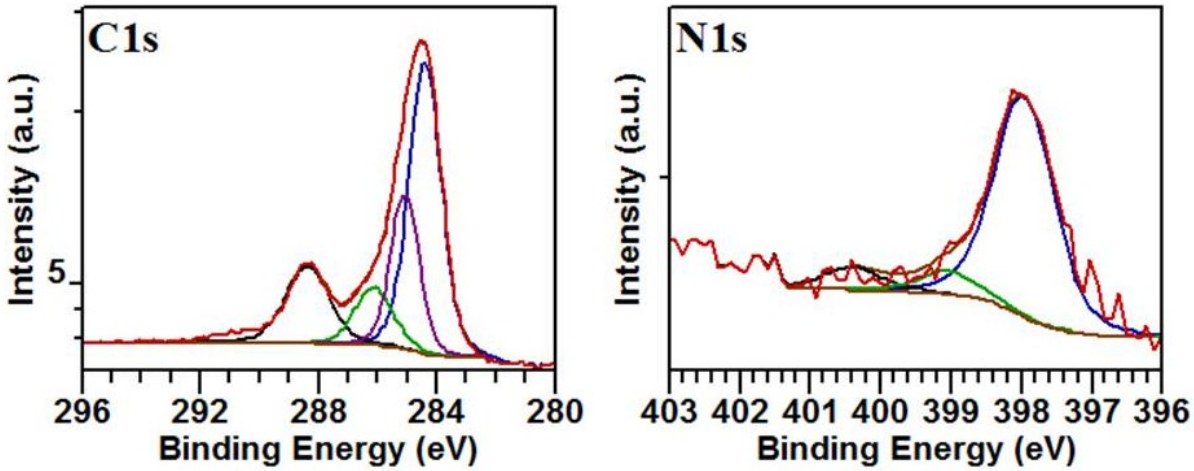


Fig S5: Core-shell XPS spectra of N-doped graphene films grown at 20 min with 10 sccm CH_4 and 5 sccm NH_3.

Table S1: Atomic compositions of N-doped graphene films

Sample	N-configurations/ N-content (N/C at. %)			
	Pyridinic-N	**Pyrrolic-N**	**Graphitic-N**	**NO_x**
2 min	2.79	1.28	0	0.61
5 min	0.82	2.51	0.13	0.28
10 min	1.24	0.081	1.83	0.11
20 min	2.36	0.24	0.24	0