Supporting Information : Selective and Regenerative Carbon Dioxide Capture by Highly Polarizing Porous Carbon Nitride

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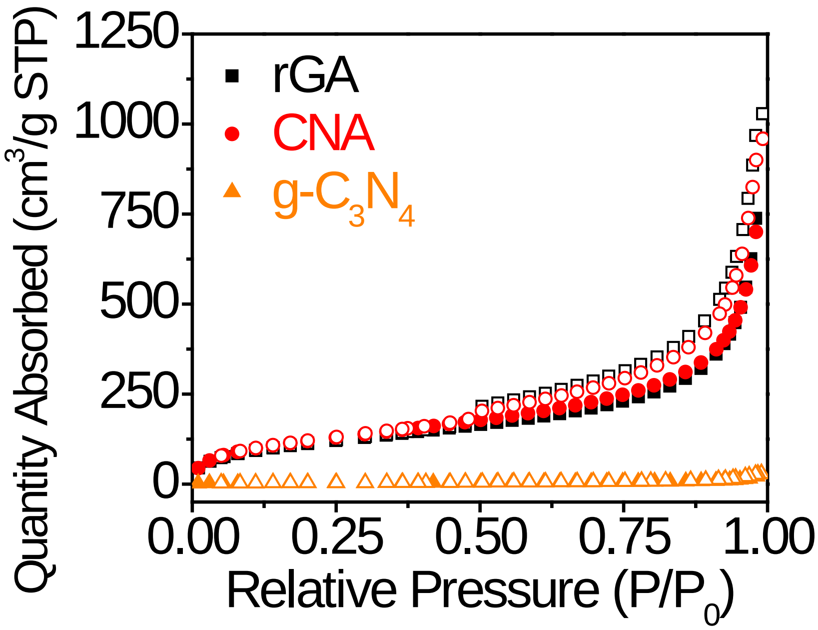
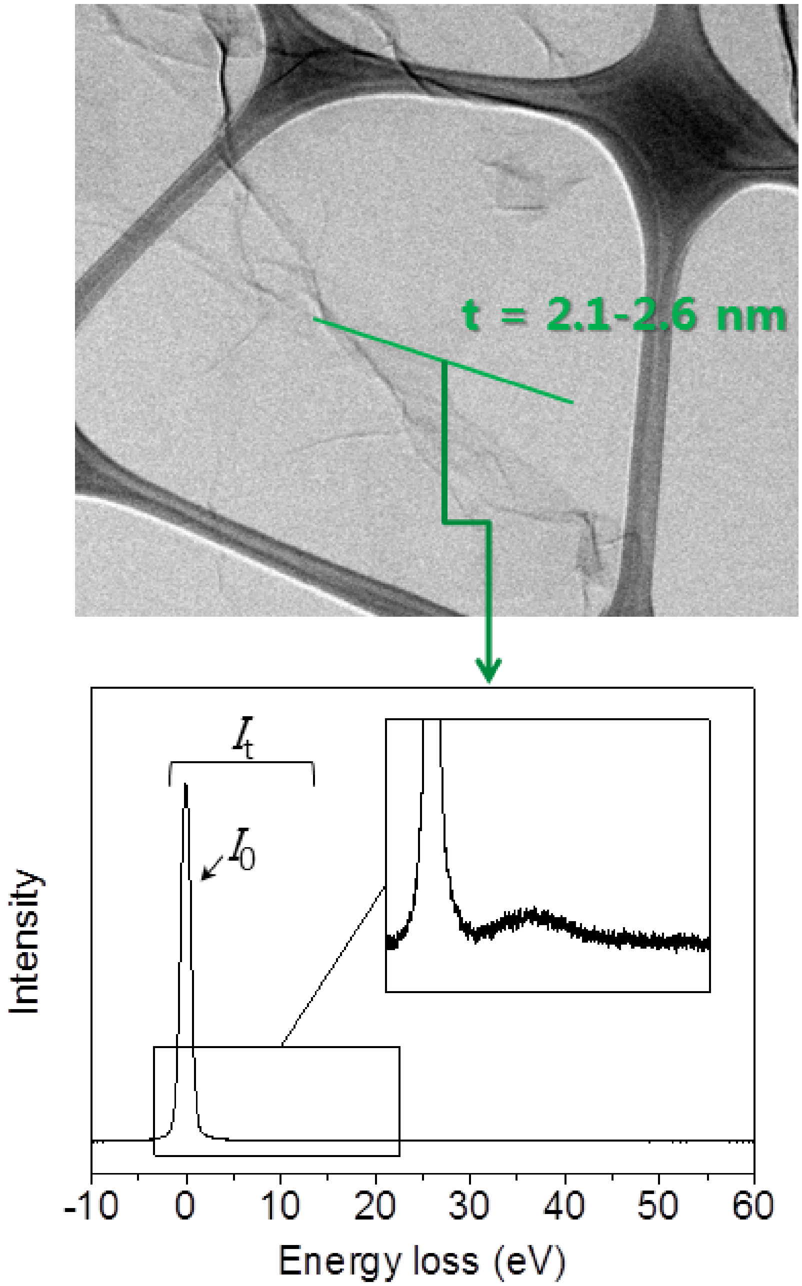
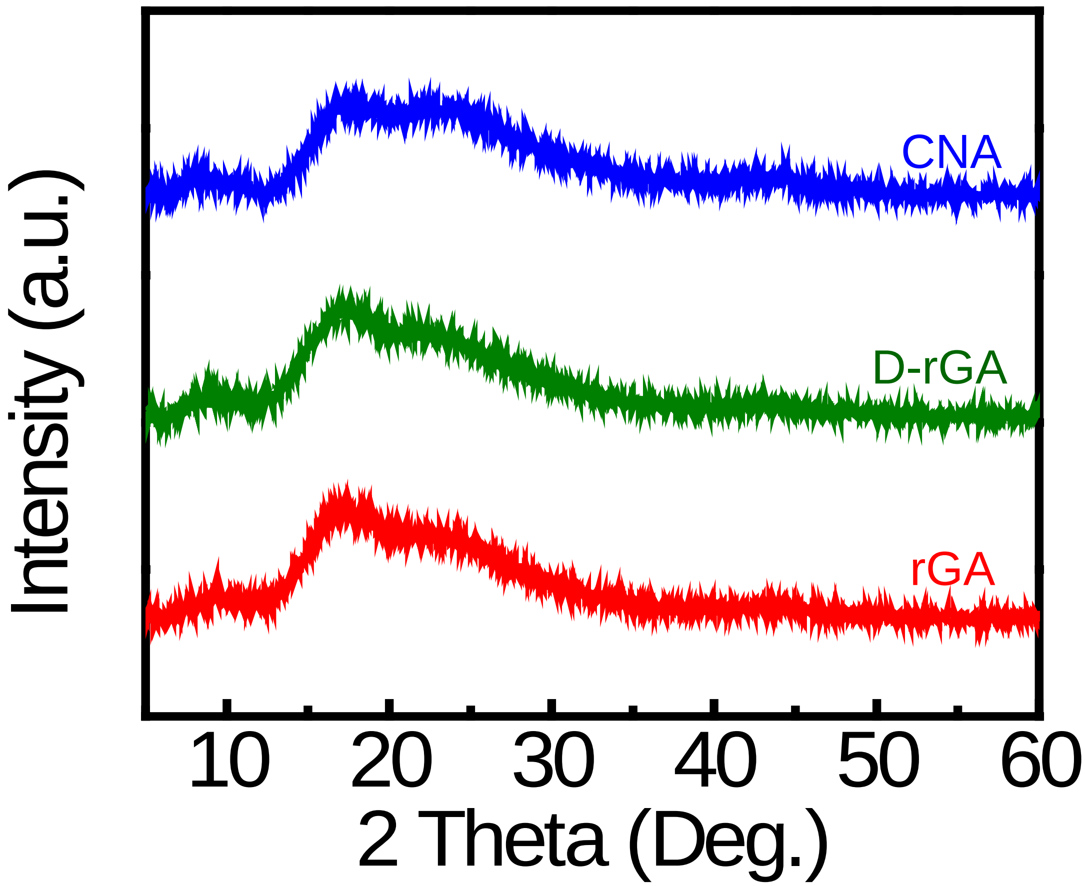


S Figure 1. Nitrogen Adsorption Isotherm of reduced graphene oxide aerogel (rGA), carbon nitride aerogel (CNA) and graphitic carbon nitride (g-C_3N_4).

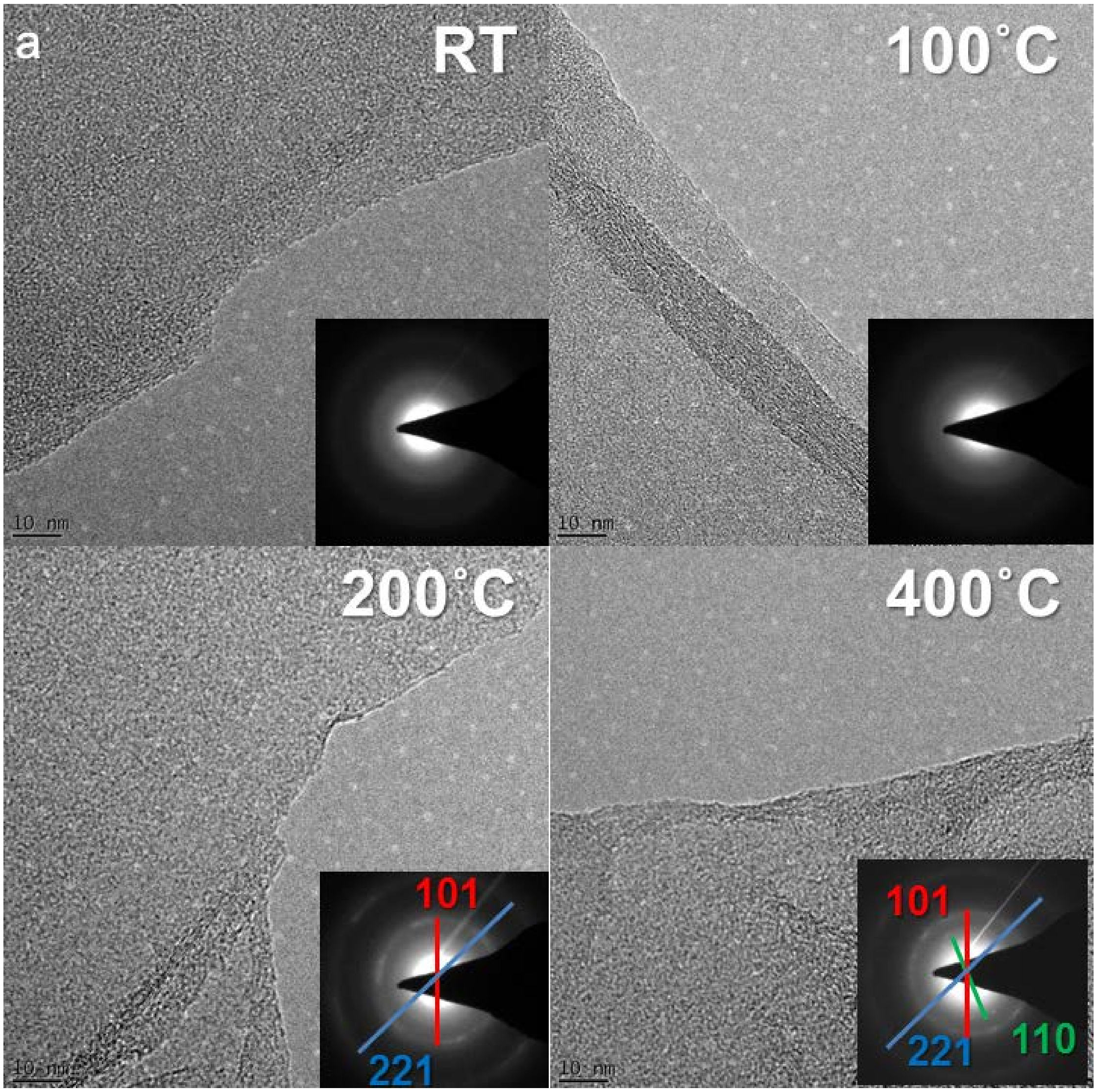


$$t = \lambda ln \frac{I_t}{I_o}$$

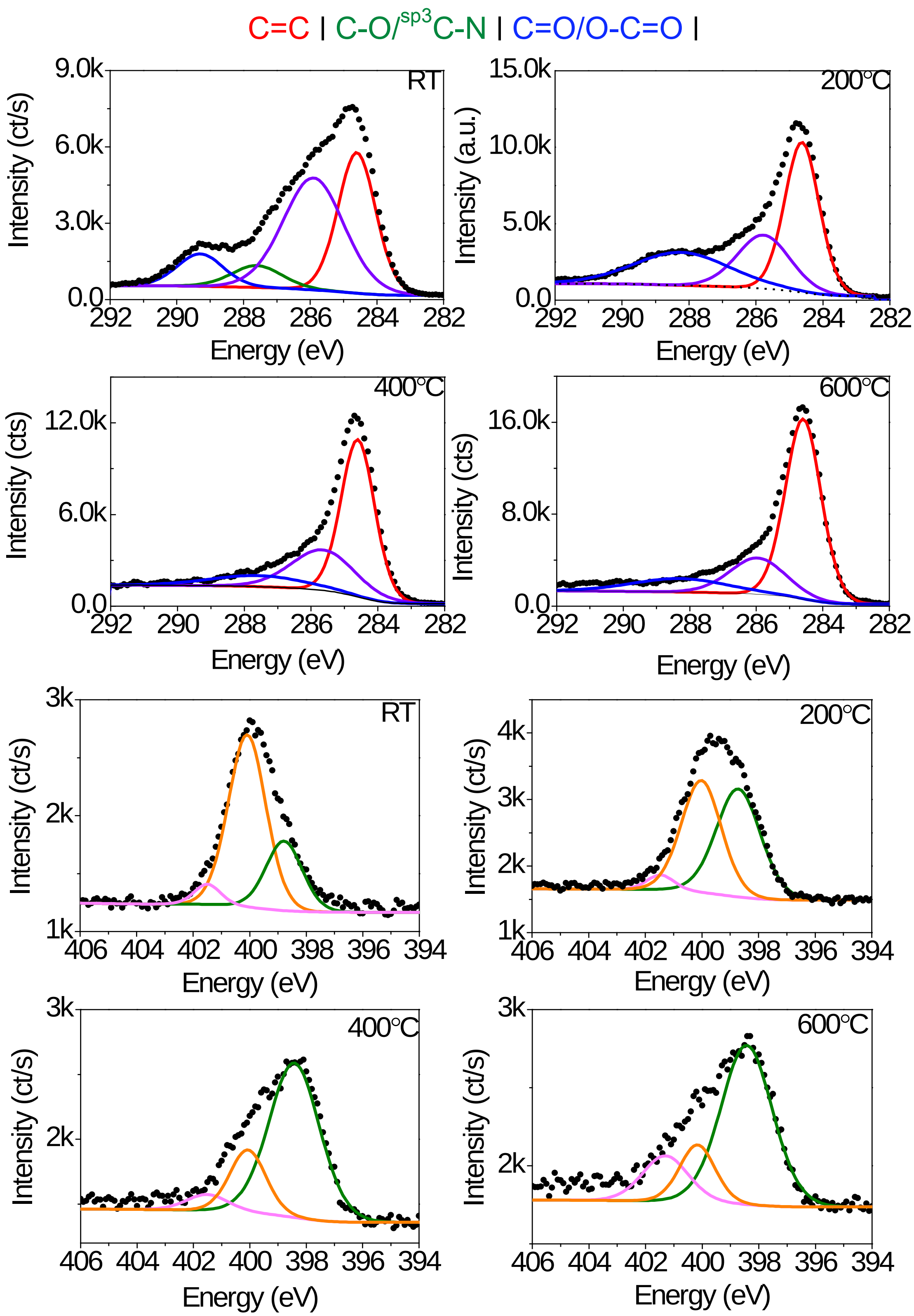
S Figure 2. TEM image with selected area of line scanning (green line) for core-loss spectrum from electron energy loss spectroscopy (EELS). Core-loss spectrum is used to measure thickness (t) of CNA, where I_0 and I_t corresponds to energy loss intensity at baseline and accumulated intensity sum in 0-20 eV range, λ as the mean path average.



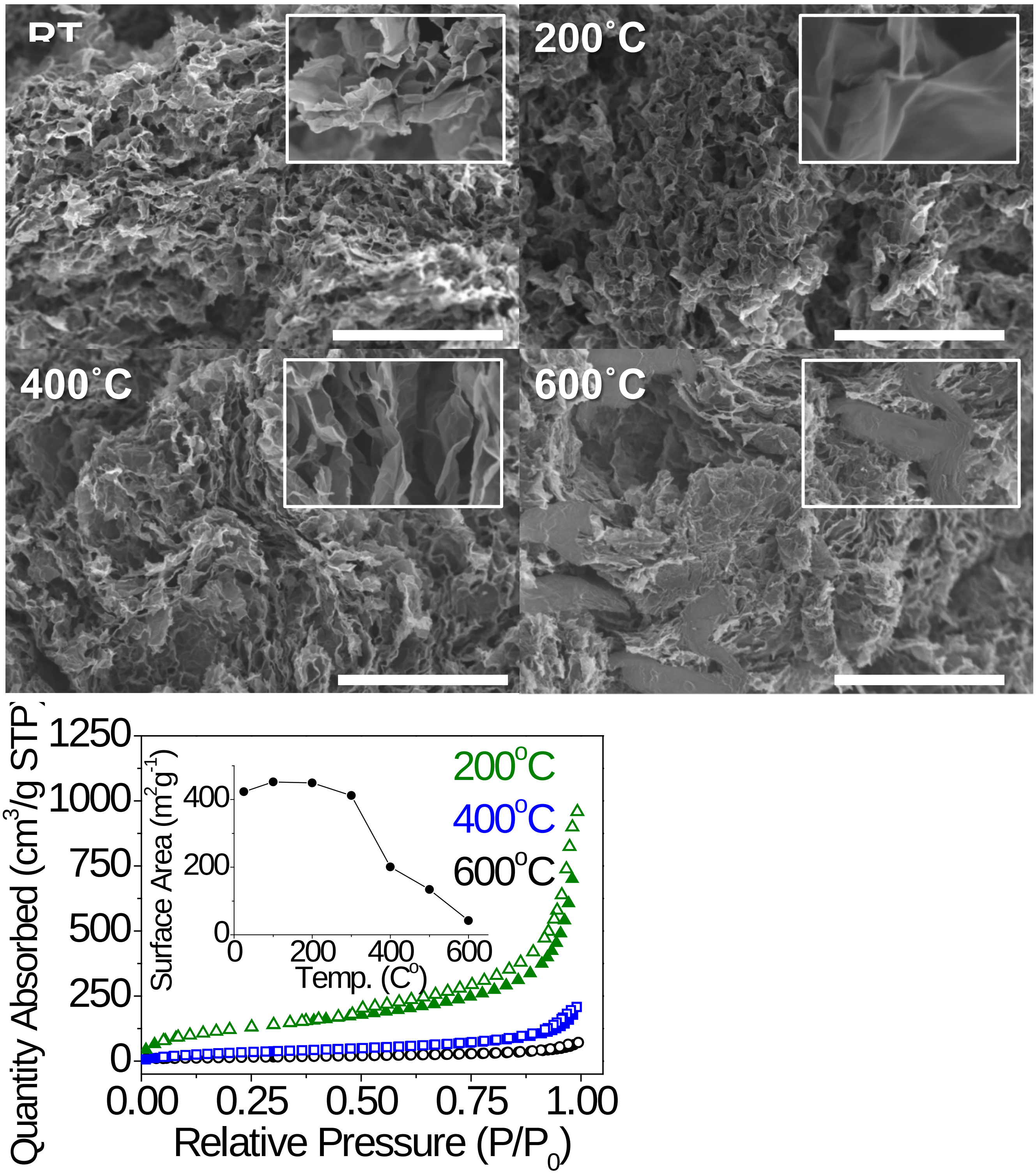
S Figure 3. PXRD pattern of CNA, D-rGA, and rGA sample showing amorphous surface of reduced graphene oxide aerogel .



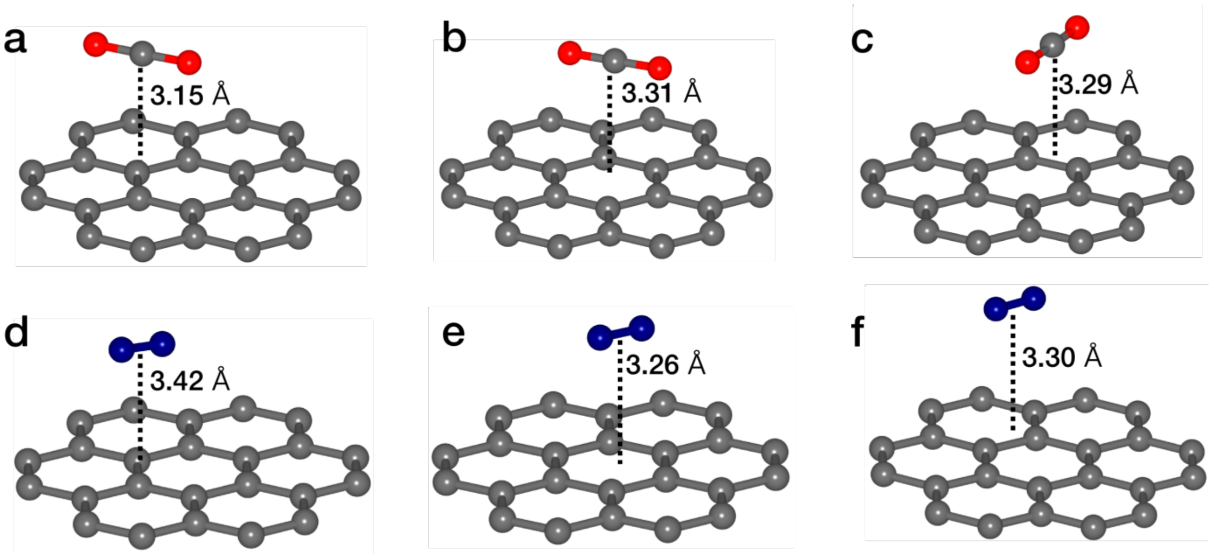
S Figure 4. TEM images and SAED (bottom right inset) of dicyandiamide functionalized reduced graphene oxide aerogel (D-rGA) transforming into carbon nitride aerogel (CNA) upon thermal treatment.


C=C | C-O/sp3C-N | C=O/O-C=O |
RT
200°C
400°C
600°C
Intensity (ct/s)
Intensity (a.u.)
Intensity (cts)
Energy (eV)

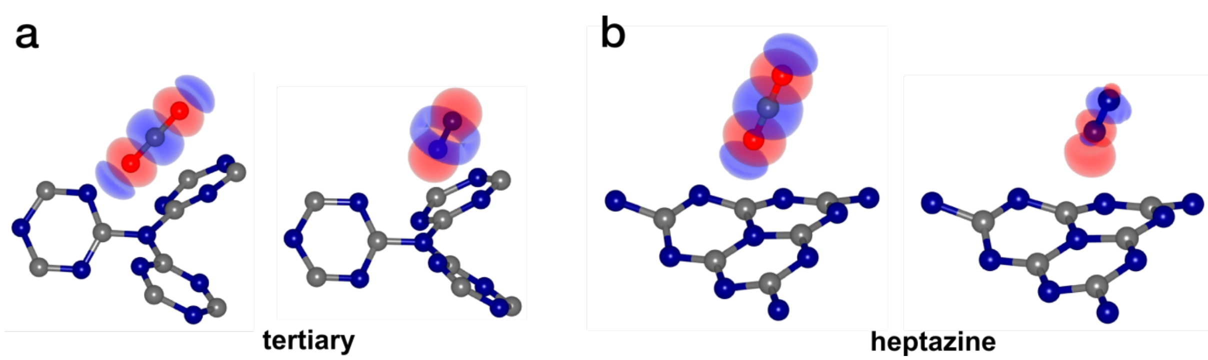
S Figure 5. XPS analysis of (top) C1s and (bottom) N1s scan showing how dicyandiamide on graphene surface transformed into condensed carbon nitride phase under temperature increase up to 600°C. Superscript ‘AP’ and ‘AR’ stands for aliphatic and aromatic state.



S Figure 6. (top) SEM analysis of CNA samples treated at various temperature conditions. Scale bar corresponds to 20 μm length. (bottom) Thermal treatment effect on surface area of CNA.



S Figure 7. Optimized geometry of CO_2 adsorptions on (a) carbon top, (b) hexagonal ring and (c) bond center of a 6 x 6 x 1 supercell of pristine graphene. Here, only a portion of the graphene substrate is shown for the sake of clarity. The corresponding optimized geometries of N_2 adsorptions are shown in (d), (e) and (f), respectively. The inset numbers represents the distance from center of gas molecule to graphene surface, in the unit of Å.



S Figure 8. (a) Iso-surface plot of the differential charge density for CO_2/N_2 adsorption at the tertiary N site. b) Same as (a) for the heptazine site of *g*-C_3N_4. Blue and red colors represent electron depletion and accumulation, respectively.

S Table 1. Experimental ATR-IR vibrational frequencies of CNA and D-rGA

	Vibrational Frequency [cm^{-1}]			
	2400-3600	1300-2400	1000-1300	600-1000
DCG	3360 [m, br] vN-H 3180 [w, br] vO-H 3020 [m, br] melamine 2941 [m, vbr] vC-H	1700, 1730 [m] vC=O (carboxylic) 1630 [vw] C=N (aliaphatic) 1550 [m] vC=C (aromatic) 1357 [w] δC-H (aromatic)	1260 [w, sh] vC-O (carbox-ylic) 1200 [m, br] vC-O-C 1100 [m, sh] vC-O 1060 [m, br] δC-C	910 [s] ωC-H 730 [s] ωN-H (amines) 620 [w] δC-C (aromatic)
CNG	3360 [m, br] vN-H 3180 [w, br] vO-H 2990[s] 2900 [m, sh] vC-H	2350 [m] vCO_2 1750 [m, br] vC=O (carboxylic) 1570 [w] vC-N (aromatic) 1530 [m, sh] vC=C (aromatic) 1480 [m] vC-N (aromatic)	1270 [m] vC-O (carbox-ylic) 1350 [w, sh] vC-N (aromatic) 1073 [m] δC-C	910 [s] ωC-H 812 [w, sh] s-triazine 736 [s] ωN-H (amines) 620 [w] δC-C (aromatic)

s : strong / m : medium / w : weak / br : broad / sh : shoulder / vw : very weak / v: vibration / δ : in-plane deformation / ω : bending