

## SUPPLEMENTARY MATERIALS

### S1. Computation of amount of aqueous ammonia and graphene oxide

The amount of aqueous ammonia (30%) used was computed using the ammonia phase diagram below. Given the pressure and temperature, density can be determined.

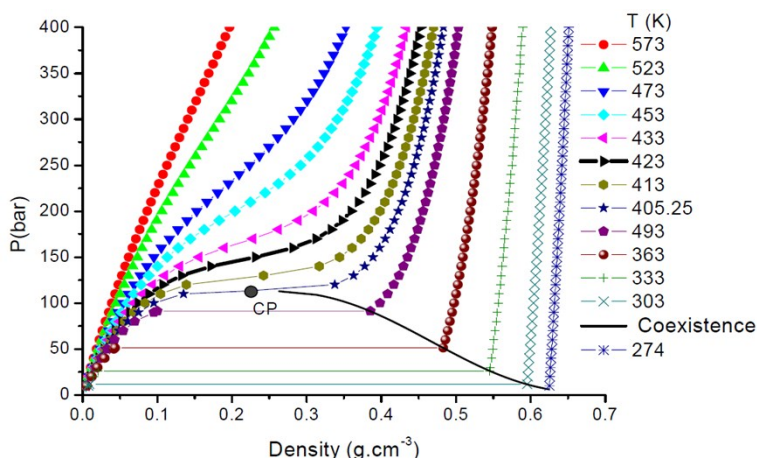


Figure S1-1. Ammonia Phase Diagram (Tassaing, 2010)

$mass\ of\ NH_3 = density \times volume\ of\ reactor$

$$mass\ of\ NH_3(aq) = mass\ of\ NH_3 \times \left(\frac{100}{30}\right)$$

$$mass\ of\ GO = mass\ of\ NH_3 \left(\frac{x\ GO}{y\ NH_3}\right)$$

Ratio of GO and NH<sub>3</sub> (x:y) were set to 0.25:3, 0.5:3 and 1:3. From preliminary runs and FTIR results, it was found that optimal ratio is 0.25:3 (see Figure S1-2). Amine peaks were more visible for the 0.25: 3 ratio.

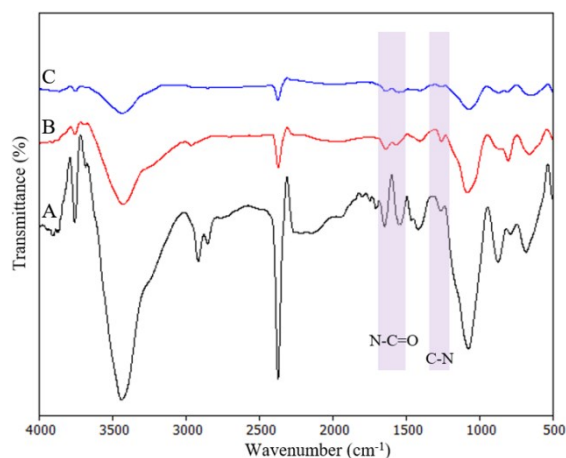


Figure S1-2. FTIR spectra showing effect of varying GO:NH<sub>3</sub> ratio: (a) 0.25:3, (b) 0.5:3, and (c) 1:3.

## S2. Optimum Pressure Evaluation

Preliminary runs were made to determine optimum value of pressure for the functionalization of GO. From the FTIR results, amine peaks were at its highest intensity at P = 30 MPa.

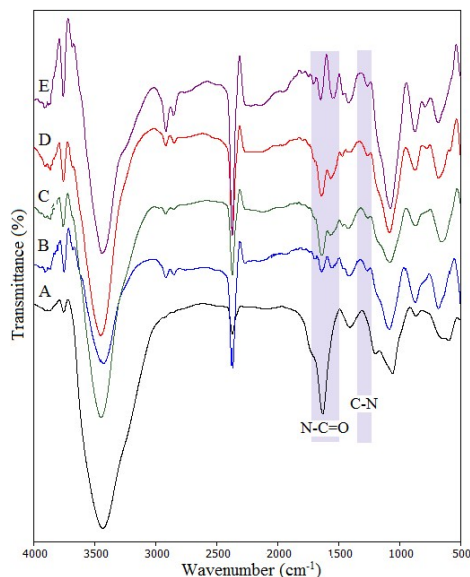


Figure S2. FTIR spectra showing effects of varying pressure: (a) GO as reference, (b) 15 MPa, (c) 20 MPa, (d) 25 MPa, and (e) 30 MPa

## S3. Optimum Temperature Evaluation

Optimum value of temperature for the functionalization of GO. From the FTIR results, amine peaks were at its highest intensity at T = 250°C.

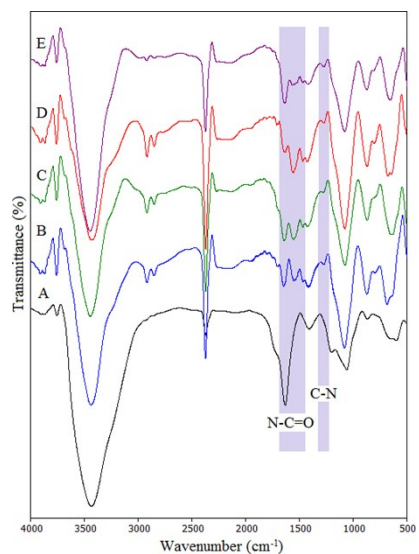


Figure S3. FTIR spectra showing effects of varying temperature: (a) GO as reference, (b) 150°C (c) 200°C, (d) 250°C, and (e) 300°C

#### S4. Optimum Run Time Evaluation

From FTR results, it was found that at longer reaction time more amine were attached to GO. However, since GO was successfully functionalized with amine groups for 1 hour, this study opted to choose a run time of 1 hour.

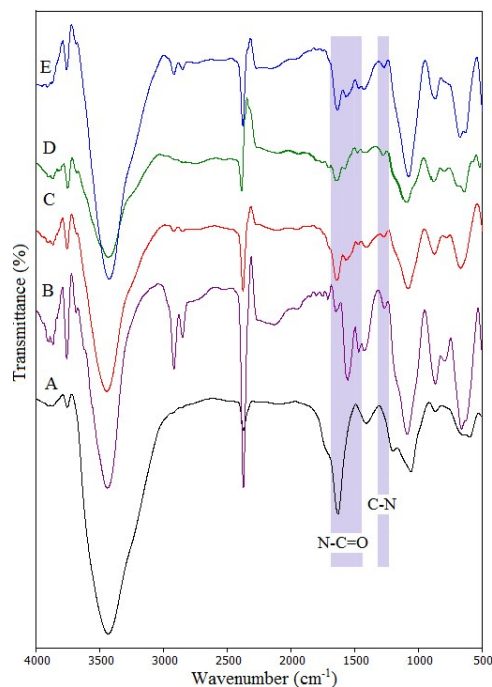


Figure S4. FTIR spectra showing effect of varying functionalization run time (a) GO as reference, (b) 4 hours, (c) 2 hours, (d) 1 hour, and (e) 30 min

#### S5. Morphological Observation

Morphologies of each sample were measured using SEM at 10,000 x modification as shown in Figure S5.

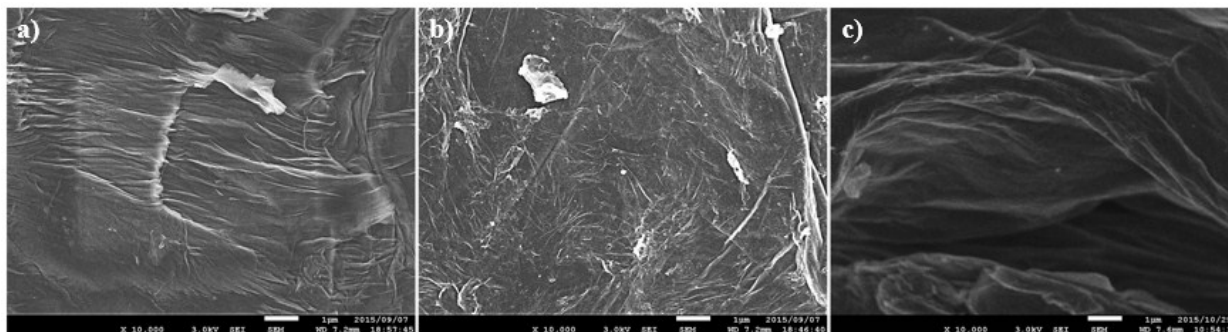


Figure S5. SEM images of (a) GO, (b) N-FGO-1, and (c) N-FGO-2

## S6. C13 NMR Results

C13 NMR data were collected to support reaction mechanism proposed in the main paper. Figures below shows NMR spectra for GO, N-FGO-1, and N-FGO-2. Further, Table S6 includes chemical shifts with corresponding type of carbon environment and examples of reaction product presented in the main paper.

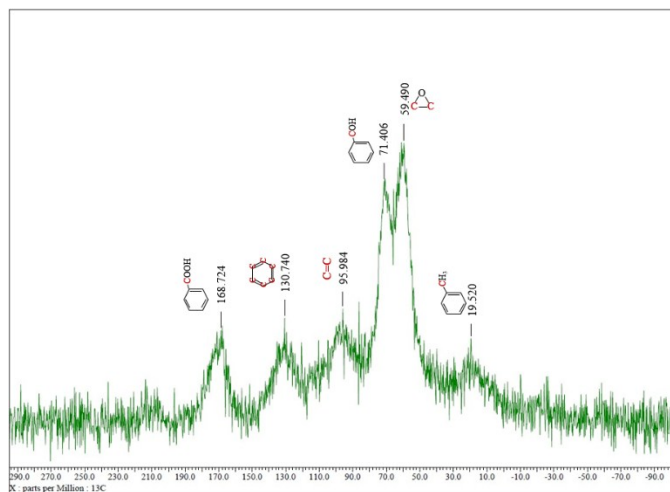


Figure S6-1. C13 NMR for GO

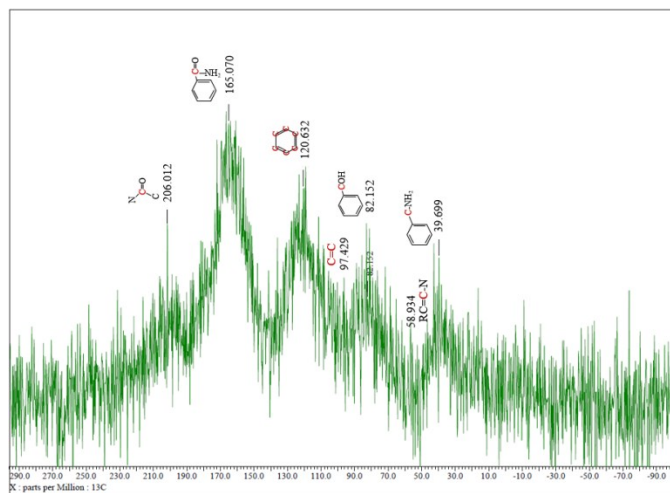


Figure S6-2. C13 NMR for N-FGO-1

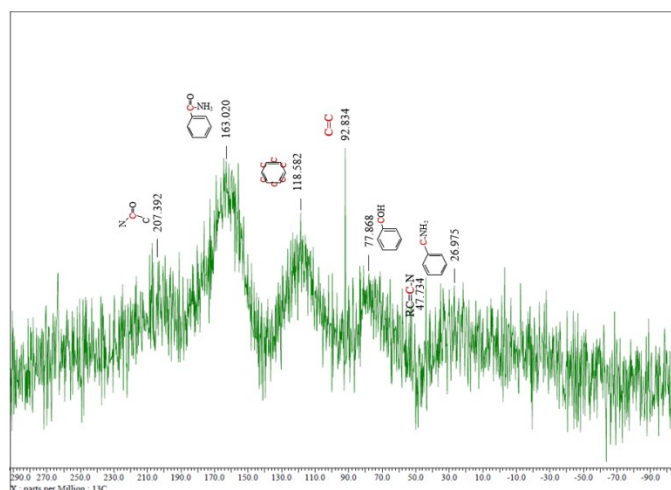


Figure S6-3.  $^{13}\text{C}$  NMR for N-FGO-2

Table S1. Chemical shifts for N-FGO-1 and N-FGO-2 and corresponding carbon environment with examples of reaction products presented in the reaction mechanism

N-FGO-1 and N-FGO-2 chemical shift (ppm)	Type of carbon environment	Corresponding product examples in the proposed reaction mechanism (main paper)
206.012 and 207.392	N-C=OC	 Imides
165.070 and 163.020	R-C=ONH <sub>2</sub>	 Amide
120.632 and 118.582	Cyclic carbon	All cyclic carbon present in the GO structure
97.429 and 92.834	C=C	All carbon with double bonds in the GO structure
82.152 and 77.868	R-COH	 R-COH
58.934 and 47.734	RC=CN	 Pyridinic N Pyrrolic N
39.699 and 26.975	R-CN <sub>2</sub>	 Amine

## REFERENCES

Tssaing, T.; Soetens, J. C.; Idrissi, A. Supercritical ammonia: A molecular dynamics simulation and vibrational spectroscopic investigation. *The Journal of Chemical Physics* 2010. DOI:10.1063/1.3506868.