Electronic Supplementary Information

Experimental Section

Materials: Graphene was purchased from Nantong Sanfeng Carbon Industry Co. Ltd.. Hydrofluoric acid (HF), sulfuric acid (H₂SO₄), hydrochloric acid (HCl) and ethanol (C₂H₅OH) were purchased from Chengdu Kelong Chemical Reagent Factory. Hydrazine hydrate (N₂H₄·H₂O), H₂O₂ (30 wt%), Nafion (5wt%) solution, sodium hypochlorite (NaClO), sodium salicylate (C₇H₅O₃Na), sodium nitroferricyanide (Na₂[Fe(CN)₅NO]·H₂O), *p*-dimethylaminobenzaldehyde (*p*-C₉H₁₁NO) were purchased from Sigma-Aldrich Chemical Reagent Co., Ltd.. Nafion 117 membrane (DuPont) was purchased from HESEN Co., Ltd. (Shanghai, China). The water used throughout all experiments was purified through a Millipore system.

Preparation of d-FG and G: d-FG was synthesized by a simple hydrothermal reaction between graphene dispersion and HF.¹ In a typical procedure, 40 mL of graphene dispersion (2 mg mL⁻¹) and 0.5 mL of HF (40 wt%) are mixed by ultrasonication for 1 min. Then, the mixture was transferred into a 50 mL Teflon-lined autoclave and maintained at 180 °C for 20, 30, 40 h, respectively. The autoclave was naturally cooled to room temperature. At last, the product was filtered using microporous membrane and throughout washed with ultrapure water, followed by freeze drying. And d-FG was obtained.

Preparation of d-FG/CP electrode: The ink was prepared by dispersing 5 mg of d-FG (30 h fluorination) catalyst dispersed into 1 mL ethanol containing 20 μ L of 5 wt% Nafion and kept ultrasonic for 1 h. Then 40 μ L of the d-FG ink was loaded on the carbon paper (1 cm × 1 cm). The d-FG/CP working electrode was prepared well.

Characterizations: Powder XRD data were acquired on a RigakuD/MAX 2550 diffractometer with Cu K α radiation (λ =1.5418 Å). The Raman spectra were collected on a Renishaw InVia Raman spectrometer under a backscattering geometry (λ = 532 nm). SEM measurements were carried out on a XL30 ESEM FEG SEM at an accelerating voltage of 20 kV. TEM measurements were performed on a HITACHI H-8100 electron microscopy (Hitachi, Tokyo, Japan) with an accelerating voltage of

200 kV. XPS data were acquired on an ESCALABMK II X-ray photoelectron spectrometer using Mg as the excitation source. The absorbance data of spectrophotometer was measured on UV-Vis spectrophotometer. A gas chromatograph (SHIMADZU, GC-2014C) equipped with MolSieve 5 A column and Ar carrier gas was used for H₂ quantifications. Gas-phase product was sampled every 1000 s using a gas-tight syringe (Hamilton). ¹H nuclear magnetic resonance (NMR) spectra were collected on a super-conducting-magnet NMR spectrometer (Bruker AVANCE III HD 500 MHz) and dimethyl sulphoxide was used as an internal to calibrate the chemical shifts in the spectra. The ion chromatography data were collected on Thermofisher ICS 5000 plus using the dual temperature heater, injection valve, conductivity detector, AERS 500 Anions suppressor. ESR data were collected by JEOL JES-FA200 on room temperature.

Electrochemical measurements: Before NRR measures, the Nafion proton exchange film was pretreated by heating in 3% H_2O_2 solution, 0.5 M H_2SO_4 and ultrapure water at 80 °C for 1.5 h, respectively. Electrochemical measurements were performed with a CHI 660D electrochemical analyzer (CH Instruments, Inc., Shanghai) in a standard three-electrode system using d-FG/CP (1.0 cm × 1.0 cm) as the working electrode, a graphite rod as the counter electrode and an Ag/AgCl electrode as the reference electrode. Electrochemical characterization of the d-FG/CP catalysts was carried out in 0.1 M Na₂SO₄ electrolytes (pH = 7). All experiments were carried out at room temperature (25 °C). For NRR, the electrolyte was bubbled with N₂ for 30 min before measurement. All potentials measured were calibrated to RHE using the following equation:

$$E_{RHE} = E_{Ag/AgCl} + 0.059 \times pH + E^{\circ}_{Ag/AgCl}$$
(E-1)

The electrode reaction of redox pairs $Fe(CN)_6^{3-}/Fe(CN)_6^{2-}$ on d-FG/CP was reversible process, so $K_3[Fe(CN)_6]$ could be used as a probe to obtain the effective surface area of modified electrode. For a reversible process, Randles–Sevcik equation[23]:

$$i = 2.69 \times 10^5 n^{2/3} A(D_0 v)^{1/2} c_0$$
 (E-2)

where i is the peak current (A), n is the number of electrons transferred, A is the electrode area (cm²), D_0 is the diffusion coefficient (cm² s⁻¹), c_0 is the bulk concentration (mol cm⁻³), and n is the scan rate (V s⁻¹).

Determination of NH₃: The produced ammonia was estimated by indophenol blue method by ultraviolet spectroscopy.² In detail, 4 mL electrolyte was removed from the cathodic chamber and added into 50 μ L oxidizing solution containing NaClO (ρ Cl = 4–4.9) and NaOH (0.75 M), followed by further adding 500 μ L coloring solution containing 0.4 M C₇H₅O₃Na and 0.32 M NaOH, and 50 μ L catalyst solution (0.1 g Na₂[Fe(CN)₅NO]·H₂O diluted to 10 mL with deionized water) in turn. After standing at 25 °C for 2 h, the UV-Vis absorption spectrum was measured. The concentration of indophenol blue was determined using the absorbance at a wavelength of 655 nm. The concentration absorbance curve was calibrated using standard ammonia chloride solution with a serious of concentrations. The fitting curve (y = 0.574x + 0.011, R² = 0.999) shows good linear relation of absorbance value with NH₄Cl concentration by three times independent calibrations.

Determination of N₂H₄: The N₂H₄ present in the electrolyte was determined by the method of Watt and Chrisp.³ The *p*-C₉H₁₁NO (5.99 g), HCI (30 mL), and C₂H₅OH (300 mL) were mixed and used as a color reagent. In detail, 5 mL electrolyte was removed from the electrochemical reaction vessel, and added into 5 mL prepared color reagent and stirred 15 min at 25 °C. The obtained calibration curve of N₂H₄ is y = 1.264x + 0.013, R² = 0.999.

Calculations of $R_{\rm NH3}$ and FE: $R_{\rm NH3}$ was calculated using the following equation:

$$R_{\rm NH3} \,(\mu g \, h^{-1} \, m g_{\rm cat.}^{-1}) = ([\rm NH_3] \times \rm V) \,/ \,(17 \times t \times m_{\rm cat.}) \tag{E-2}$$

Where $[NH_3]$ (µg mL⁻¹) is the measured NH₃ concentration; V (mL) is the volume of electrolyte; t (h) is the reaction time; m (mg) is the mass loading of catalyst on CP. FE was calculated according to following equation:

$$FE = 3 \times F \times [NH_3] \times V / (17 \times Q)$$
 (E-3)

Where F is the Faraday constant (96500 C mol⁻¹); and Q (C) is the quantity of applied electricity.

Computational details: The d-FG model is built based on a 6×6 graphene sheet. All

electron spin-polarized DFT methods implemented in the DMol³ module of Material Studio package have been employed for all present calculations.^{4,5} The generalized gradient approximation Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional is adopted.⁶ The van der Waals interaction is described using the empirical correction scheme of Grimme.⁷ To expand the electronic wavefunction, the double numerical plus polarization (DNP) basis set is used.⁸ A $5 \times 5 \times 1$ Monkhorst–Pack k-points are used in the DFT calculations. The vacuum layer of about 20 Å is used between two neighboring slabs to avoid artificial interaction. Self-consistent field (SCF) calculations are performed with a total energy convergence criterion of 10^{-6} hartree. Since bulk water layer slightly stabilizes NRR intermediates,⁹ we have therefore adopted the conductor-like screening model (COSMO) to implicitly consider solvent effects.¹⁰

 N_2 reduction reaction involves six net coupled proton and electron transfer (CPET) steps ($N_2 + 6H^+ + 6e^- \rightarrow NH_3$). The Gibbs free energy change (ΔG) of NRR is calculated by using the standard hydrogen electrode (SHE) model proposed by Nørskov et al.,^{11–13}

$$\Delta G = \Delta E + \Delta Z P E - T \Delta S + \Delta G_{pH} + \Delta G_{U}$$
(E-4)

where ΔE is the reaction energy directly obtained from DFT calculations; ΔZPE is the change in zero-point energy; T is temperature (298.15 K); and ΔS is the change in entropy. ΔG_{pH} is the correction of the H⁺ free energy by the concentration, and the value in this work is assumed as 0. ΔG_U is the free energy contribution connected to electrode potential U. The zero-point energies and entropies of the NRR species are determined from the vibrational frequencies calculations. For N₂(g) and NH₃(g), the entropies and vibrational frequencies are taken from the NIST database. [http://cccbdb.nist.gov/]

We have also calculated adsorption energy of N_2/NNH on the catalyst sheet. The adsorption energy of A species $E_{ads}(A)$ is defined as :

$$E_{ads}(A) = -[E(total) - E(surface) - E(A)]$$
(E-5)

where E(surface), E(A), and E(total) are the energies for the clean surface, A species in the gas phase, and A species adsorbed on the surface, respectively.



Fig. S1. XRD patterns of graphene and d-FG.



Fig. S2. Raman spectra of graphene and d-FG.



Fig. S3. Room-temperature ESR spectra of graphene and d-FG.



Fig. S4. EDX spectrum of d-FG.



Fig. S5. (a) XPS survey spectrum for d-FG. XPS spectra for d-FG in (b) C 1s, (c) O 1s, and (d) F 1s region. (e) XPS survey spectrum for graphene. (f) XPS spectrum for graphene in C 1s region. The C 1s binding energies (BEs) at 284.8, 285.3, 286.9, 288.8, 292.9, and 295.7 eV (Fig. S5b) are attributed to C=C/C–C, defect (attributed to carbon atoms no longer in the regular tubular structure), C–O, O–C=O, C–F, and C=C–F, respectively.¹⁴ For the O 1s region (Fig. S5c), the spectrum can be deconvoluted into the following two bands: oxygen doubly bound to carbon (C=O) at

531.6 eV and C–O at 533.2 eV.¹⁵ In F 1s region (Fig. S5d), the BEs at 685.2 and 688.1 eV are derived from C–F and C=C–F bonds, respectively.¹⁴ Compared with XPS survey spectrum of graphene (Fig. S5e and f), it can be found that hydrothermal fluorination reaction introduces O and F elements.



Fig. S6. Optical photograph of the reactor.



Fig. S7. Cyclic voltammogram of 0.1 mM $K_3[Fe(CN)_6]$ in 0.1 M KCl solution for d-FG/CP with scan rate 1 mV s⁻¹. It can be calculated that effective surface area (A_E) of d-FG/CP is 1.17 cm² through equation of (E-2).



Fig. S8. Photographs of pH indicator paper in $0.1 \text{ M Na}_2\text{SO}_4$.



Fig. S9. (a) UV-Vis absorption spectra of indophenol assays with different NH_4^+ concentrations after incubated for 2 h at room temperature. (b) Calibration curve used for calculation of NH_4^+ concentrations.



Fig. S10. (a) UV-Vis absorption spectra of various N_2H_4 concentrations after incubated for 10 min at room temperature. (b) Calibration curve used for calculation of N_2H_4 concentrations.



Fig. S11. (a) Ion chromatogram analysis for the NH_4^+ ions. (b) Calibration curve used for estimation of NH_4^+ . (c) Ion chromatogram for the electrolytes at a series of potentials after electrolysis for 4 h. (d) R_{NH3} for d-FG/CP at corresponding potentials.



Fig. S12. (a) Amounts of $H_2(n_{H2})$ from gas chromatography data of the gas from the headspace of the cell for NRR on the d-FG/CP catalyst in N₂-saturated 0.1 M Na₂SO₄ at various potentials. (b) The calculated FEs of HER and NRR. Combing the data with the obtained NH₃ selectivity, the unaccounted value may be attributed to uncontrollable experimental error and the capacitance of the support as well as dynamic hydrogen adsorption and absorption on the catalyst.¹⁶



Fig. S13. (a) Photographs of the electrolytes. (b) UV-Vis absorption spectra of the electrolytes stained with p-C₉H₁₁NO indicator after NRR electrolysis at a series of potentials.



Fig. S14. (a) Time-dependent current density curves of CP in 0.1 M $Na_2SO_4 N_2$ saturated solution at different potentials. (b) UV-Vis absorption spectra of the electrolytes stained with indophenol indicator after 2-h NRR electrolysis at different potentials.



Fig. S15. (a) Time-dependent current density curves of d-FG/CP in $0.1 \text{ M Na}_2\text{SO}_4$ Arsaturated solution at different potentials. (b) UV-Vis absorption spectra of the electrolytes stained with indophenol indicator after 2-h NRR electrolysis at different potentials.



Fig. S16. (a) UV-Vis absorption spectra of the electrolytes stained with indophenol indicator after 2-h NRR electrolysis at different potentials. (b) $m_{\rm NH3}$ of d-FG/CP in 0.1 M Na₂SO₄ N₂-saturated solution.



Fig. S17. (a) Time-dependent current density curves of d-FG/CP in 0.1 M Na₂SO₄ N₂or Ar-saturated solution at -0.7 V *vs.* RHE. (b) UV-Vis absorption spectra of the electrolytes stained with indophenol indicator after 2-h NRR electrolysis at different potentials.



Fig. S18. The ¹H NMR spectra for ${}^{15}NH_4^+$ standard sample (red curve) and electrolysis using ${}^{15}N_2$ (blue curve) as the feeding gas.



Fig. S19. (a) Time-dependent current density curves of graphene/CP in $0.1 \text{ M Na}_2\text{SO}_4$ N₂-saturated solution at different potentials. (b) UV-Vis absorption spectra of the electrolytes stained with indophenol indicator after 2-h NRR electrolysis at different potentials.



Fig. S20. EDX spectra of d-FG with (a) 20 h, (b) 30 h, and (c) 40 h fluorination reaction.



Fig. S21. (a) Time-dependent current density curves of d-FG/CP with different fluorination time in 0.1 M Na₂SO₄ solution at -0.7 V *vs.* RHE. (b) UV-Vis absorption spectra of the electrolytes stained with indophenol indicator after NRR electrolysis for 2 h. (c) $R_{\rm NH3}$ and corresponding FE of d-FG/CP with different fluorination time for NRR.



Fig. S22. (a) Time-dependent current density curves of d-FG/CP in 0.1 M Na₂SO₄ N₂-saturated solution at -0.7 V *vs.* RHE. (b) UV-Vis absorption spectra of the electrolytes stained with indophenol indicator after NRR electrolysis for 2 h. (c) Stability test of d-FG/CP during repeated NRR at -0.7 V with alternating 2-h cycles in N₂-saturated solution.



Fig. S23. Time-dependent current density curve for d-FG/CP at -0.7 V after electrolysis for 28 h in N₂-saturated solution. All experiments were carried out in 0.1 M Na₂SO₄.



Fig. S24. XRD patterns of bare CP, d-FG/CP, and post-NRR d-FG/CP.



Fig. S25. XPS spectra of d-FG in the (a) C 1s and (b) F 1s regions.



Fig. S26. (a) TEM and (b) HRTEM images of post-NRR d-FG.



Fig. S27. Optimized geometric structures of intermediates along the reaction path proceeded on the d-FG. Colour code: grey, C; blue, N; Cyan, F; white, H.

Table S1. D and G-band peak intensities (I_G) and I_D/I_G ratios for d-FG and graphene obtained by Raman spectroscopy.

Sample	I _D	I _G	I_D/I_G
d-FG	267	246	1.10
graphene	259	261	0.99

Catalyst	Catalyst Electrolyte NH ₃ yi		FE (%)	Ref.
d-FG/CP	0.1 M Na ₂ SO ₄	9.3 μg h ⁻¹ mg _{cat.} ⁻¹	4.2	This work
Mo ₂ N	0.1 M HCl	78.4 μ g h ⁻¹ mg _{cat.} ⁻¹	4.5	16
γ-Fe ₂ O ₃	0.1 M KOH	$0.212 \ \mu g \ h^{-1} \ m g_{cat.}^{-1}$	1.9	17
Fe ₂ O ₃ -CNT	0.1 M KHCO ₃	$0.22 \ \mu g \ h^{-1} \ cm_{cat.}^{-2}$	0.15	18
Fe ₃ O ₄ /Ti	0.1 M Na ₂ SO ₄	$3.43 \ \mu g \ h^{-1} \ cm_{cat.}^{-2}$	2.60	19
Mo nanofilm	0.01 M H ₂ SO ₄	$1.89 \ \mu g \ h^{-1} \ m g_{cat.}^{-1}$	0.72	20
MoO ₃	0.1 M HCl	29.43 μ g h ⁻¹ mg _{cat.} ⁻¹	1.9	21
MoS ₂ /CC	0.1 M Na ₂ SO ₄	4.94 μ g h ⁻¹ cm _{cat.} ⁻²	1.17	22
MoN	0.1 M HCl	$18.42 \ \mu g \ h^{-1} \ cm_{cat.}^{-2}$	1.15	23
VN	0.1 M HCl	5.14 μ g h ⁻¹ cm _{cat.} ⁻²	2.25	24
Au NRs	0.1 M KOH	$1.64 \ \mu g \ h^{-1} \ cm_{cat.}^{-2}$	3.88	25
a-Au/CeO _x -RGO	0.1 M HCl	8.3 μ g h ⁻¹ mg _{cat.} ⁻¹	10.1	26
Pd/C	0.1 M PBS	4.5 $\mu g h^{-1} m g_{cat.}^{-1}$	8.2	27
Pd _{0.2} Cu _{0.8} /rGO	0.1 M KOH	$2.80 \ \mu g \ h^{-1} \ m g_{cat.}^{-1}$	4.5	28
carbon nitride	0.1 M HCl	$8.09 \ \mu g \ h^{-1} \ m g_{cat.}^{-1}$	11.59	29
PCN	0.05 M H ₂ SO ₄	$27.2 \ \mu g \ h^{-1} \ m g_{cat.}^{-1}$	1.42	30
N-doped carbon	0.1 M HCl	$15.7 \ \mu g \ h^{-1} \ m g_{cat.}^{-1}$	1.45	31
hollow Cr ₂ O ₃	0.1 M Na ₂ SO ₄	25.3 μ g h ⁻¹ mg _{cat.} ⁻¹	6.78	32
TiO ₂ -rGO	0.1 M Na ₂ SO ₄	$15.13 \ \mu g \ h^{-1} \ cm_{cat.}^{-2}$	3.3	33
Nb ₂ O ₅ nanofiber	0.1 M HCl	43.6 μ g h ⁻¹ mg _{cat.} ⁻¹	9.26	34
Fe ₂ O ₃ nanorods	0.1 M Na ₂ SO ₄	$15.9 \ \mu g \ h^{-1} \ m g_{cat.}^{-1}$	0.94	35
defect-rich MoS ₂	0.1 M Na ₂ SO ₄	29.28 μ g h ⁻¹ mg _{cat.} ⁻¹	8.34	36
B ₄ C	0.1 M HCl	$26.57 \ \mu g \ h^{-1} \ m g_{cat.}^{-1}$	15.95	37

Table S2. Comparison of the NH₃ electrosynthesis activity for d-FG/CP with other aqueous-based NRR electrocatalysts at ambient conditions.

Table S3.	The amount of the	element in d-FG w	ith the different	fluorination r	eaction
time.					

Time	C content (%)	F content (%)
20 h	97.86	0.98
30 h	94.02	3.12
40 h	90.01	5.31

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