

Electronic Supplementary Information (ESI) for

Graphitic nitrogen in carbon catalysts important for the reduction of nitrite revealed by ^{15}N NMR spectroscopy at natural abundance

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Supplement:

Fig. S1 The calibration curves used to calibrate the concentrations of NO_2^- and NH_4^+ ion by spectrophotometer at 520 nm and 655 nm absorbance spectra, respectively.

Fig. S2 CV curves of N-doped C (N_2) and N-doped C (NH_3) on HER (a, c) and OER (b, d) in 0.05 M H_2SO_4 (pH 1) electrolyte and 0.1 M NaOH (pH 13) electrolyte. A carbon paper (c, d) instead of a Pt wire as a counter electrode in order to ensure that there is no migration of Pt ions to the working electrode during the electrocatalytic experiments, HER and OER results show that the electrocatalytic activities of N-doped C (NH_3) and N-doped C (N_2) C are similar and without majorly change.

Fig. S3 The selectivity, Faradaic efficiency and NH_4^+ yield of N-doped C (NH_3) toward nitrite reduction to ammonia.

Fig. S4 T1 NMR relaxation curve for ^{13}C in N-doped C (NH_3). The same dataset fitted with biexponential function [$A1*(1-\exp(-x/T1)) + A2*(1-\exp(-x/T2))$] provided A1 and A2 of 0.55 and 0.45, and T1 and T2 of 0.21 and 7.01 s, respectively (simple exponential function does not fit).

Fig. S5 XRD of N-doped C (N_2) and N-doped C (NH_3).

Fig. S6 SEM micrographs of N-doped C (NH_3) (a) and N-doped C (N_2) (b).

Fig. S7 TEM images of N-doped C (NH_3) before (a, b) and after (c, d) the electrocatalytic tests; (e) XPS N 1s spectra of the N-doped C (NH_3) electrode catalysts after the electrocatalytic tests. For structural characterization, we prepared the different amounts of N-doped C (NH_3) (1mg and 5mg) electrodes for electrocatalytic experiments and then physically removed the catalyst of the electrodes for TEM and XPS characterization.

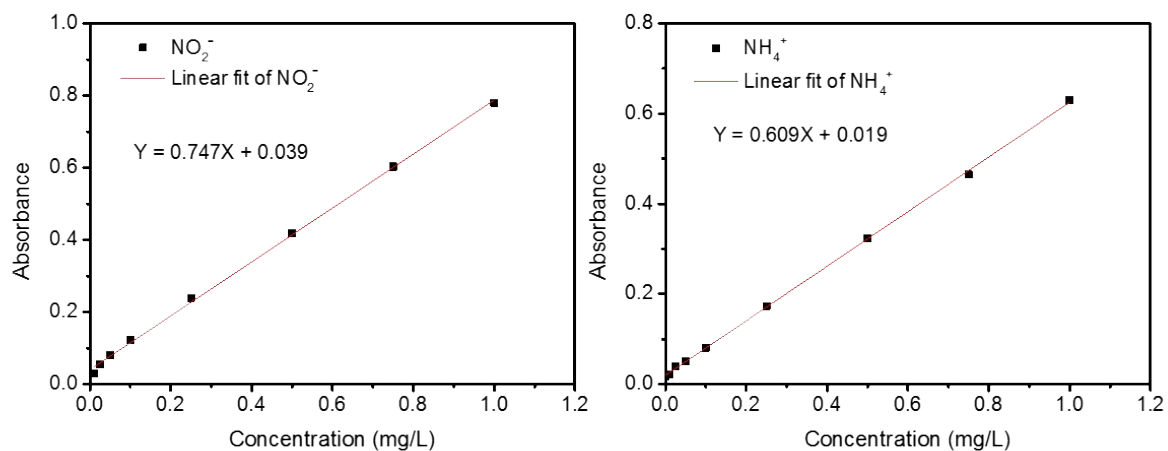


Fig. S1 The calibration curves used to calibrate the concentrations of NO_2^- and NH_4^+ ion by spectrophotometer at 520 nm and 655 nm absorbance spectra, respectively.

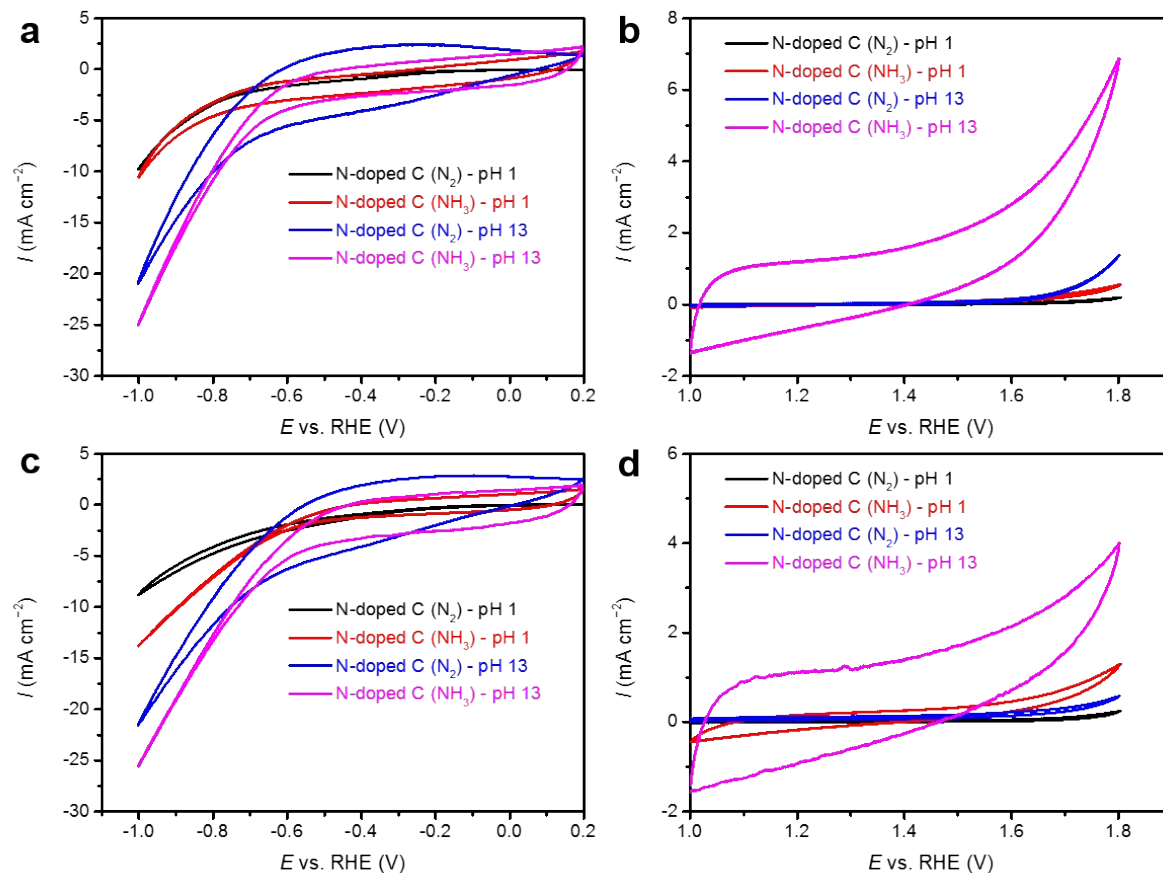


Fig. S2 CV curves of N-doped C (N_2) and N-doped C (NH_3) on HER (a, c) and OER (b, d) in 0.05 M H_2SO_4 (pH 1) electrolyte and 0.1 M NaOH (pH 13) electrolyte. A carbon paper (c, d) instead of a Pt wire as a counter electrode in order to ensure that there is no migration of Pt ions to the working electrode during the electrocatalytic experiments, HER and OER results show that the electrocatalytic activities of N-doped C (NH_3) and N-doped C (N_2) C are similar and without major change.

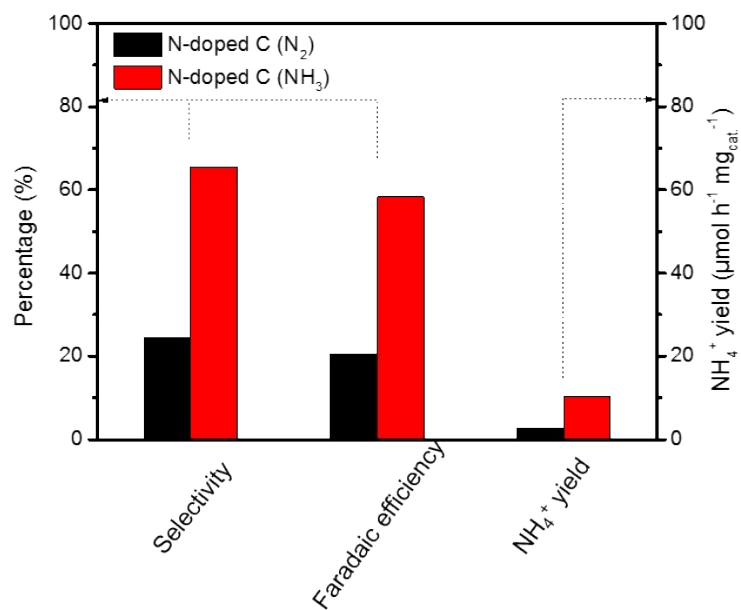


Fig. S3 The selectivity, Faradaic efficiency and NH₄⁺ yield of N-doped C (NH₃) toward nitrite reduction to ammonia.

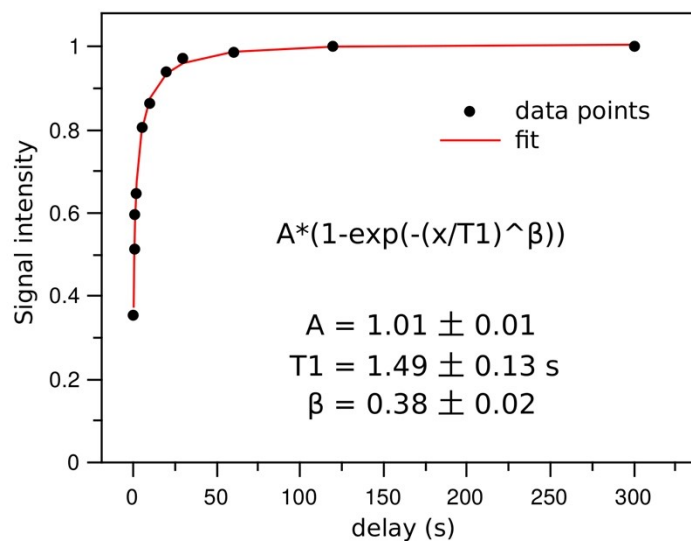


Fig. S4 T₁ NMR relaxation curve for ¹³C in N-doped C (NH₃). The same dataset fitted with biexponential function [$A1 \cdot (1 - \exp(-x/T1)) + A2 \cdot (1 - \exp(-x/T2))$] provided A1 and A2 of 0.55 and 0.45, and T1 and T2 of 0.21 and 7.01 s, respectively (simple exponential function does not fit).

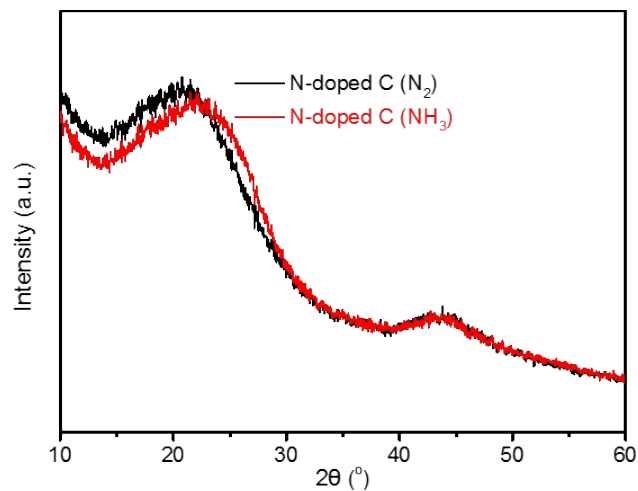


Fig. S5 XRD of N-doped C (N_2) and N-doped C (NH_3).

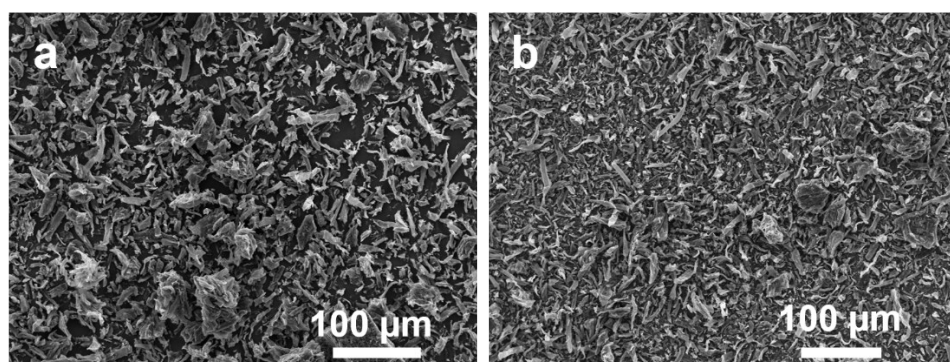


Fig. S6 SEM micrographs of N-doped C (NH_3) (a) and N-doped C (N_2) (b).

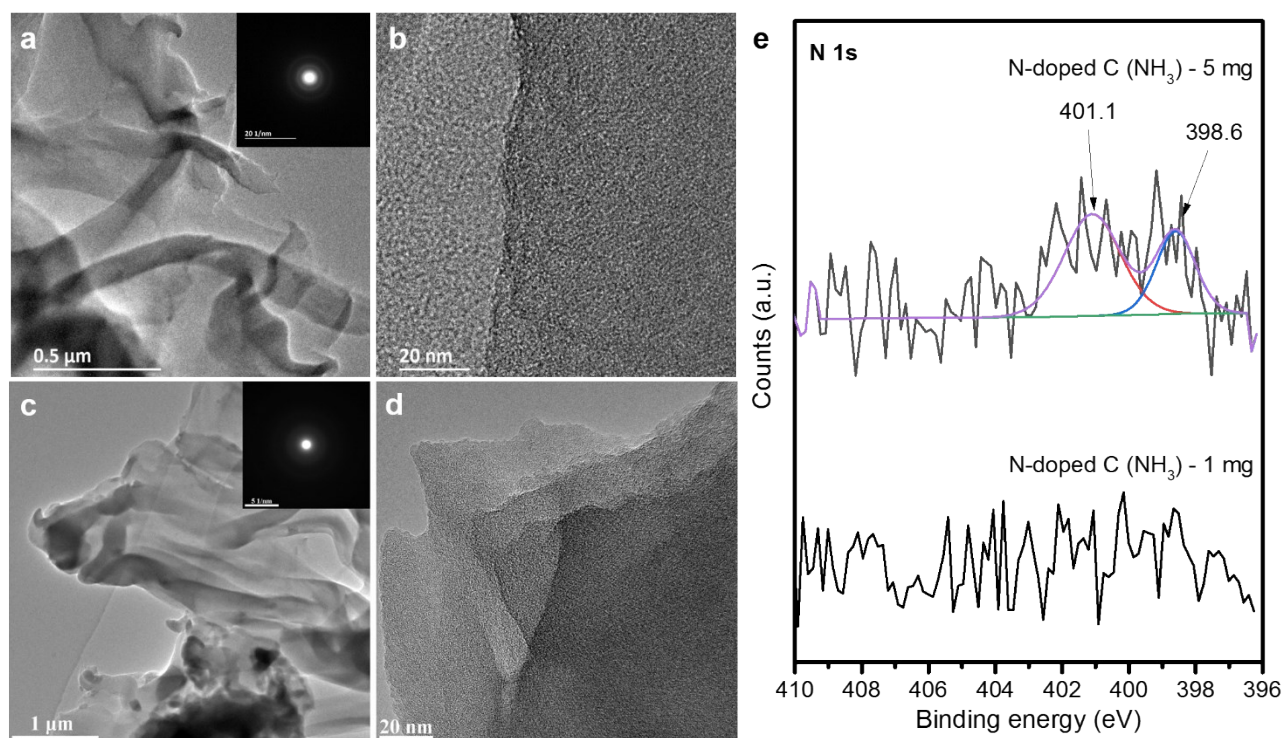


Fig. S7 TEM images of N-doped C (NH_3) before (a, b) and after (c, d) the electrocatalytic tests; (e) XPS N 1s spectra of the N-doped C (NH_3) electrode catalysts after the electrocatalytic tests. For structural characterization, we prepared the different amounts of N-doped C (NH_3) (1mg and 5mg) electrodes for electrocatalytic experiments and then physically removed the catalyst of the electrodes for TEM characterization. For XPS analysis, the N-doped carbon component was not removed from the composite electrode and Figure e shows the XPS spectrum of the complete thin film electrode.

