Supplementary Information for

Carbon doped hexagonal boron nitride as efficient metal-free catalyst for NO capture and reduction

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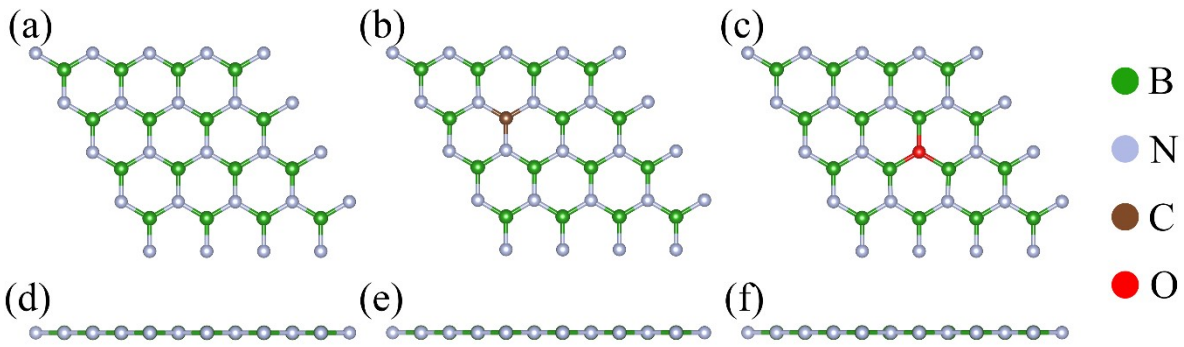

Fig. S1 Top view and side view of possible configurations of (a) h-BN, (b) C-doped h-BN, and (c) O-doped h-BN after lattice relaxation

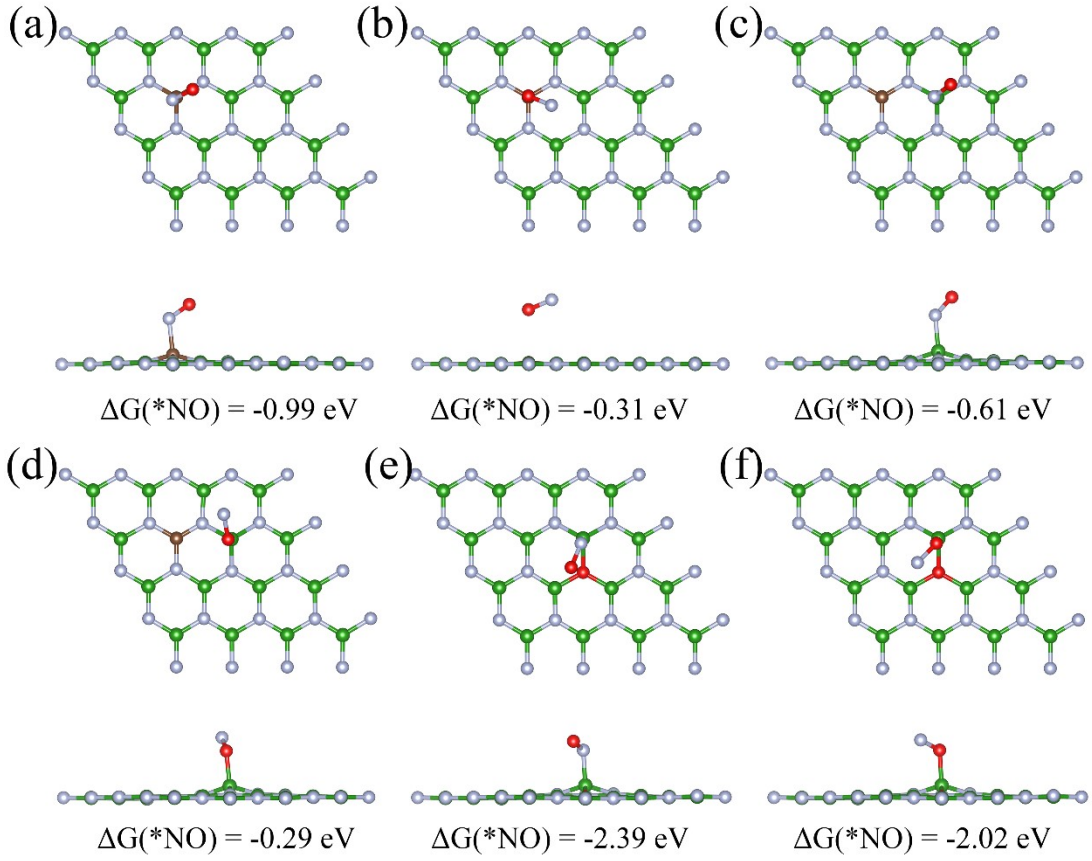

Fig. S2 Top view and side view of adsorption configurations of C_BBN and O_NBN after lattice relaxation

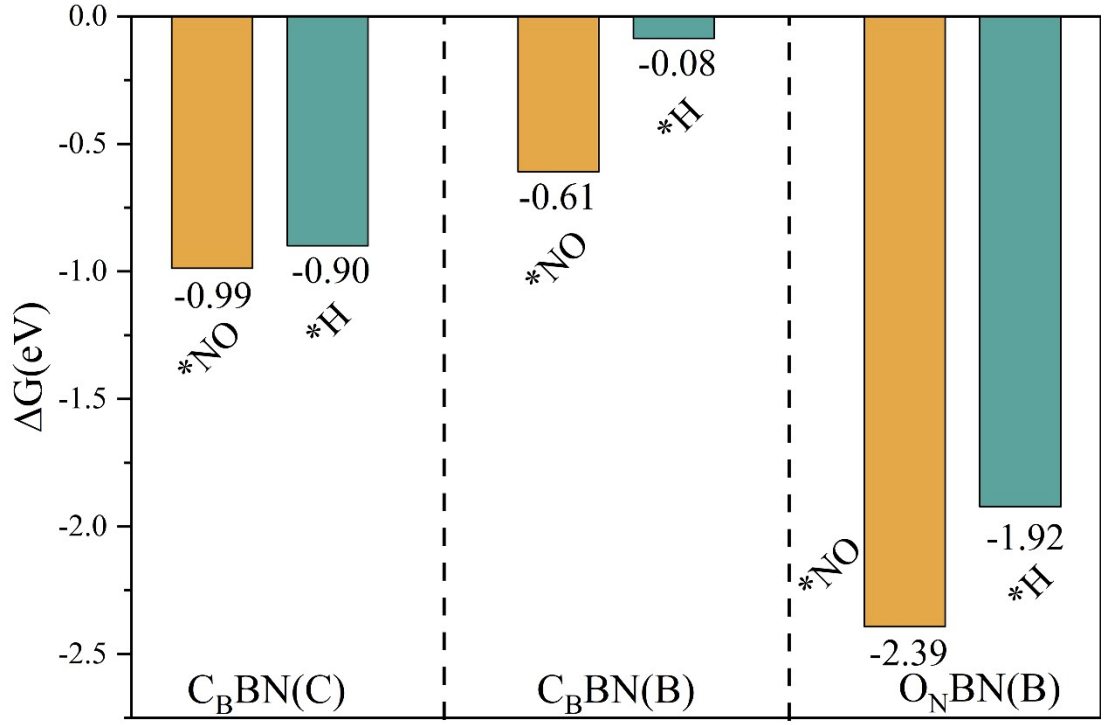

Fig. S3 Adsorption free energies of *H and *NO at C_BBN and O_NBN.

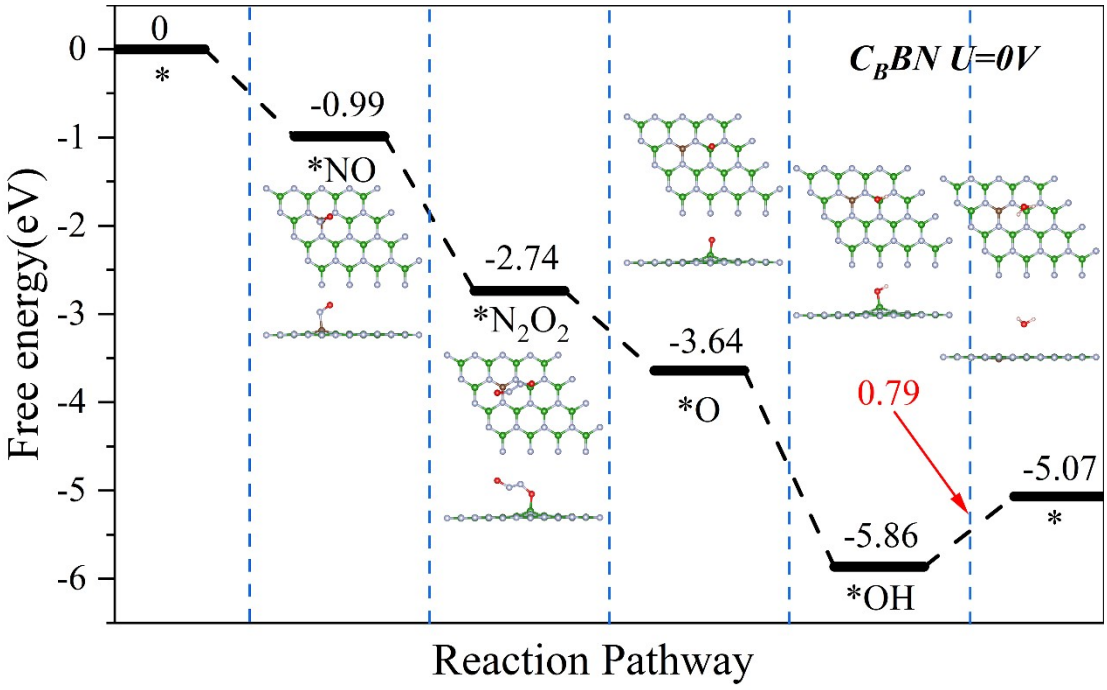

Fig. S4 The Gibbs free energy diagram for NO reduction to N_2O through N_2O_2 dimer formation on C_BBN at $U = 0$ V.

Table S1 Zero-point energy (ZPE), and entropic correction (TS, T = 298.15K) of the free molecule. The unit for energy is eV.

Gas	ΔZPE (relevant free molecule)	TS (relevant free molecule)
NO	0.12	0.66
NOH	0.35	0.69
HNO	0.36	0.69
HNOH	0.71	0.73
H_2NO	0.70	0.74
H_2NOH	1.06	0.73
NH	0.20	0.56
NH_2	0.51	0.61
NH_3	0.91	0.60
H_2O	0.57	0.59

Table S2 Zero-point energy (ZPE), and entropic correction (TS, T = 298.15K) of the intermediates for the NORR. The unit for energy is eV.

C_BBN (C site)	ΔZPE (intermediates for the NORR)	TS (intermediates for the NORR)
*NO	0.19	0.14
*NOH	0.49	0.14
*HNO	0.50	0.14
*HNOH	0.84	0.13
*H_2NO	0.83	0.09
*N	0.06	0.08
*NH	0.38	0.07

*NH_2	0.75	0.06
*NH_3	1.06	0.09

Table S3 Zero-point energy (ZPE), and entropic correction (TS, T = 298.15K) of the intermediates for the NORR. The unit for energy is eV.

C_BBN (B site)	**ΔZPE (intermediates for the NORR)**	**TS (intermediates for the NORR)**
*NO	0.27	0.08
*NOH	0.48	0.17
*HNO	0.48	0.16
*HNOH	0.81	0.17
*H_2NO	0.83	0.14
*N	0.05	0.09
*NH	0.29	0.05
*NH_2	0.70	0.09
*NH_3	0.93	0.16

Table S4 Zero-point energy (ZPE), and entropic correction (TS, T = 298.15K) of the intermediates for the NORR. The unit for energy is eV.

O_NBN (B site)	**ΔZPE (intermediates for the NORR)**	**TS (intermediates for the NORR)**
*NO	0.18	0.13
*NOH	0.49	0.14
*HNO	0.49	0.15
*HNOH	0.82	0.16
*H_2NO	0.89	0.09
*H_2NOH	1.06	0.09
*N	0.06	0.07
*NH	0.35	0.10
*NH_2	0.72	0.08

*NH_3	1.01	0.09

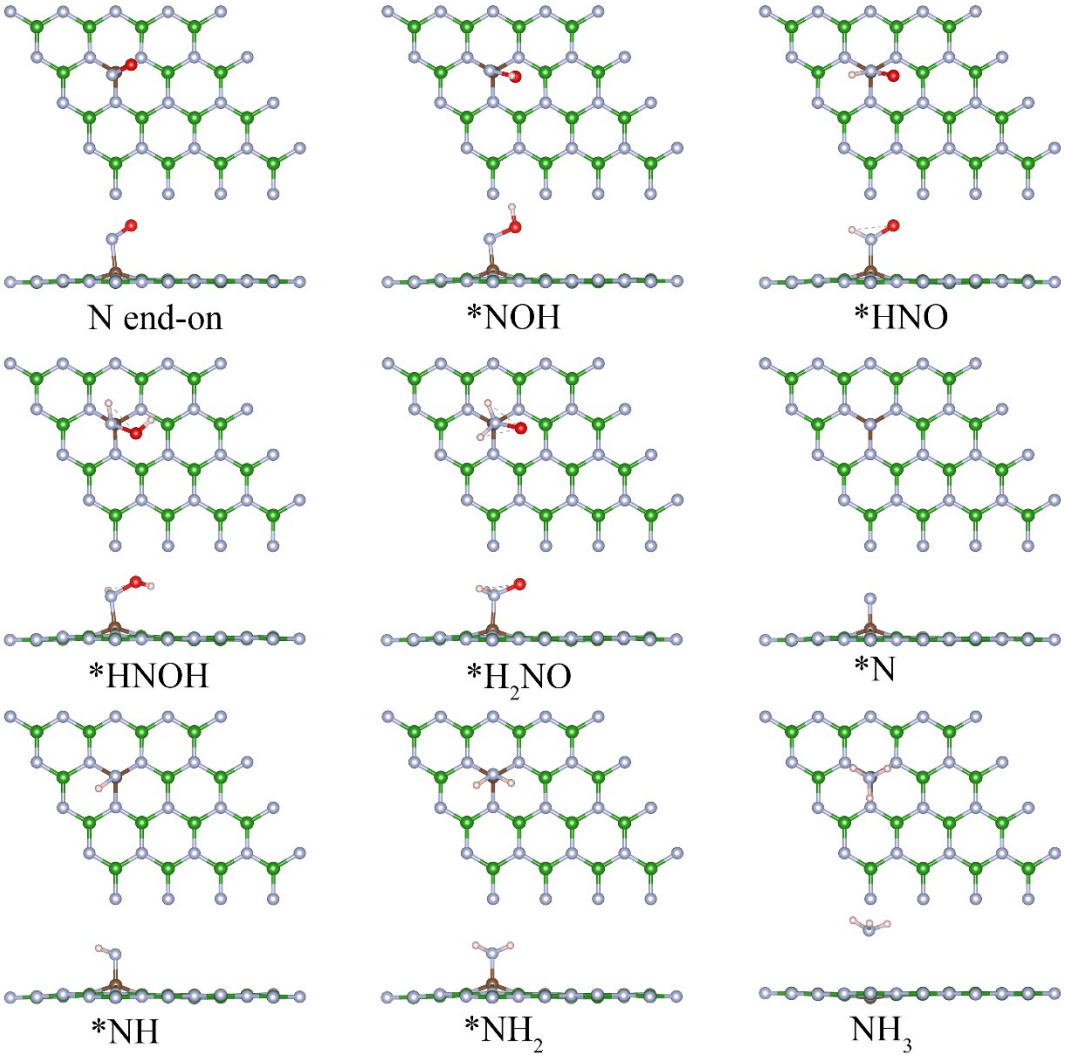

Fig. S5 The optimized configurations of all NORR intermediates on C site of C_BBN with solvent effect.

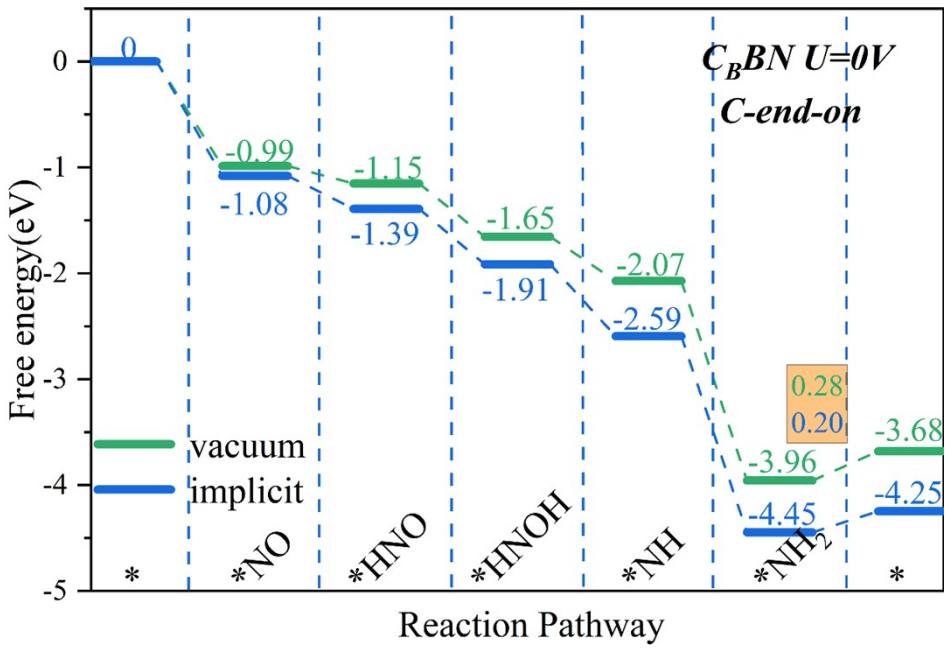

Fig. S6 NORR free energy diagrams of the considered pathways on C site with solvent effect. The applied potential is zero.

Table S5 Computed Gibbs free energy of each elementary step on the C site of C_BBN without and with solvation effects. (ΔG, units in eV)

C_BBN (C site)	**ΔG (without solvation effects)**	**ΔG (with solvation effects)**
* + NO → *NO (N end-on)	-0.99	-1.08
*NO + H^+ + e^- → *HNO	-0.16	-0.31
*NO + H^+ + e^- → *NOH	0.34	0.14
*HNO + H^+ + e^- → *HNOH	-0.50	-0.52
*HNO + H^+ + e^- → *H_2NO	-0.01	-0.07
*NOH + H^+ + e^- → *N +H_2O	-0.04	-0.25
*NOH + H^+ + e^- → *HNOH	-1.00	-0.98
*HNOH + H^+ + e^- → *H_2NOH	------	------
*HNOH + H^+ + e^- → *NH +H_2O	-0.42	-0.68
*H_2NO + H^+ + e^- → *H_2NOH	------	------
*N + H^+ + e^- → *NH	-1.38	-1.40
*H_2NOH + H^+ + e^- → *NH_2	------	------

$*NH + H^+ + e^- \rightarrow *NH_2$	-1.89	-1.85
$*NH_2 + H^+ + e^- \rightarrow * + NH_3$	0.28	0.20

Table S6 Computed Gibbs free energy of the optimal pathway for ammonia formation on the B site of C_BBN without and with solvation effects. (ΔG, units in eV)

C_BBN (B site)	**ΔG (without solvation effects)**	**ΔG (with solvation effects)**
$*+NO \rightarrow *NO$	-0.61	-0.90
$*NO + H^+ + e^- \rightarrow *HNO$	-0.66	-0.75
$*HNO + H^+ + e^- \rightarrow *HNOH$	-0.39	-0.31
$*HNOH + H^+ + e^- \rightarrow *NH + H_2O$	0.36	0.01
$*NH + H^+ + e^- \rightarrow *NH_2$	-2.65	-2.56
$*NH_2 + H^+ + e^- \rightarrow * + NH_3$	0.27	0.26

Table S7 Computed Gibbs free energy of the optimal pathway for ammonia formation on the B site of O_NBN without and with solvation effects. (ΔG, units in eV)

O_NBN (B site)	**ΔG (without solvation effects)**	**ΔG (with solvation effects)**
$* + NO \rightarrow *NO$	-2.39	-2.55
$*NO + H^+ + e^- \rightarrow *HNO$	-0.74	-0.80
$*HNO + H^+ + e^- \rightarrow *HNOH$	-0.40	-0.40
$*HNOH + H^+ + e^- \rightarrow *NH + H_2O$	-0.28	-0.60
$*NH + H^+ + e^- \rightarrow *NH_2$	-2.23	-2.16
$*NH_2 + H^+ + e^- \rightarrow * + NH_3$	2.35	2.26