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Decomposition of Nitrous Oxide on Fe-doped Boron Nitride Nanotubes: The Ligand Effect

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Natcha Injan,[a] Jakkapan Sirijaraensre[b] and Jumras Limtrakul*[bc]

Table S1. Electronic energies with zero-point energy (ZPE) and computed $\langle S^2 \rangle$ values of various spin states of $Fe_{(B)}$–BNNT and $Fe_{(N)}$–BNNT.

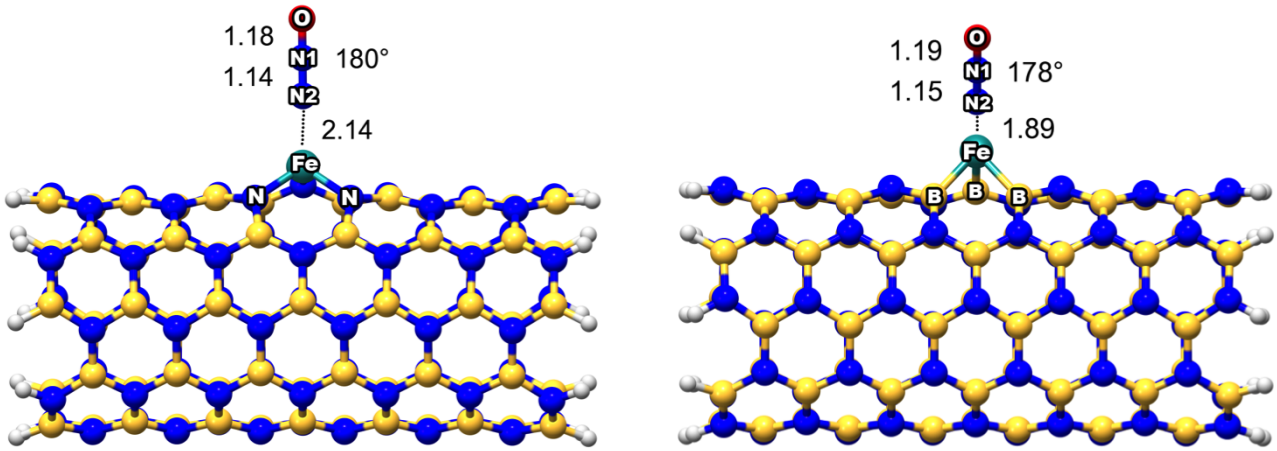
System	Spin multiplicity	Relative energy (kcal mol^{-1})	$\langle S^2 \rangle$	$\langle S^2 \rangle_{cont}$ [a]
$Fe_{(B)}$–BNNT	doublet	52.1	0.7890	5.20
	quartet	11.2	3.7523	0.06
	sextet	0.00	8.7501	0.00
$Fe_{(N)}$–BNNT	doublet	0.00	0.7861	4.81
	quartet	10.1	3.7591	0.24
	sextet	24.0	8.7503	0.00

[a] The percent spin contamination $\langle S^2 \rangle_{cont} = [\langle S^2 \rangle - S(S+1)]100/[S(S+1)]$

Table S2. Partial natural charges of the adsorption complex (q_{Ads}) and charge-transfer of all species in the adsorption complexes ($^{\Delta q_{Ads}}$) and transition states ($^{\Delta q_{TS}}$).[a]

System	q_{Ads}			Δq_{Ads}			Δq_{TS}		
	tube	Fe	N_2O	tube	Fe	N_2O	tube	Fe	N_2O
sextet $Fe_{(B)}$–BNNT:N_2O	–1.333	1.241	0.091	–0.037	–0.055	0.091	0.548	–0.069	–0.479
doublet $Fe_{(N)}$–BNNT:N_2O	0.357	–0.039	–0.318	0.442	–0.083	–0.360	0.026	–0.007	–0.019

[a] calculated from equations $\Delta q_{Ads} = q_{Ads} - q_{iso}$ and $\Delta q_{TS} = q_{TS} - q_{Ads}$ where q_{Ads}, q_{TS} and q_{iso} are charges of all species in the adsorption complexes, the transition states and the isolated N_2O and nanotubes, respectively.



Adsorption energy = –15.0 kcal mol^{-1}

(a) sextet Fe$_{(B)}$–BNNT

Adsorption energy = –26.4 kcal mol^{-1}

(b) doublet Fe$_{(N)}$–BNNT

Fig. S1 Geometrical structures and adsorption energy with ZPE correction of N-bound complexes over (a) sextet Fe$_{(B)}$–BNNT and (b) doublet Fe$_{(N)}$–BNNT. Distances and angles are given in Å and degrees, respectively.

Table S3. Relative energies with zero-point energy (ZPE) of N_2O decomposition on $Fe_{(B)}$–BNNT and $Fe_{(N)}$–BNNT.

	Relative energy (kcal mol^{-1})		
	Doublet state	Quartet state	Sextet state
$Fe_{(B)}$–BNNT + N_2O	52.1	11.2	0.0
Ads	5.2	2.1	-12.9
TS	15.5	9.6	12.3
Prod	-43.3	-39.2	-14.4
$FeO_{(B)}$–BNNT + N_2	-40.9	-36.4	-15.4
$Fe_{(N)}$–BNNT + N_2O	0.0	10.1	24.0
Ads	-25.0	-3.1	1.9
TS	-25.2	-19.2	4.8
Prod	-51.3	-67.3	-32.8
$FeO_{(N)}$–BNNT + N_2	-48.7	-64.7	-32.5