Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Single transition metal doped monolayer C_9N_4 as an efficient catalyst for CO oxidation: A first-principles study

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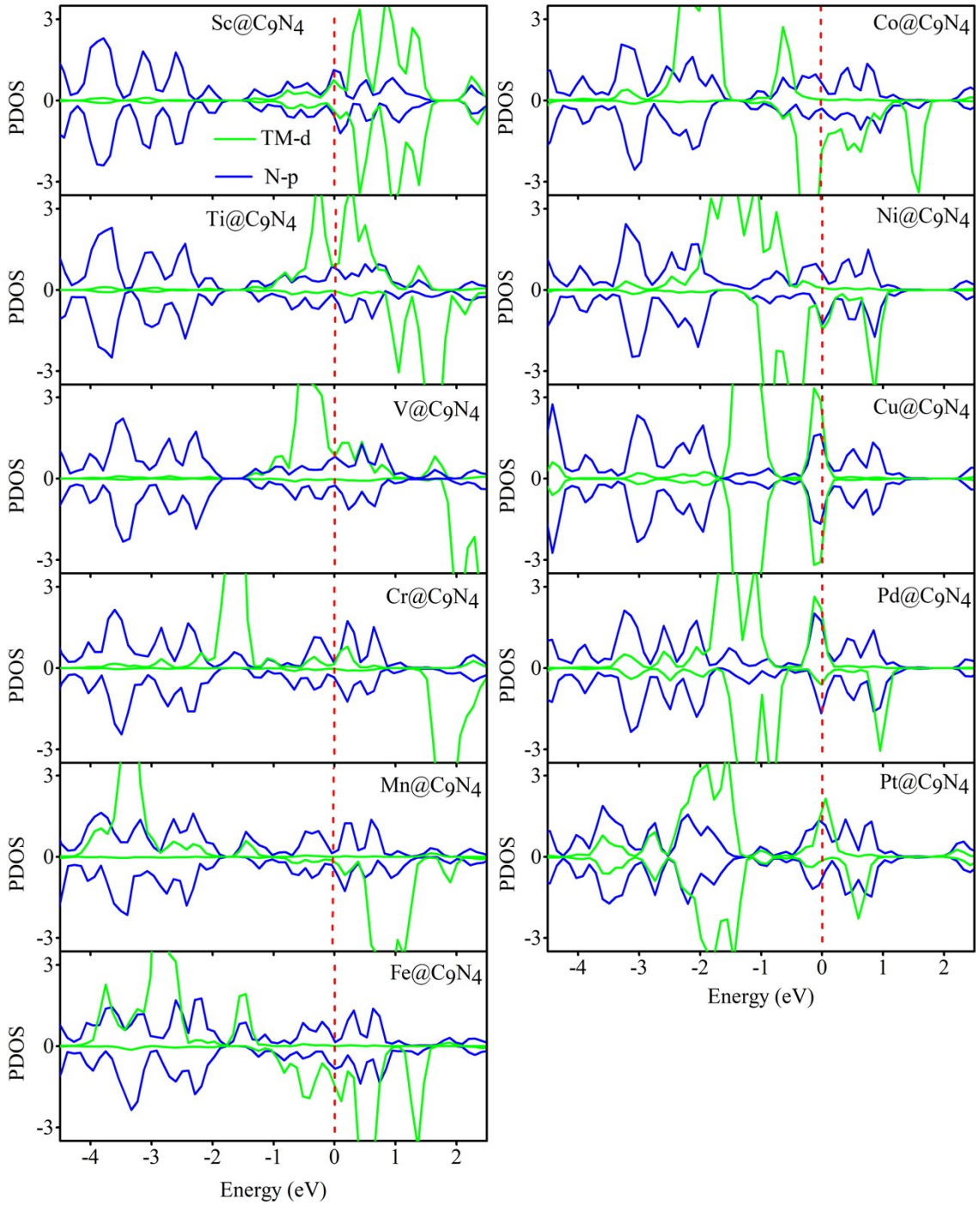


Fig. S1 Calculated PDOS illustrating the p orbital of N and the d orbital of TM@C_9N_4. The Fermi level is set at zero and highlighted by red dash line.

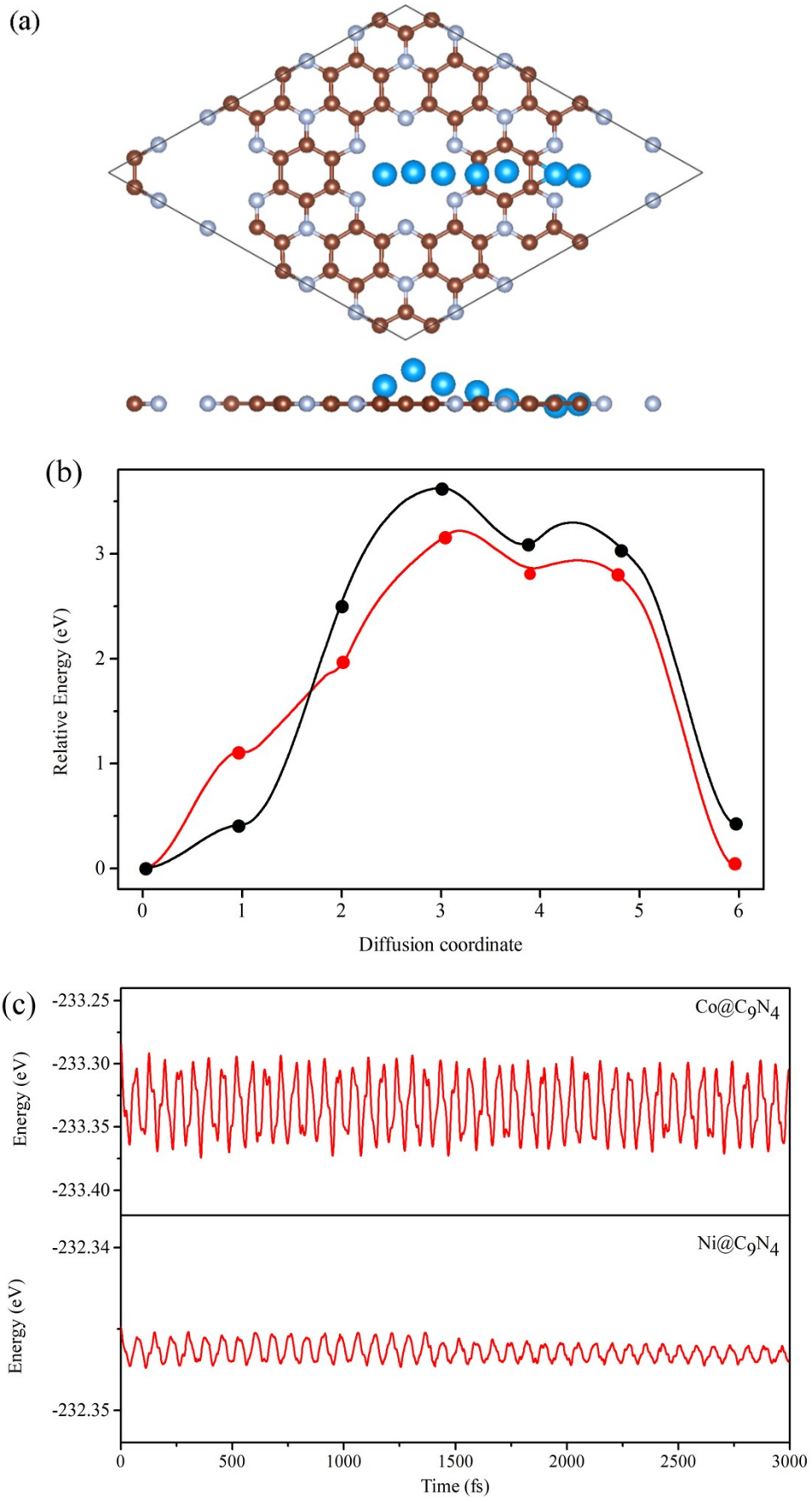


Fig. S2 (a) The view of migration pathway and (b) corresponding minimum diffusion barriers for Co (black) and Ni (red) shifting between two nearby hexagonal holes on C_9N_4 sheet. (c) The total energy as a function of time for Co@C_9N_4 and Ni@C_9N_4 at 300 K.

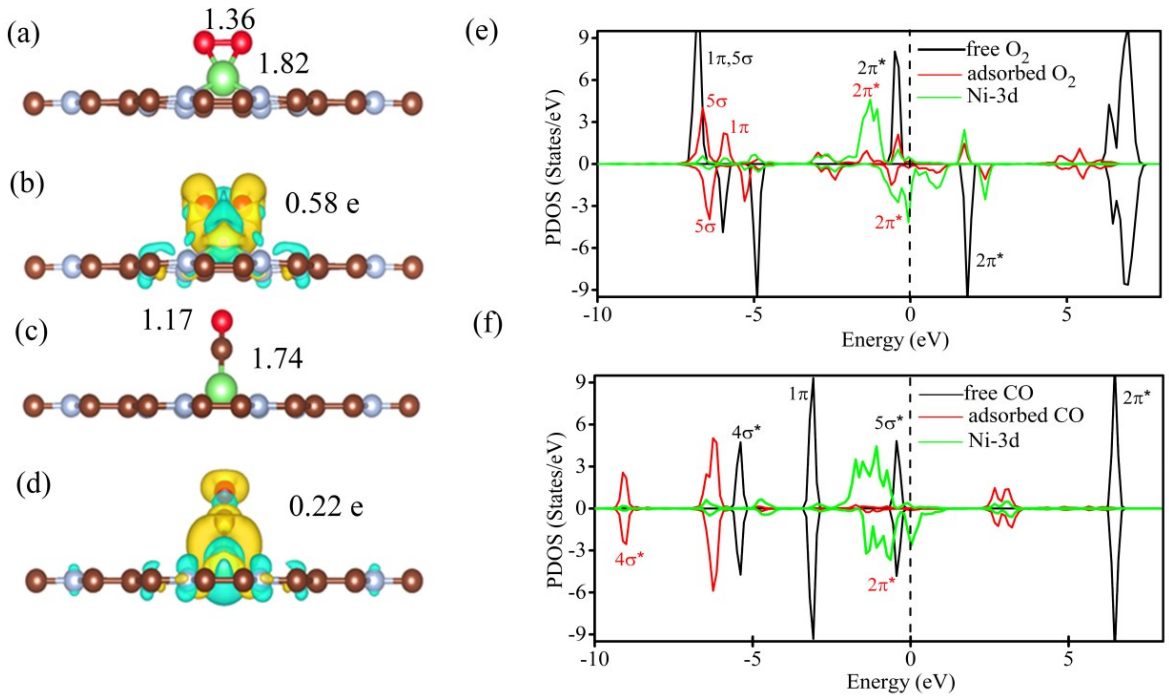
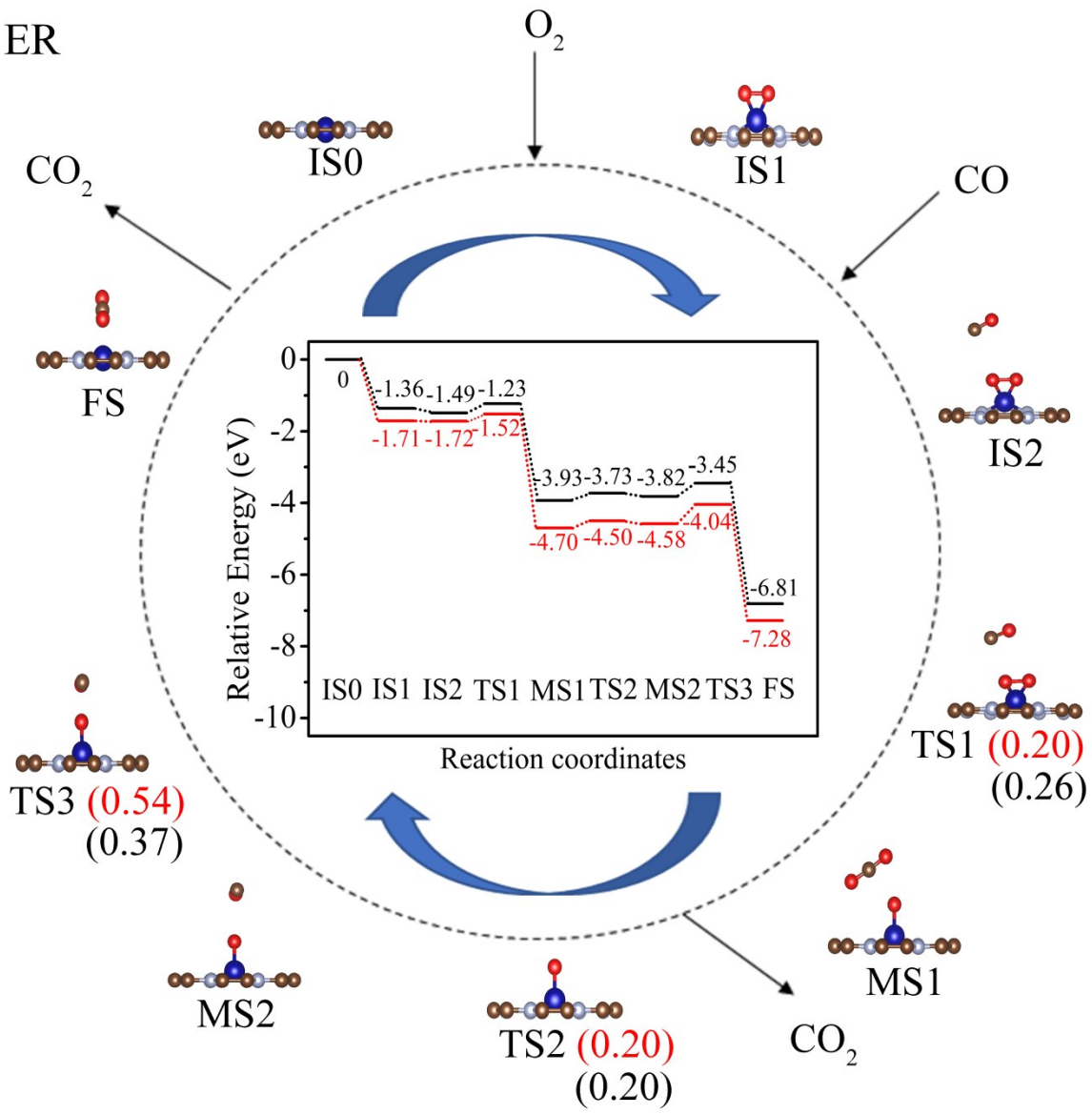


Fig. S3 The side views of optimized structures, corresponding charge density difference, spin-polarized PDOS for O_2 (a) (b) (e) and CO (c) (d) (f) adsorption on Ni@C_9N_4, respectively. The yellow and blue isosurfaces represent the electron accumulation and depletion, and the Fermi level is set zero.


ER
O2
IS0
IS1
CO2
CO
FS
IS2
0
-2
-4
-6
-8
-10
Relative Energy (eV)
0
-1.36
-1.49
-1.23
-1.71
-1.72
-1.52
-3.93
-3.73
-3.82
-3.45
-4.70
-4.50
-4.58
-4.04
-6.81
-7.28
IS0 IS1 IS2 TS1 MS1 TS2 MS2 TS3 FS
Reaction coordinates
TS1 (0.20)
(0.26)
TS3 (0.54)
(0.37)
MS1
MS2
TS2 (0.20)
(0.20)
CO2

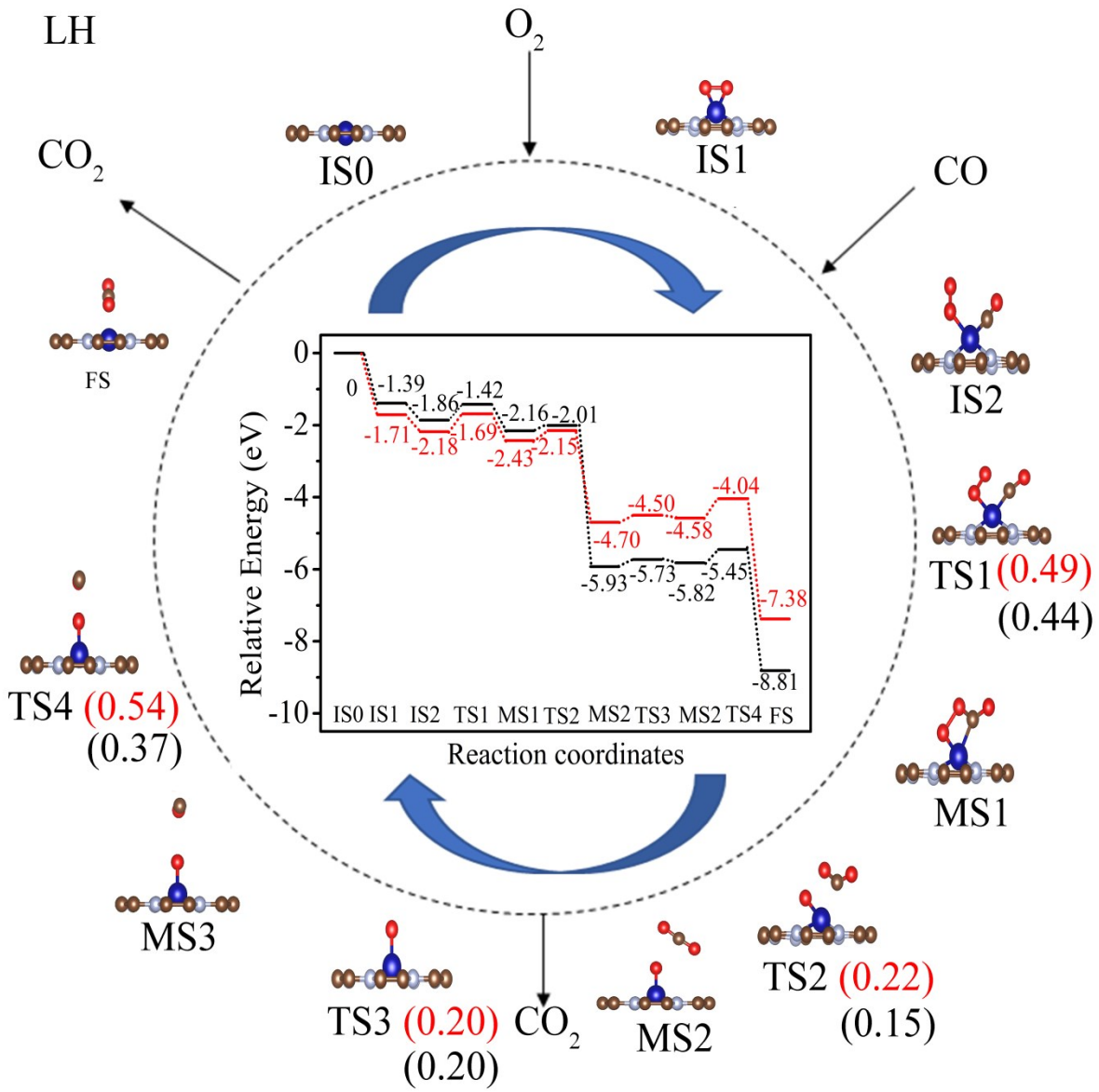
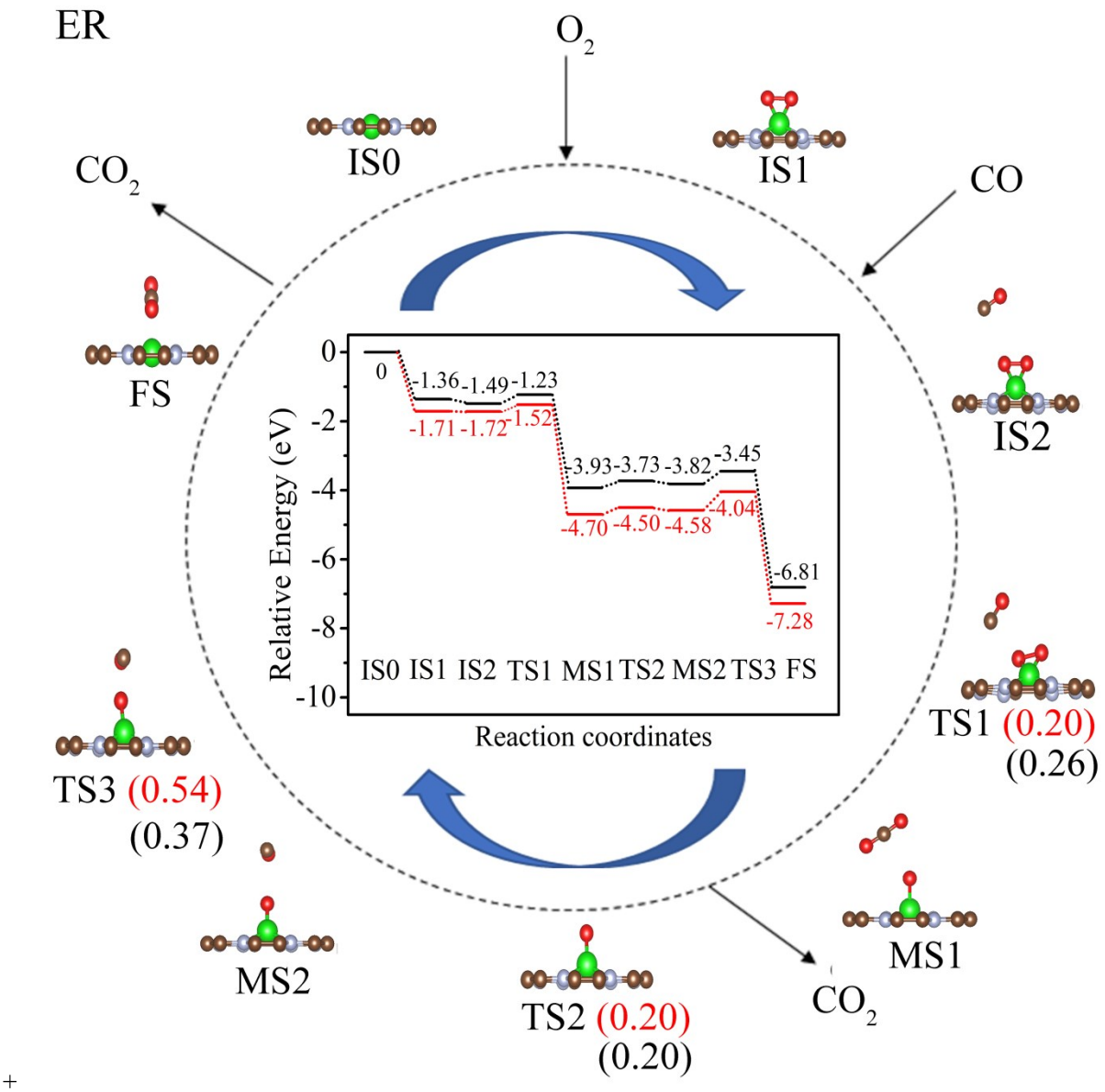
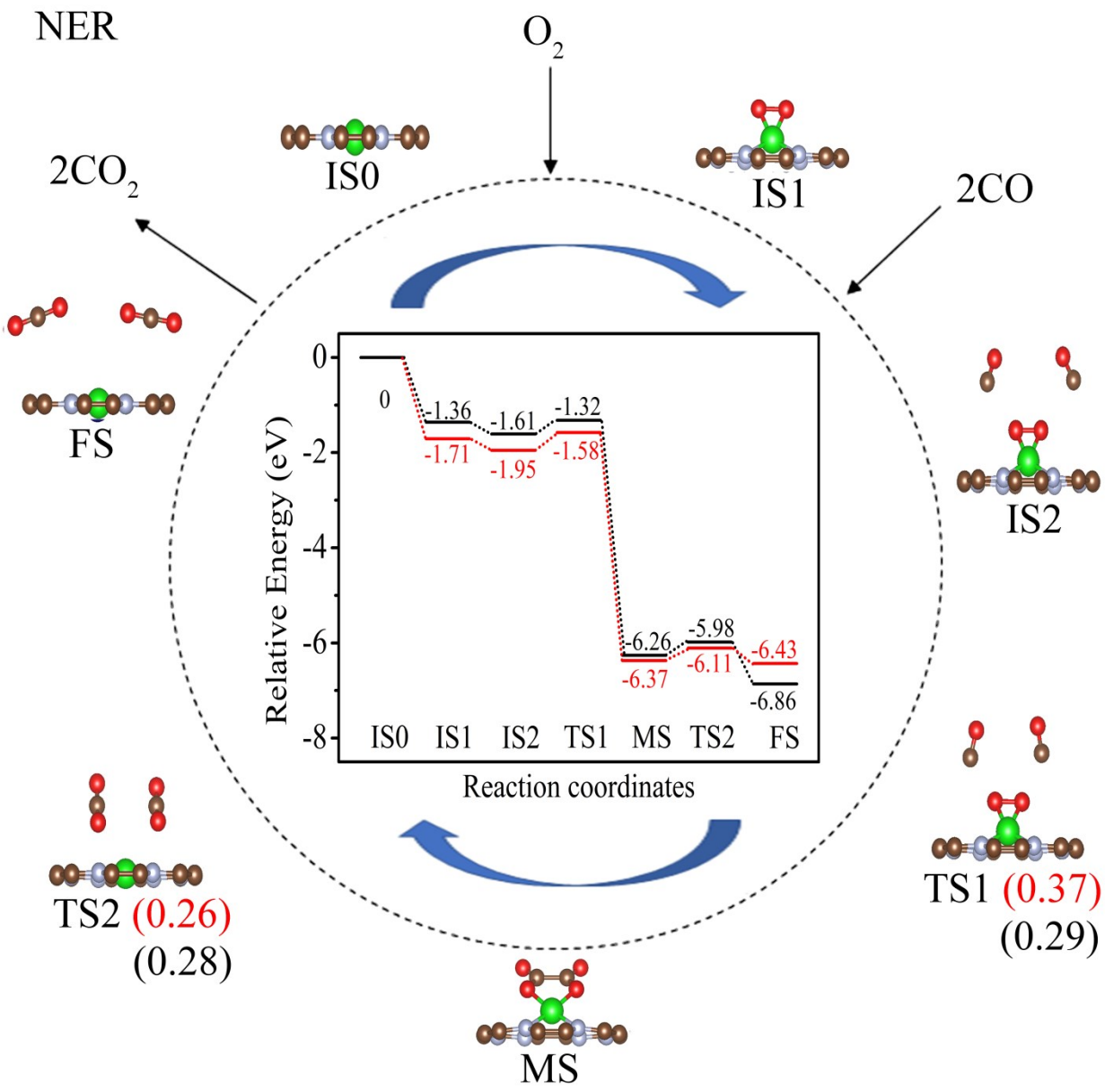
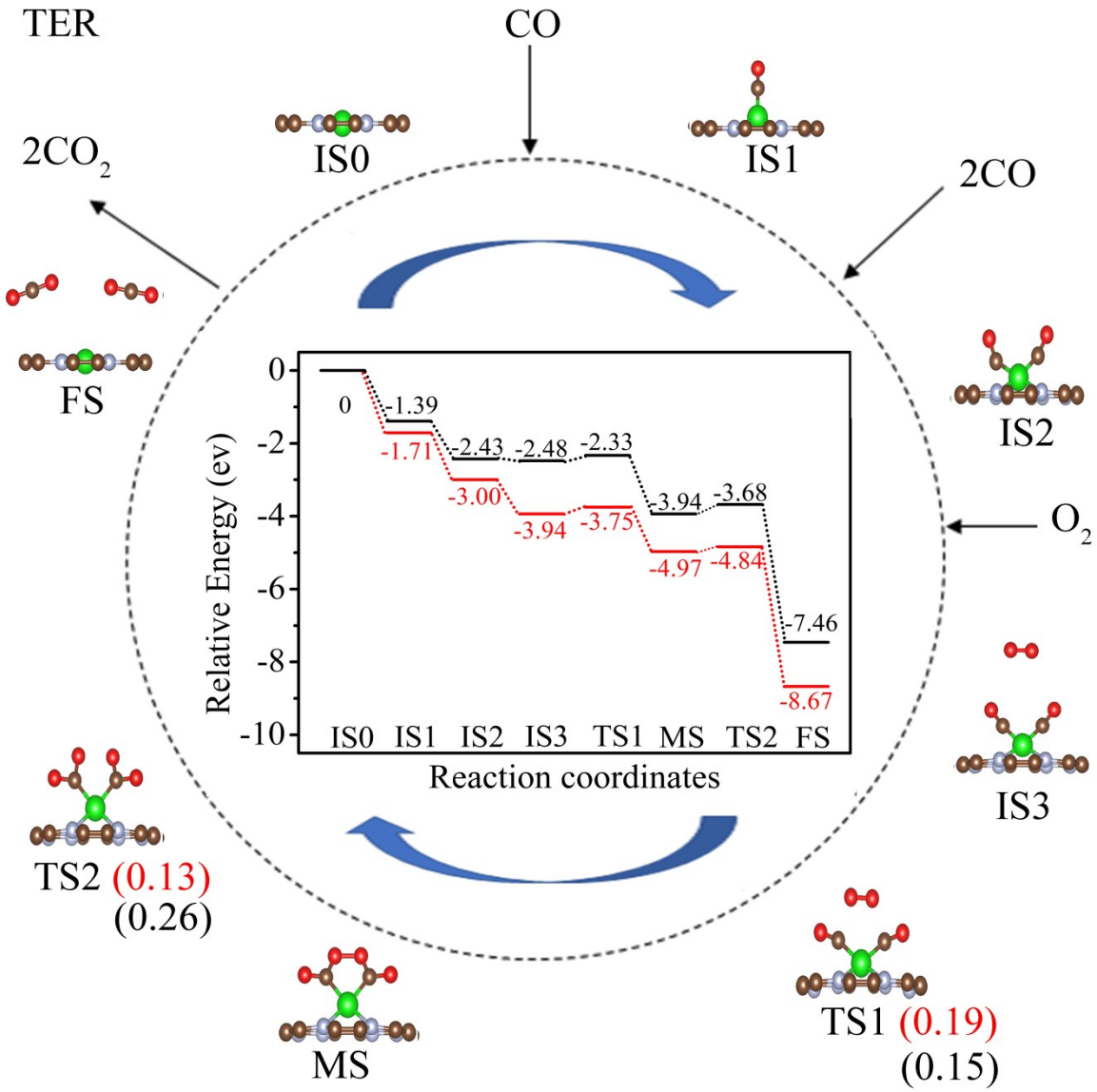


Fig. S4 The structural diagram of CO oxidation on catalyst Ni@C_9N_4 along the minimum energy path via ER and LH mechanism, respectively.


ER
O2
IS0
IS1
CO2
CO
FS
IS2
0
-2
-4
-6
-8
-10
Relative Energy (eV)
0
-1.36
-1.49
-1.23
-1.71
-1.72
-1.52
-3.93
-3.73
-3.82
-3.45
-4.70
-4.50
-4.58
-4.04
-6.81
-7.28
IS0 IS1 IS2 TS1 MS1 TS2 MS2 TS3 FS
Reaction coordinates
TS1 (0.20)
(0.26)
TS3 (0.54)
(0.37)
MS2
MS1
TS2 (0.20)
(0.20)
CO2


NER
O_2
$2CO_2$
IS0
IS1
2CO
FS
IS2
Relative Energy (eV)
0
-2
-4
-6
-8
0
-1.36
-1.61
-1.32
-1.71
-1.95
-1.58
-6.26
-5.98
-6.43
-6.37
-6.11
-6.86
IS0 IS1 IS2 TS1 MS TS2 FS
Reaction coordinates
TS2 (0.26)
(0.28)
TS1 (0.37)
(0.29)
MS


TER
CO
2CO2
IS0
IS1
2CO
FS
IS2
O2
Relative Energy (ev)
0
-2
-4
-6
-8
-10
0
-1.39
-1.71
-2.43
-2.48
-2.33
-3.00
-3.94
-3.75
-3.94
-3.68
-4.97
-4.84
-7.46
-8.67
IS0 IS1 IS2 IS3 TS1 MS TS2 FS
Reaction coordinates
IS3
TS2 (0.13)
(0.26)
MS
TS1 (0.19)
(0.15)

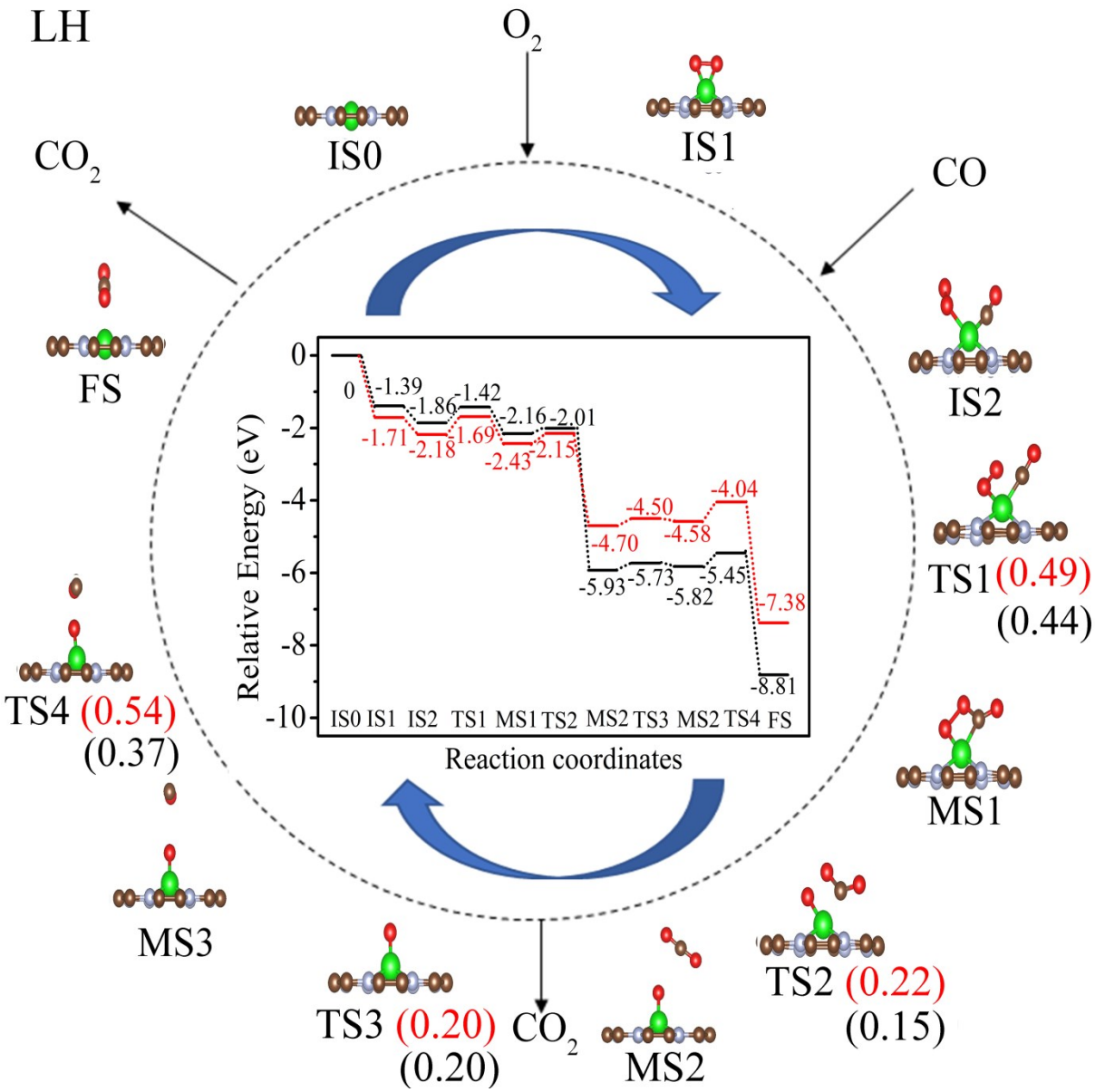


Fig.S5 The structural diagram of CO oxidation on catalyst Ni@C_9N_4 along the minimum energy path via ER, NER, TER and LH mechanisms, respectively.