Supplementary Information

Revisiting the Bonding Nature of Pyramidane: Analogue of the CO Molecule

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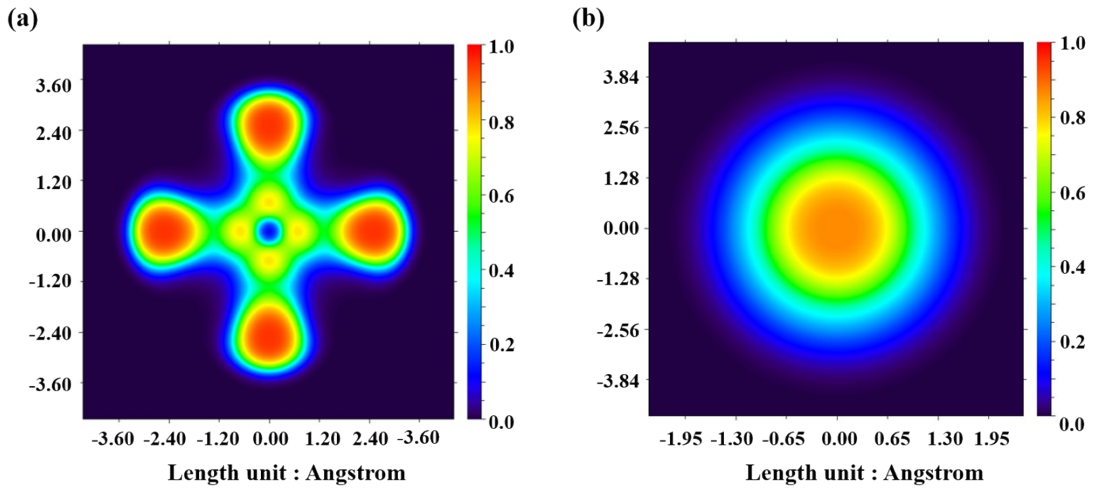

Fig. S1 ELF maps of the (a) $C(C_4H_4)$ and (b) CO monolayers on the x-y planes.

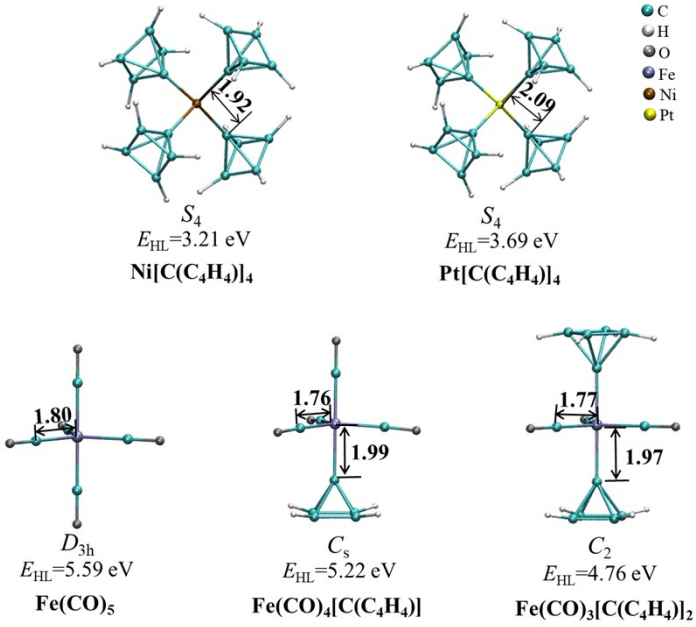

Fig. S2 The optimized structure and the energy (E_{HL}) gap between the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) for $Ni[C(C_4H_4)]_4$, $Pt[C(C_4H_4)]_4$, $Fe(CO)_5$, $Fe(CO)_4[C(C_4H_4)]$, and $Fe(CO)_4[C(C_4H_4)]_2$.

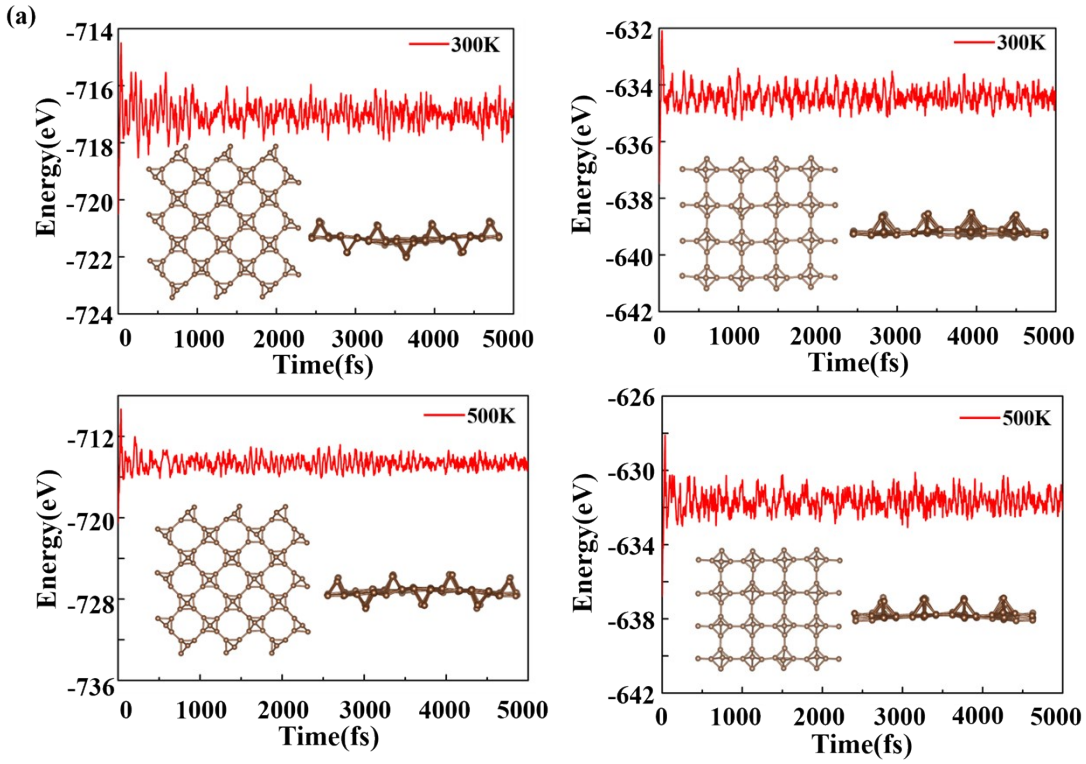

Fig. S3 AIMD simulations at 300 and 500 K for the (a) **C_5-A** and (b) **C_5-B** monolayers.

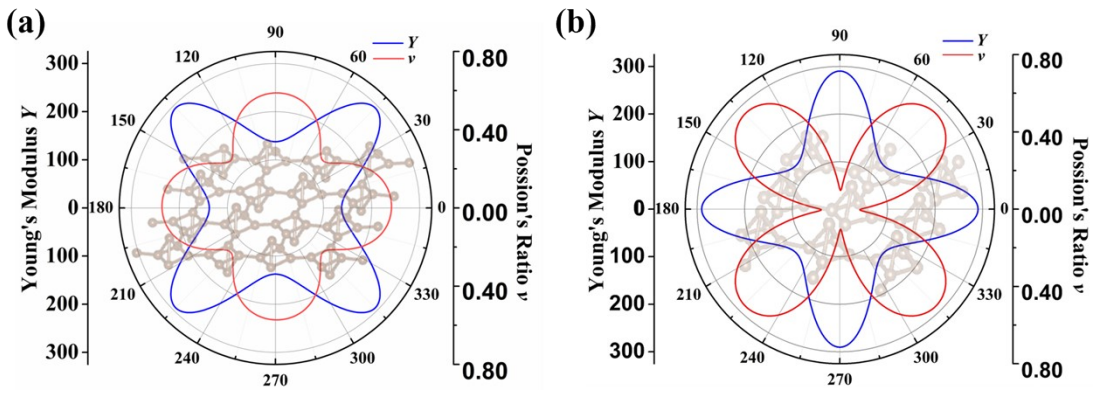

Fig. S4 Young's modulus and Poisson's ratio for the (a) **C_5-A** and (b) **C_5-B** monolayers.

Table S1 Binding energy (E_b) and HOMO-LUMO gap (E_{H-L}) of C_4H_4-*M* (*M* = C, Si, Ge, Sn).

C_4H_4-M	E_b (KJ/mol)	$E_{(H-L)}$ (eV)
C_4H_4-C	-772.75	8.14
C_4H_4-Si	-664.05	7.03
C_4H_4-Ge	-602.44	6.67
C_4H_4-Sn	-528.09	5.47

Table S2 Optimized Cartesian coordinates (in angstrom) of the unit cell for CO, $C(C_4H_4)$, $Pd(CO)_{1\text{-}4}$, $Pd[C(C_4H_4)]_{1\text{-}4}$, **$C_5$-A**, and **$C_5$-B**.

CO

C	0.00000000	0.00000000	-0.64457100
O	0.00000000	0.00000000	0.48342900

$C(C_4H_4)$

C	0.00000000	1.01959600	-0.24261000
C	1.01959600	0.00000000	-0.24261000
C	0.00000000	-1.01959600	-0.24261000
C	-1.01959600	0.00000000	-0.24261000
H	0.00000000	2.08507900	-0.08702700
H	2.08507900	0.00000000	-0.08702700
H	0.00000000	-2.08507900	-0.08702700
H	-2.08507900	0.00000000	-0.08702700
C	0.00000000	0.00000000	1.02845600

Pd(CO)

C	0.00000000	0.00000000	-1.25352500
Pd	0.00000000	0.00000000	0.57951700
O	0.00000000	0.00000000	-2.39207600

$Pd(CO)_2$

Pd	0.00000000	0.00000000	0.00000000
C	0.00000000	0.00000000	1.92975400
O	0.00000000	0.00000000	3.05915000
C	0.00000000	0.00000000	-1.92975400
O	0.00000000	0.00000000	-3.05915000

$Pd(CO)_3$

Pd	0.00000000	0.00000000	0.00000000
C	0.00000000	1.96716800	0.00000000
O	0.00000000	3.09912000	0.00000000
C	1.70361800	-0.98358400	0.00000000
O	2.68391700	-1.54956000	0.00000000
C	-1.70361800	-0.98358400	0.00000000
O	-2.68391700	-1.54956000	0.00000000

$Pd(CO)_4$

Pd	-0.00001800	0.00000700	-0.00001500
C	-1.87244800	-0.62615200	-0.34423800
O	-2.92869200	-0.97935900	-0.53840500
C	1.23570900	-0.81380000	-1.35177900

O	1.93280900	-1.27288300	-2.11427600
C	0.07366400	1.99754900	-0.14478200
O	0.11526600	3.12436300	-0.22641600
C	0.56307200	-0.55760100	1.84079500
O	0.88072500	-0.87215600	2.87918600

$Pd[C(C_4H_4)]$

C	0.00000000	1.02312800	-1.93447800
C	-1.02312800	0.00000000	-1.93447800
C	1.02312800	0.00000000	-1.93447800
C	0.00000000	-1.02312800	-1.93447800
H	0.00000000	2.08624700	-1.76824000
H	-2.08624700	0.00000000	-1.76824000
H	0.00000000	-2.08624700	-1.76824000
H	2.08624700	0.00000000	-1.76824000
C	0.00000000	0.00000000	-0.68478200
Pd	0.00000000	0.00000000	1.25237200

$Pd[C(C_4H_4)]_2$

C	0.00000000	1.02280300	3.24484200
C	-1.02280300	0.00000000	3.24484200
C	1.02280300	0.00000000	3.24484200
C	0.00000000	-1.02280300	3.24484200
H	0.00000000	2.08536600	3.07468000
H	-2.08536600	0.00000000	3.07468000
H	0.00000000	-2.08536600	3.07468000
H	2.08536600	0.00000000	3.07468000
C	0.00000000	0.00000000	1.99581900
C	0.72323100	0.72323100	-3.24484200
C	0.72323100	-0.72323100	-3.24484200
C	-0.72323100	0.72323100	-3.24484200
C	-0.72323100	-0.72323100	-3.24484200
H	1.47457700	1.47457700	-3.07468000
H	1.47457700	-1.47457700	-3.07468000
H	-1.47457700	-1.47457700	-3.07468000
H	-1.47457700	1.47457700	-3.07468000
C	0.00000000	0.00000000	-1.99581900
Pd	0.00000000	0.00000000	0.00000000

$Pd[C(C_4H_4)]_3$

C	-0.72081600	0.72306200	3.34094400
C	-0.72081600	-0.72306200	3.34094400
C	0.72081600	0.72306200	3.34094400
C	0.72081600	-0.72306200	3.34094400

H	-1.47443400	1.47287900	3.17380300
H	-1.47443400	-1.47287900	3.17380300
H	1.47443400	-1.47287900	3.17380300
H	1.47443400	1.47287900	3.17380300
C	0.00000000	0.00000000	2.06355700
C	-1.01932700	-2.92903700	-1.64330400
C	0.00000000	-2.45963000	-2.55214200
C	0.00000000	-3.44021400	-0.75701500
C	1.01932700	-2.92903700	-1.64330400
H	-2.08465700	-2.79427900	-1.56987700
H	0.00000000	-1.79196100	-3.39572900
H	2.08465700	-2.79427900	-1.56987700
H	0.00000000	-3.78998000	0.26045600
C	0.00000000	-1.81850000	-1.03491500
C	0.00000000	3.44021400	-0.75701500
C	1.01932700	2.92903700	-1.64330400
C	-1.01932700	2.92903700	-1.64330400
C	0.00000000	2.45963000	-2.55214200
H	0.00000000	3.78998000	0.26045600
H	2.08465700	2.79427900	-1.56987700
H	0.00000000	1.79196100	-3.39572900
H	-2.08465700	2.79427900	-1.56987700
C	0.00000000	1.81850000	-1.03491500
Pd	0.00000000	0.00000000	-0.02480500

$Pd[C(C_4H_4)]_4$

C	1.62074400	-2.13811200	2.35371500
C	1.37961800	-3.05635200	1.26762400
C	0.24092500	-2.25490600	2.75931600
C	0.00000000	-3.17524500	1.67390200
H	2.42438900	-1.46885200	2.60751200
H	1.93234400	-3.34626500	0.39098600
H	-0.88476200	-3.58909500	1.22218300
H	-0.39306700	-1.71187800	3.43852300
C	0.49966900	-1.66956800	1.24861900
C	-2.62050900	-1.73415100	-1.74512600
C	-2.06885500	-0.90733200	-2.79173900
C	-3.32186000	-0.59109500	-1.21236500
C	-2.77160500	0.23498400	-2.25923000
H	-2.41115800	-2.72543500	-1.38239600
H	-1.28678300	-1.03889800	-3.51918600
H	-2.71756100	1.29618200	-2.42976500
H	-3.84217200	-0.39133400	-0.29181800
C	-1.68414300	-0.47735600	-1.25576500

C	0.00000000	3.17524500	1.67390200
C	-0.24092500	2.25490600	2.75931600
C	-1.37961800	3.05635200	1.26762400
C	-1.62074400	2.13811200	2.35371500
H	0.88476200	3.58909500	1.22218300
H	0.39306700	1.71187800	3.43852300
H	-2.42438900	1.46885200	2.60751200
H	-1.93234400	3.34626500	0.39098600
C	-0.49966900	1.66956800	1.24861900
C	2.77160500	-0.23498400	-2.25923000
C	3.32186000	0.59109500	-1.21236500
C	2.06885500	0.90733200	-2.79173900
C	2.62050900	1.73415100	-1.74512600
H	2.71756100	-1.29618200	-2.42976500
H	3.84217200	0.39133400	-0.29181800
H	2.41115800	2.72543500	-1.38239600
H	1.28678300	1.03889800	-3.51918600
C	1.68414300	0.47735600	-1.25576500
Pd	0.00000000	0.00000000	-0.01172800

C_5-A

C	0.722046675	1.752976656	1.406448260
C	2.788263312	1.752976656	1.406448260
C	1.755154967	0.721150600	1.406448260
C	1.755154967	2.784802608	1.406448260
C	1.755154967	1.752976656	2.752306759

C_5-B

C	1.740515176	0.731621869	7.436013669
C	3.197779235	4.226578035	7.436013669
C	1.740514881	4.226578331	7.436013669
C	3.197779529	0.731621647	7.436013669
C	0.728632103	3.210721932	7.563985884
C	4.209662381	1.747478046	7.563985884
C	0.728632324	1.747478342	7.563985884
C	4.209662087	3.210721636	7.563985884
C	2.469147205	0.000000000	8.766694665
C	0.000000000	2.479099989	6.233305782