

Supporting information

The Effect of Defects on the Catalytic Activity of Single Au Atom Supported on Carbon Nanotube and Reaction Mechanism for CO Oxidation

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Table S1. The structure information and atom number in the cell of the investigated carbon nanotubes.

System	Number of atoms			Diameter(Å)	C-C bond length (Å)	Angles degree
	m-SWCNT	di-SWCNT	SW-SWCNT/pristine			
(8,0) SWCNT	95	94	96	6.41	1.42 and 1.44	117.2° and 119.4°
(5,5) SWCNT	99	98	100	6.87	1.43	118.9°

Table S2. The comparison of binding energy of Au in different exchange correlation functions

Exchange correlation functionals	Au/SWCNT	Au/m-SWCNT	Au/di-SWCNT	Au/SW-SWCNT
	E_b	E_b	E_b	E_b
RPBE	-0.38	-2.74	-0.176	-0.74
VDW_DF	-0.63	-2.77	-0.24	-1.273
VDW_DF2	-0.66	-2.53	-0.34	-1.274

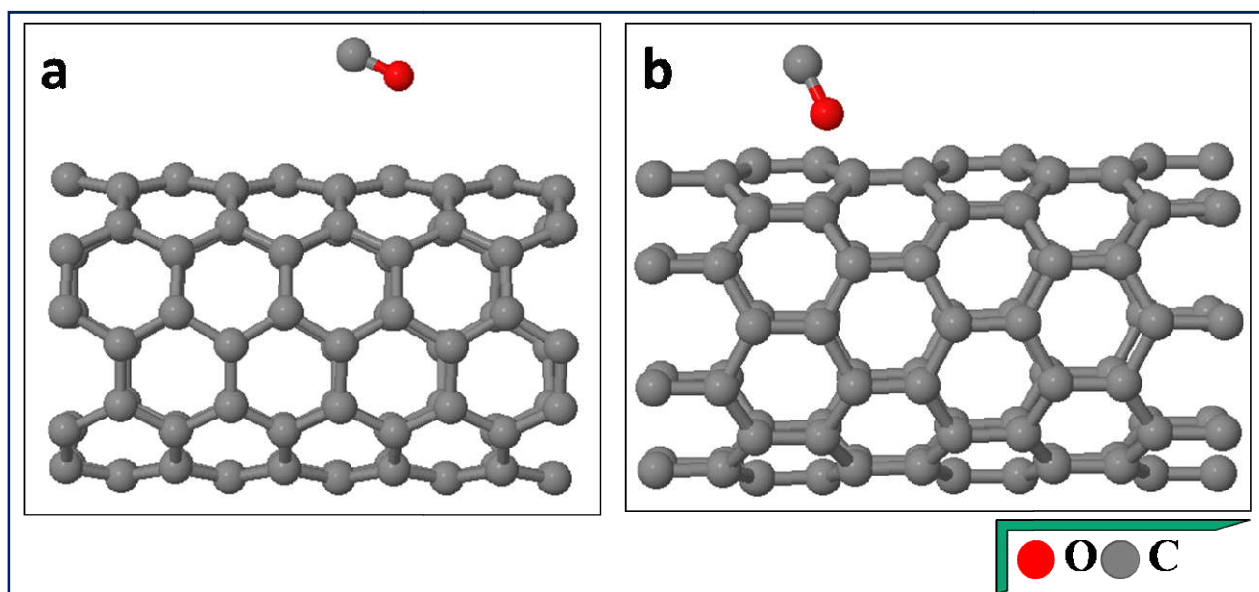


Fig. S1 Optimized geometry of CO on pristine armchair (5,5) and zigzag (5,5) SWCNT.

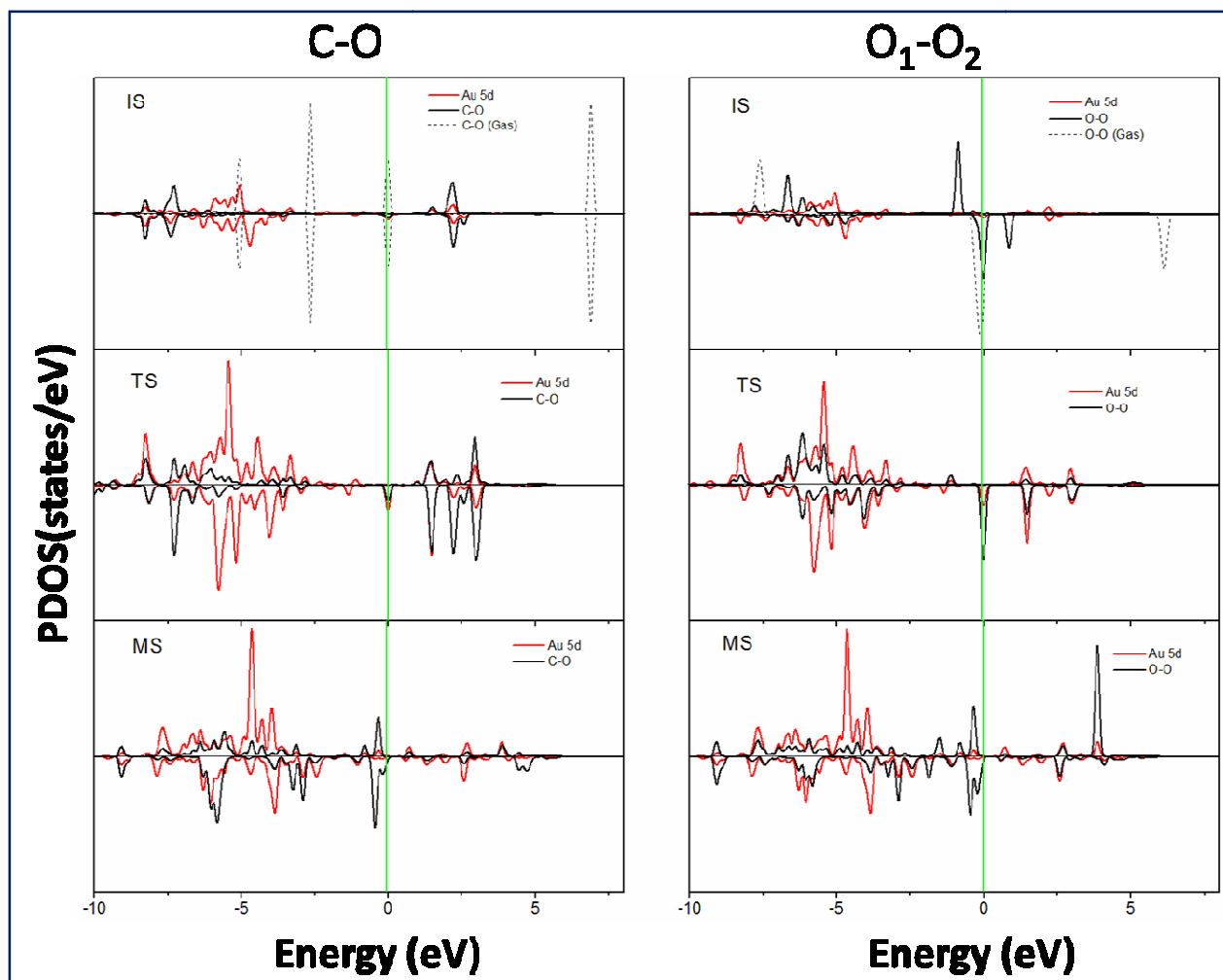


Fig. S2 Spin-polarized partial density of states (PDOS) projected onto C-O (left panel) and O₁-O₂ (right panel) on the Au/m-SWCNT (Fig. 6b), together with the d-projected PDOS of the Au atom in the IS, TS, and MS. Black dashed curve, gas-phase CO or O₂; black solid curve, C-O or O₁-O₂ on Au/m-SWCNT; red curve, d-projected PDOS of the Au atom. The Fermi level is set to zero. The spin density is defined as the difference between spin-up and spin-down electron densities, $\rho_{\uparrow} - \rho_{\downarrow}$.

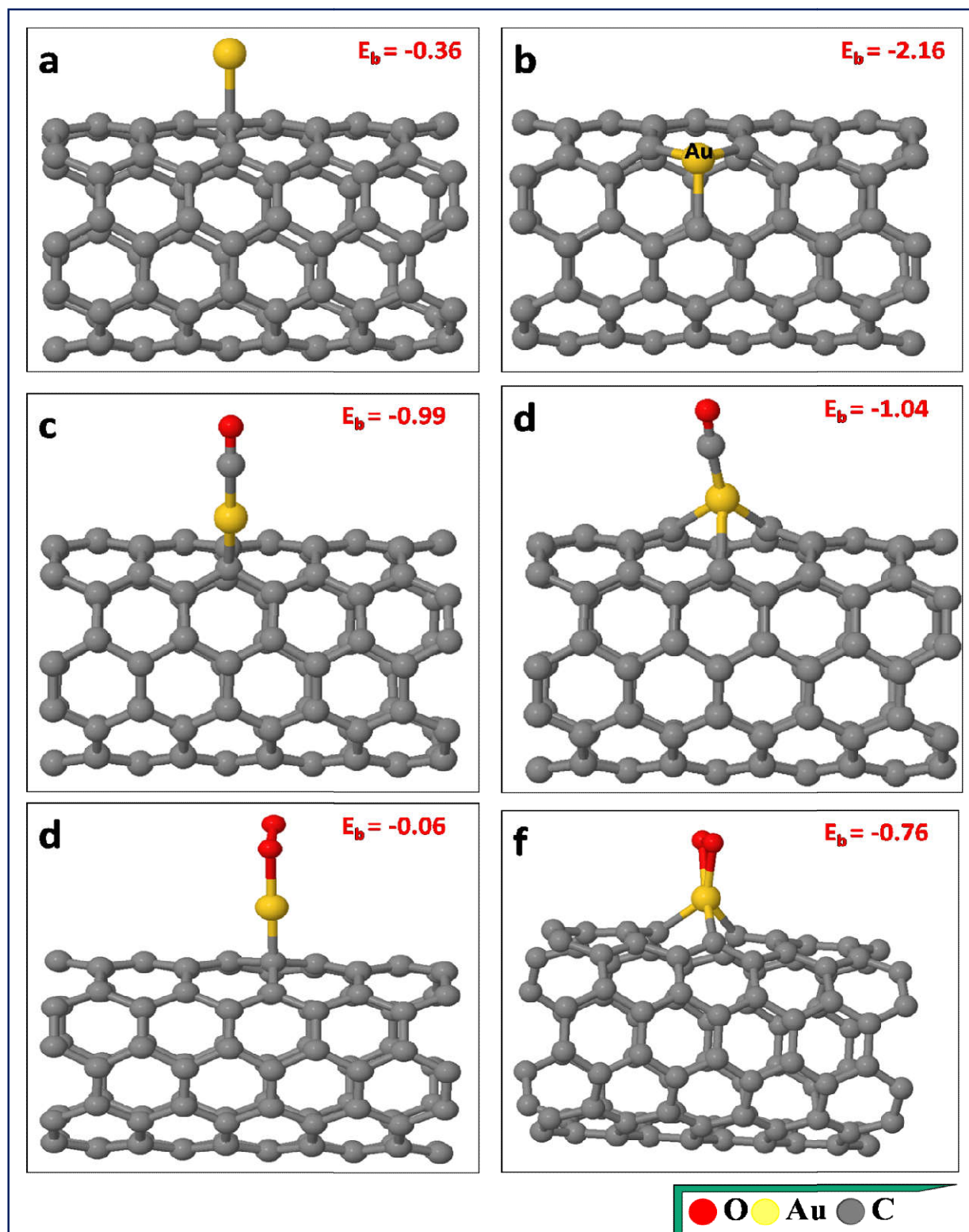


Fig. S3 Au, CO and O₂ adsorption on; (a) Au/SWCNT (5,5) (a,c,d); (b) Au/m-SWCNT (5,5) (b,e,f); E_b represent binding energy in eV. Only a part of SWCNTs are shown.

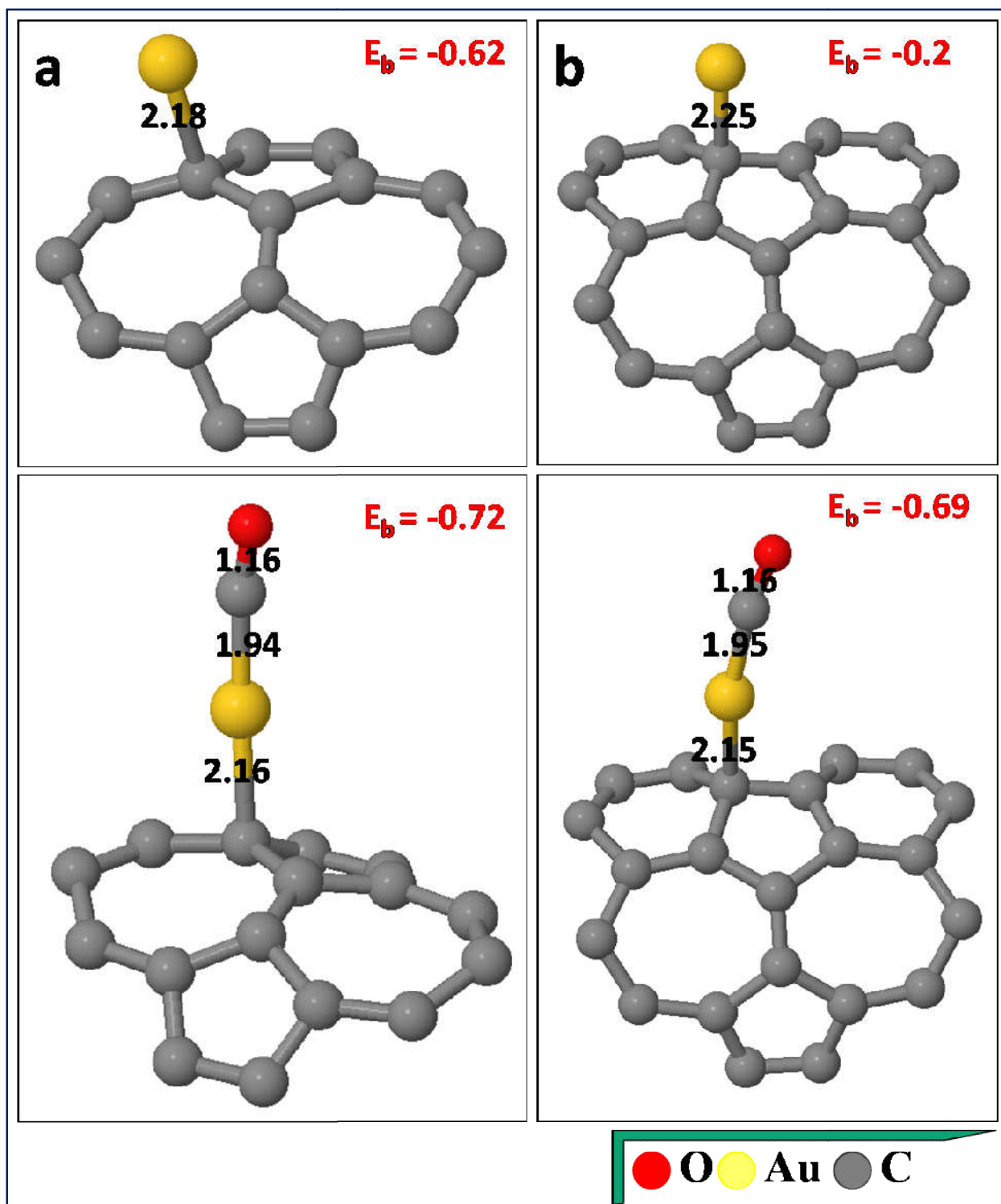


Fig. S4 Au and CO adsorption on Au/SW-SWCNT (8,0) with different binding sites; E_b represent binding energy in eV. The interatomic distances are given in Å. Only a part of SWCNTs are shown.

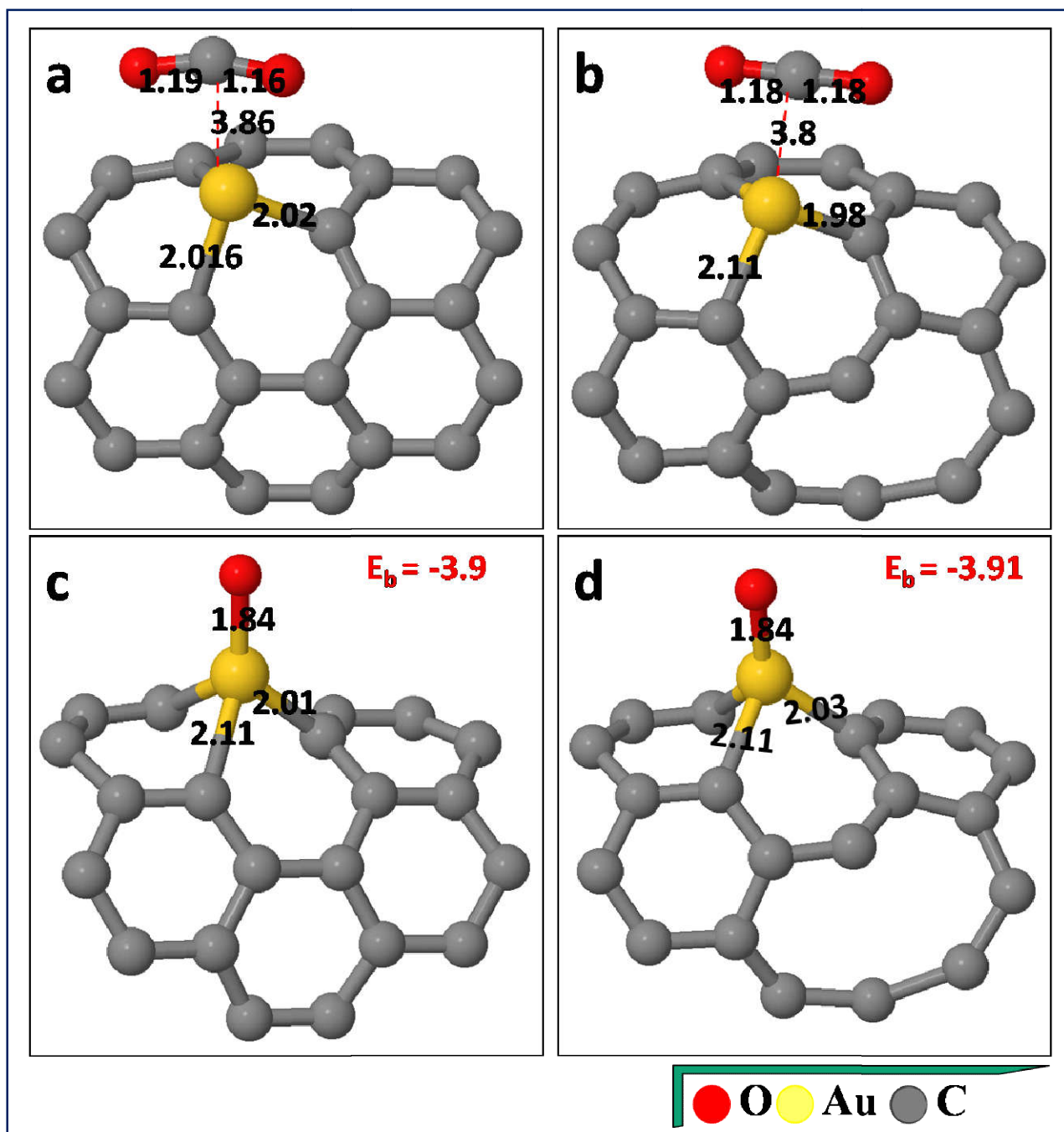


Fig. S5 CO₂ and O adsorption on; (a) Au/m-SWCNT (8,0) (a, c); (b) Au/di-SWCNT (8,0) (b, d); E_b represent binding energy in eV. The interatomic distances are given in Å. Only a part of SWCNTs are shown.

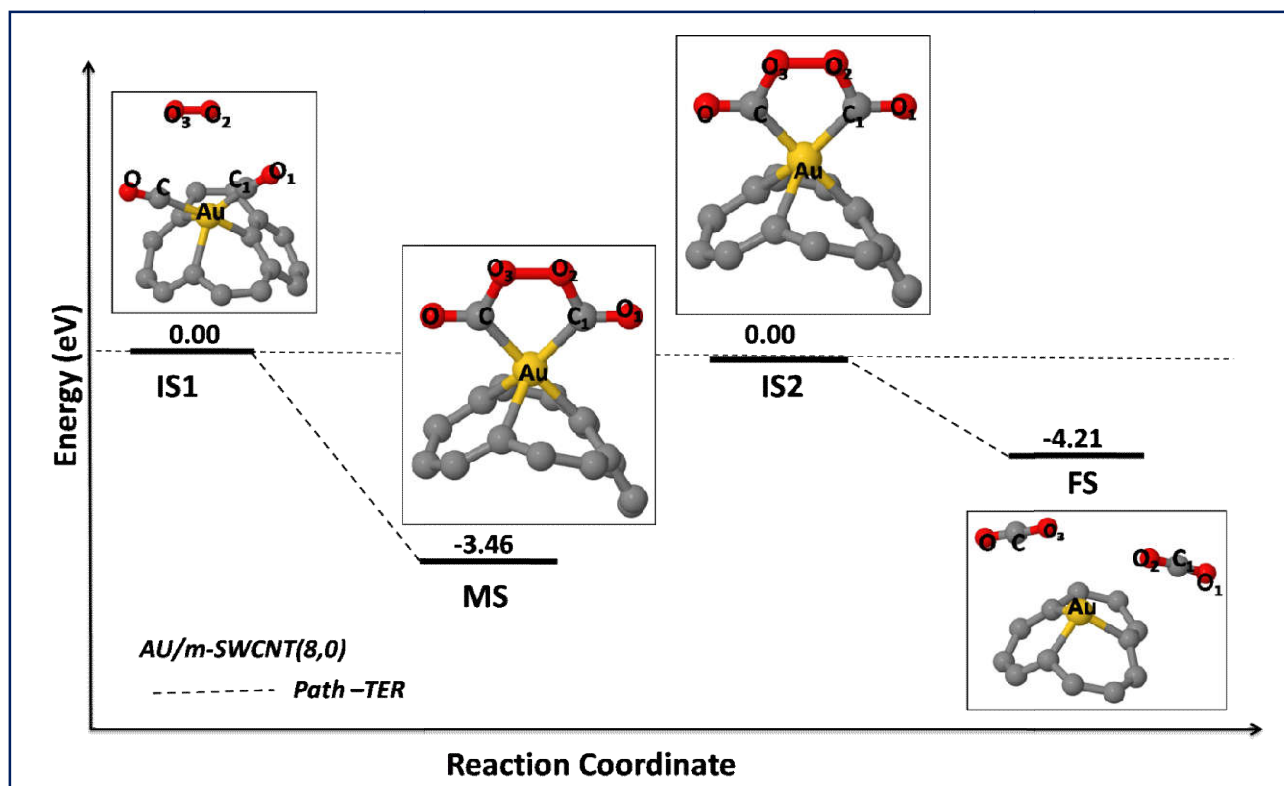


Fig. S6. Reaction pathways of CO on Au supported m-SWCNT. The Schematic energy profile corresponding to local configurations along the minimum-energy pathway via the TER $2\text{CO} + \text{O}_2 \rightarrow \text{OOCOCO} \rightarrow 2\text{CO}_2$ route. All energies are given with respect to the reference energy, i.e., the sum of energies of the Au/m-SWCNT and individual CO and O₂ molecules.

Table S3. The magnetic moment of Au (M Au) and C (M C) at defected sites of SWCNT are summarized.

Magnetic moment	Au/SWCNT	Au/m-SWCNT	Au/di-SWCNT	Au/SW-SWCNT
M Au (μ_B)	0.463	0.095	0.103	0.291
M C1 (μ_B)	0.004	0.061	0.242	0.044
M C2 (μ_B)	0.052	0.202	0.002	0.027
M C3 (μ_B)	0.053	0.061	0.028	0.016
M C4 (μ_B)	0.060	-	0.182	0.192

Table S4. Structural parameters for the intermediate states (IS, TS, MS, and FS) along the MEP for the CO oxidation on the Au/m-SWCNT: (a) $\text{CO} + \text{O}_2 \rightarrow \text{OOCO} \rightarrow \text{CO}_2 + \text{O}$; (b) $\text{CO} + \text{O} \rightarrow \text{CO}_2$

(a)	IS	TS	MS	FS
$d_{(\text{O1-O2})}$	1.29	1.32	1.51	3.38
$d_{(\text{C-O})}$	1.16	1.16	1.22	1.18
$d_{(\text{C-Au})}$	2.01	2.09	2.08	4.08
$d_{(\text{O2-Au})}$	2.31	2.18	2.13	1.84
$\delta q [\text{Au}]$	+0.53	+0.58	+0.59	+0.67
$\delta q [\text{C+O}]$	$1.05+(-1.06) = 0.01$	$1.09+(-1.07) = 0.02$	$1.4+(-1.1) = 0.28$	$2.1+(-1.1) = 1.0$
$\delta q [\text{O}_2+\text{O}_1]$	$-0.15+(-0.22) = -0.37$	$-1.1+(-0.7) = -1.8$	$-0.57+(-0.41) = -0.98$	-0.8, -1.06
(b)	IS	TS	FS	
$d_{(\text{O-C})}$	3.13	1.7	1.18	
$d_{(\text{O-Au})}$	1.84	1.92	3.34	
$d_{(\text{C-O1})}$	1.15	1.17	1.18	
$\delta q [\text{Au}]$	+0.67	+0.63	+0.34	
$\delta q [\text{C+O}_1]$	$1.1+(-1.12) = -0.02$	$1.14+(-1.13) = 0.01$	$2.1+(-1.05) = 1.05$	
$\delta q [\text{O}]$	-0.69	-0.68	-1.07	

The units of the bond distance and charge transfer are Å and e respectively. The structure of IS, TS, MS and FS are displayed in Fig. 6b. Charge transfer is calculated using the Bader charge analysis. Negative (positive) δq denotes charge gain (loss).