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

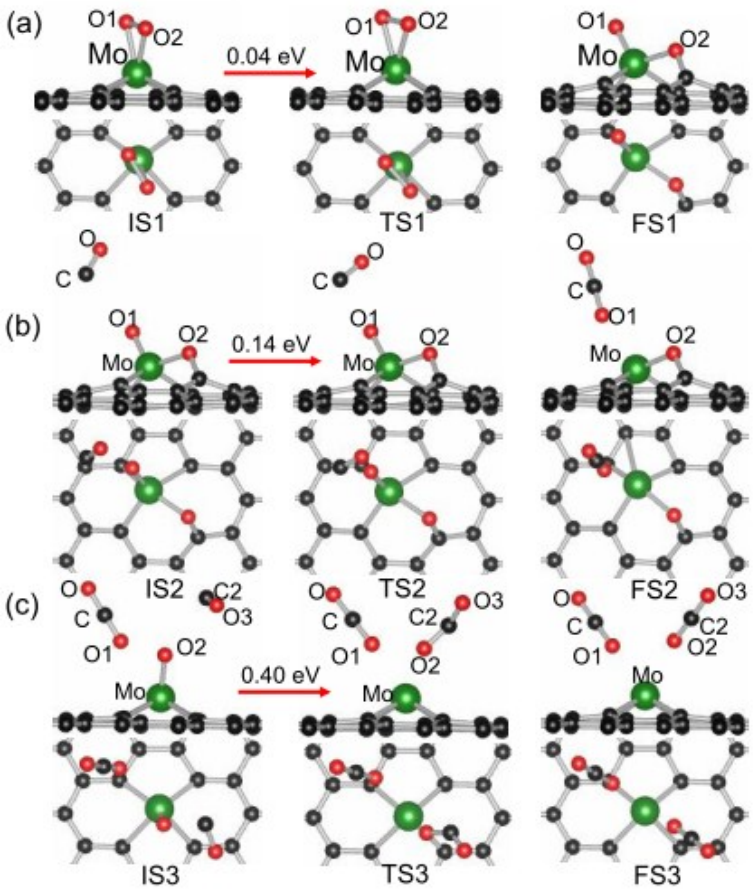

Fig.S1. The minimum energy profiles and the configurations of different states for CO oxidation reaction on Mo-DG sheet, including (a) the dissociative adsorption of O_2 molecule and (b)-(c) $2CO + 2O_{ads}$ reactions by the ER mechanism. Red, green and black balls represent O, Mo and C atoms, respectively.

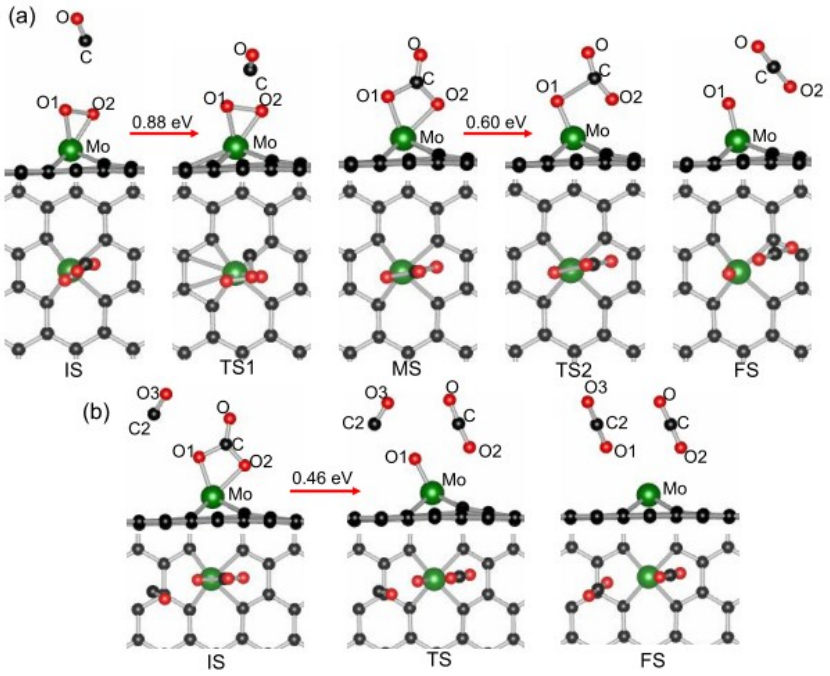

Fig.S2. The minimum energy profiles and the configurations of different states for CO oxidation reaction on Mo-DG sheet, including (a) $CO + O_2 \rightarrow CO_3 \rightarrow CO_2 + O_{ads}$ and (b) $CO_3 + CO \rightarrow 2CO_2$ reactions by the ER mechanism. Red, green and black balls represent O, Mo and C atoms, respectively.

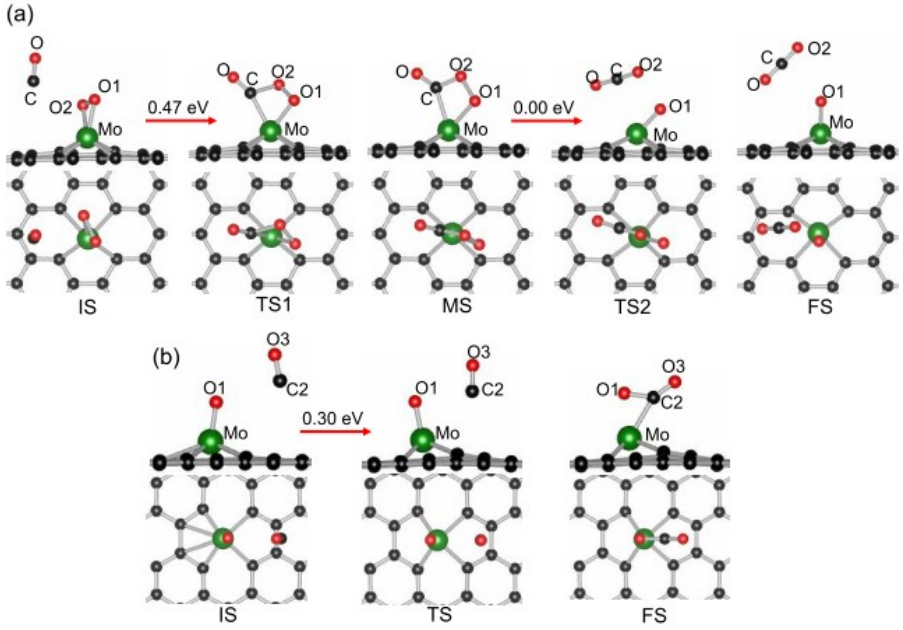

Fig.S3. The minimum energy profiles and the configurations of different states for CO oxidation reaction on Mo-DG sheet, including (a) $CO + O_2 \rightarrow OOCO \rightarrow CO_2 + O_{ads}$ by the LH mechanism, (b) $CO + O_{ads}$ reaction by the ER mechanism. Red, Green and black balls represent O, Mo and C atoms, respectively.

Table S1. Adsorption energies (E_{ads}, eV) of different adsorbates (including the reactants, intermediates and products) on Co-DG sheet are calculated by GGA-PBE and DFT-D2 methods.

adsorbates	CO	O_2	O	CO_2	CO_3	CO, O_2	2OCO
GGA-PBE	1.32	1.61	4.83	0.16	4.56	1.79	0.33
DFT-D2	1.47	1.74	4.91	0.27	4.81	2.09	0.85

Table.S2. Structural parameters for the CO oxidation on the Mo-DG (IS, TS and FS), (a) the dissociated reaction of O_2 ($O_2 \rightarrow O1 + O2$), two CO reacting with O_{ads} including (b) $CO + O1 \rightarrow CO_2$ and (c) $CO + O2 \rightarrow CO_2$, as display in Fig. S1(a)-(c).

$O_2 \rightarrow O1 + O2$	IS1 (Å)	TS1 (Å)	FS1 (Å)
(a)			
d_{O1-O2}	1.44	1.45	3.04
d_{Mo-O1}	1.94	1.95	1.71
d_{Mo-O2}	1.98	1.96	1.94

$O1 + CO \rightarrow CO_2$	IS2 (Å)	TS2 (Å)	FS2 (Å)
(b)			
d_{O1-O2}	3.02	3.03	3.54
d_{O1-Mo}	1.72	1.71	2.48
d_{C-O}	1.14	1.14	1.17
d_{C-O1}	2.89	2.34	1.18

$O2 + CO \rightarrow CO_2$	IS3 (Å)	TS3 (Å)	FS3 (Å)
(c)			
d_{O2-Mo}	1.73	2.06	2.46
d_{O2-C2}	2.69	1.23	1.18
d_{C2-O3}	1.14	1.18	1.17

Table.S3. Structural parameters for the preadsorbed O_2 with CO for the IS, TS, MS and FS along the MEP for CO oxidation on the Mo-DG sheet through ER reactions, which includes (a) $CO + O_2 \rightarrow CO_3 \rightarrow CO_2 + O_{ads}$ and (b) $CO + CO_3 \rightarrow 2CO_2$, the corresponding reaction pathways are display in Fig. S2(a) and (b).

distance (Å)	IS	TS1	MS	TS2	FS
(a)					
d_{C-O}	1.14	1.15	1.21	1.18	1.17
d_{C-O1}	3.47	2.22	1.39	1.95	2.72
d_{C-O2}	3.49	2.12	1.33	1.22	1.18
d_{O1-Mo}	1.94	1.90	1.96	1.80	1.73
d_{O2-Mo}	1.98	1.97	2.11	2.46	3.53
d_{O1-O2}	1.44	1.51	2.18	2.41	3.02

distance (Å)	IS	TS	FS
(b)			
d_{O3-C2}	1.14	1.15	1.17
d_{O1-C}	1.39	2.58	3.19
d_{O1-C2}	2.98	2.50	1.18
d_{O1-Mo}	1.96	1.74	2.99

Table.S4. Structural parameters for the coadsorption of CO and O_2 for the IS, TS, MS and FS along the MEP for CO oxidation on the Mo-DG, (a) LH reaction ($CO + O_2 \rightarrow OOCO \rightarrow CO_2 + O_{ads}$), (b) ER reaction ($CO + O \rightarrow CO_2$), as display in Fig. S3(a)-(b).

distance (Å)	IS	TS1	MS	TS2	FS
(a)					
d_{C-O}	1.14	1.19	1.21	1.18	1.17
d_{C-Mo}	3.92	2.20	2.21	2.99	4.16
d_{C-O1}	3.23	2.30	2.18	2.79	2.91
d_{C-O2}	2.99	1.56	1.38	1.16	1.17
d_{O1-Mo}	1.94	2.00	2.03	1.76	1.72
d_{O2-Mo}	2.00	2.20	2.74	3.13	4.63
d_{O1-O2}	1.44	1.45	1.48	2.24	3.13

distance (Å)	IS	TS	FS
(b)			
d_{O1-Mo}	1.72	1.74	2.03
d_{O1-C2}	2.91	2.61	1.32
d_{C2-O3}	1.14	1.15	1.20