Supporting Information for

Adsorption energy as a promising single-parameter descriptor for single atom catalysis in oxygen evolution reaction

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Note 1

In this study, the oxygen evolution reaction (OER) is considered as four elementary steps. The Gibbs free energy (ΔG) for each step was calculated as follows:

$$(1)\ H_2O\ (l) + * \rightarrow OH* + e^- + H^+$$

$$\begin{aligned}\Delta G_1 &= \mu_{OH*} + \mu_{H^+} + \mu_{e^-} - \mu_{H_2O} - \mu_* \\ &= \Delta E_{OH} + \Delta E_{ZPE(OH)} - T\Delta E_{OH} + G_{solv(OH*)} \\ &= \Delta G_{OH}\end{aligned}$$

$$(2)\ OH* \rightarrow O* + e^- + H^+$$

$$\begin{aligned}\Delta G_1 &= \mu_{O*} + \mu_{H^+} + \mu_{e^-} - \mu_{OH*} \\ &= (\Delta E_O + \Delta E_{ZPE(O)} - T\Delta E_O + G_{solv(O*)}) - (\Delta E_{OH} + \Delta E_{ZPE(OH)} - T\Delta E_{OH} + G_{solv(OH*)}) \\ &= \Delta G_O - \Delta G_{OH}\end{aligned}$$

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(3) $H_2O\ (l) + O^* \rightarrow OOH^* + e^- + H^+$

$$\Delta G_1 = \mu_{OOH^*} + \mu_{H^+} + \mu_{e^-} - \mu_{O^*} - \mu_{H_2O}$$
$$= (\Delta E_{OOH} + \Delta E_{ZPE(OOH)} - T\Delta E_{OOH} + G_{solv(OOH^*)}) - (\Delta E_O + \Delta E_{ZPE(O)} - T\Delta E_O + G_{solv(O^*)})$$
$$= \Delta G_{OOH} - \Delta G_O$$

(4) $OOH^* \rightarrow O_2\ (g) + e^- + H^+$

$$\Delta G_1 = \mu_* + \mu_{O_2^*} + \mu_{H^+} + \mu_{e^-} - \mu_{OOH^*}$$
$$= 4.92 - (\Delta E_{OOH} + \Delta E_{ZPE(OOH)} - T\Delta E_{OOH} + G_{solv(OOH^*)})$$
$$= 4.92 - \Delta G_{OOH}$$

The overpotential for OER is obtained by:

$$\eta_{OER} = \frac{\max(\Delta G_1, \Delta G_2, \Delta G_3, \Delta G_4)}{e} - 1.23\text{V}$$

Although the zero-point energy and entropy of various OER intermediates for different SACs are different, the difference between them is very small which can be ignored, as shown in Table S3. Therefore, for the convenience of calculation, we adopted the data of previous work[S1] about the zero-point energy and entropy of various OER intermediates, as listed in Table S4.

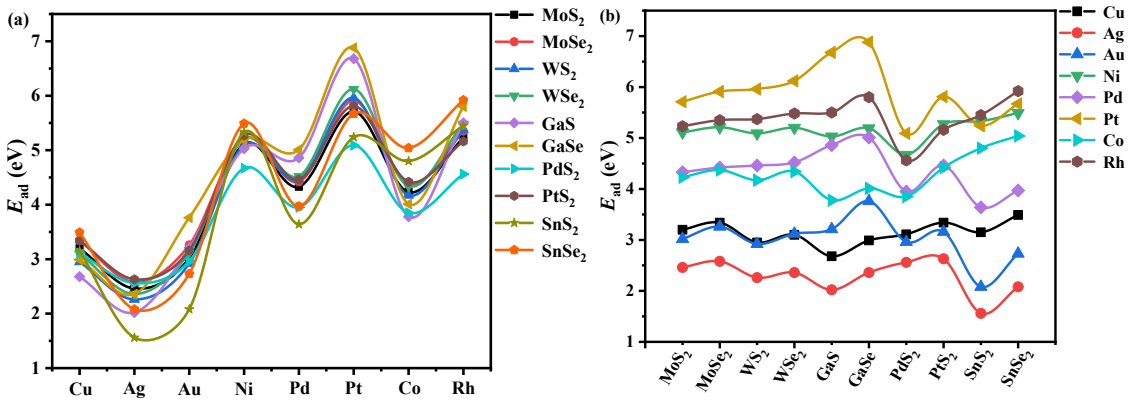


Fig. S1 Adsorption energies of various SACs. (some data are obtained from our previous work[S2])

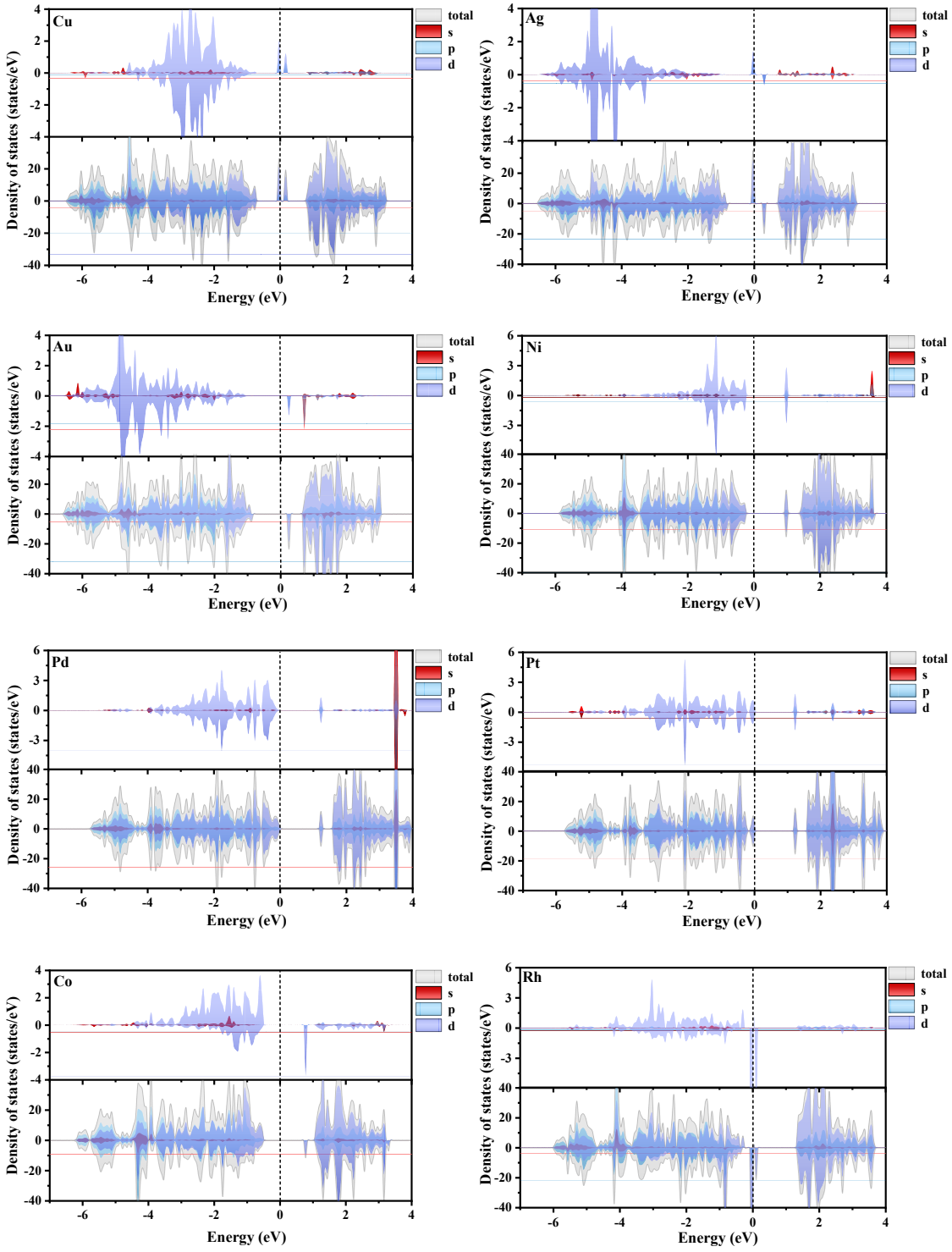


Fig. S2 Density of states for TM-$MoSe_2$.

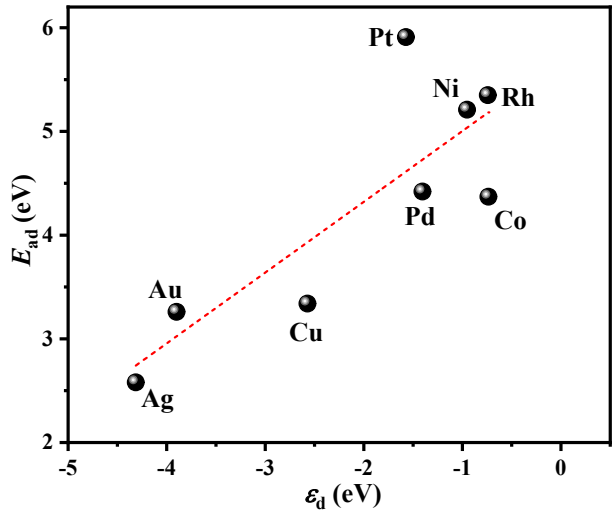
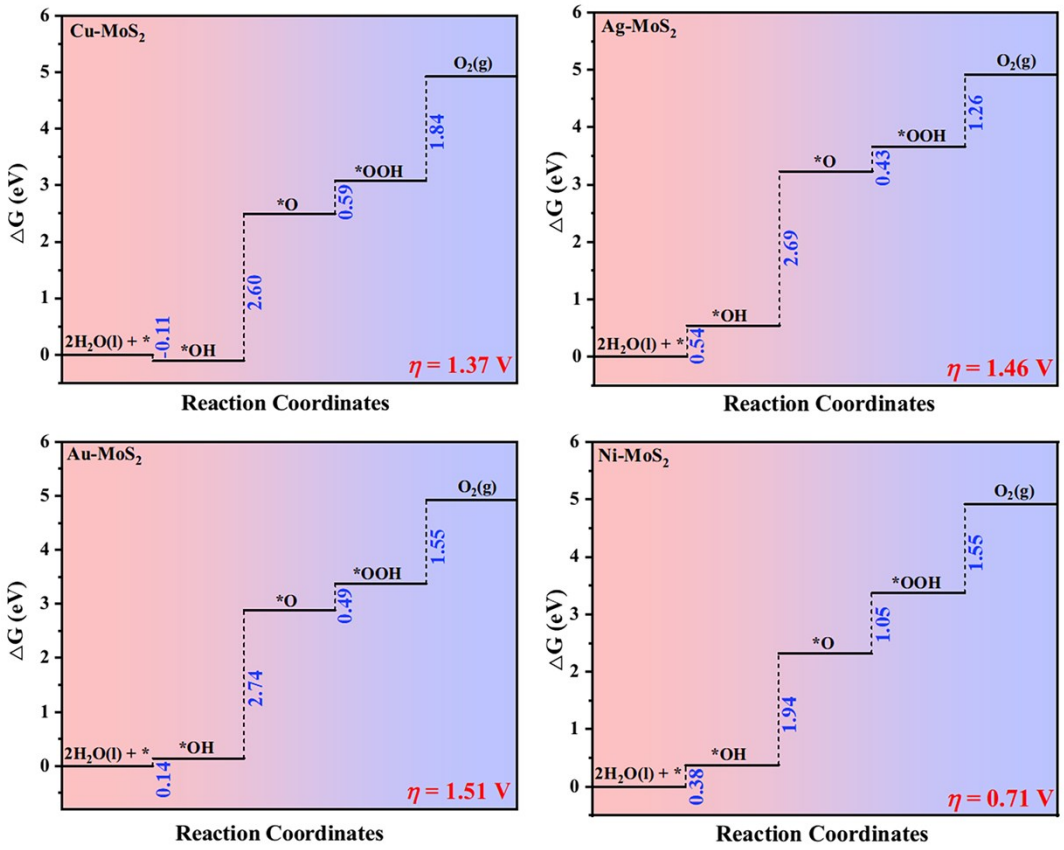
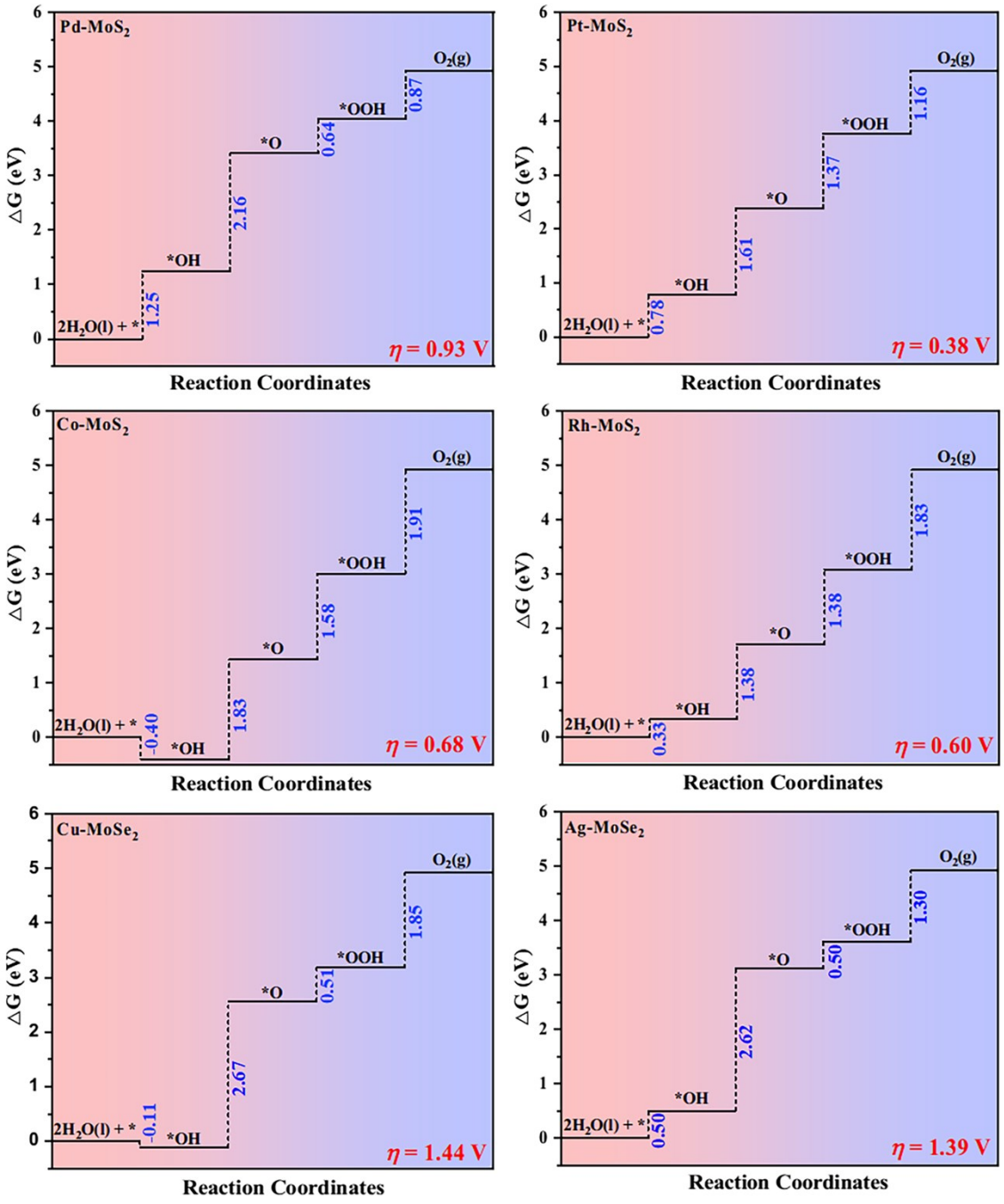
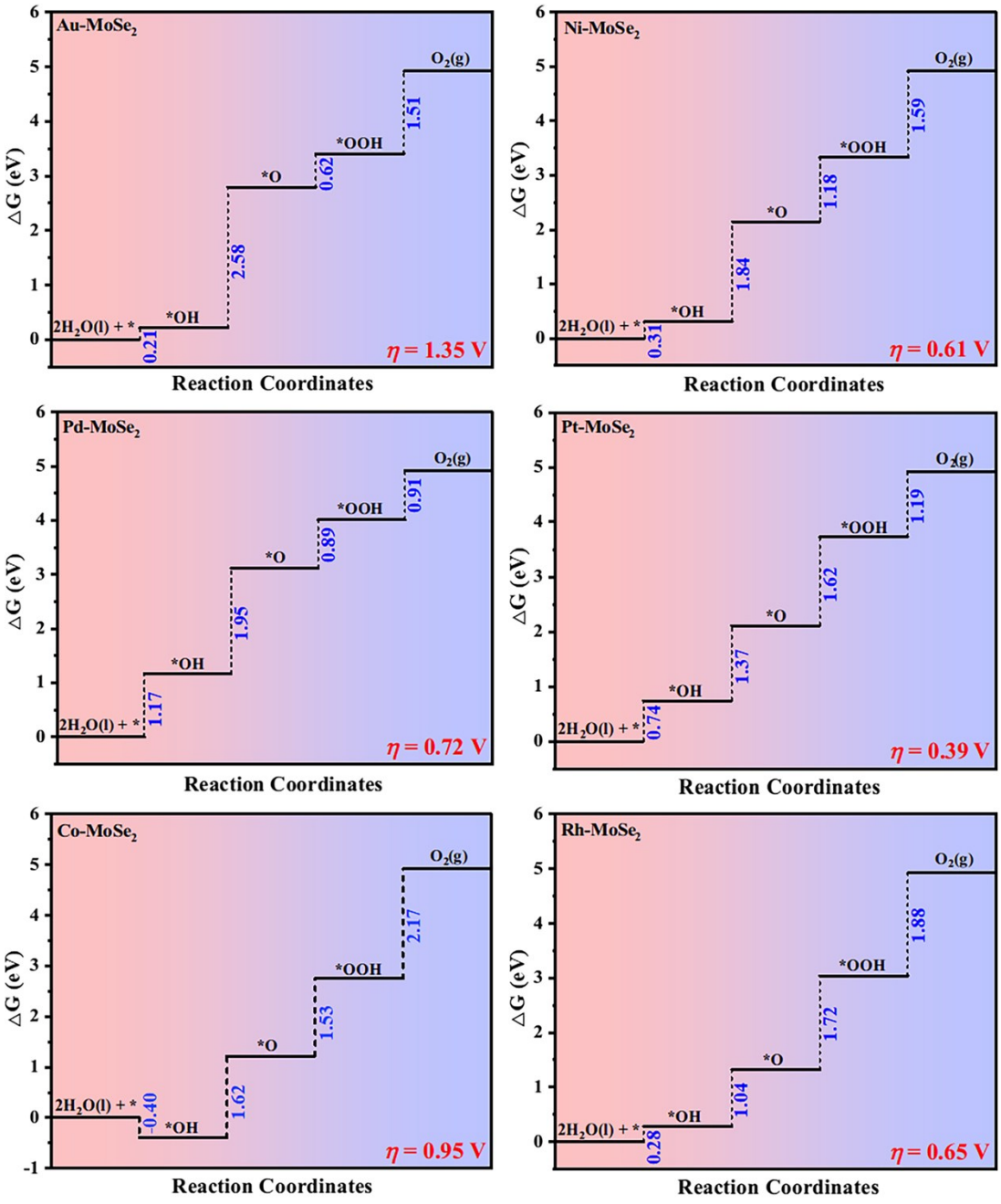
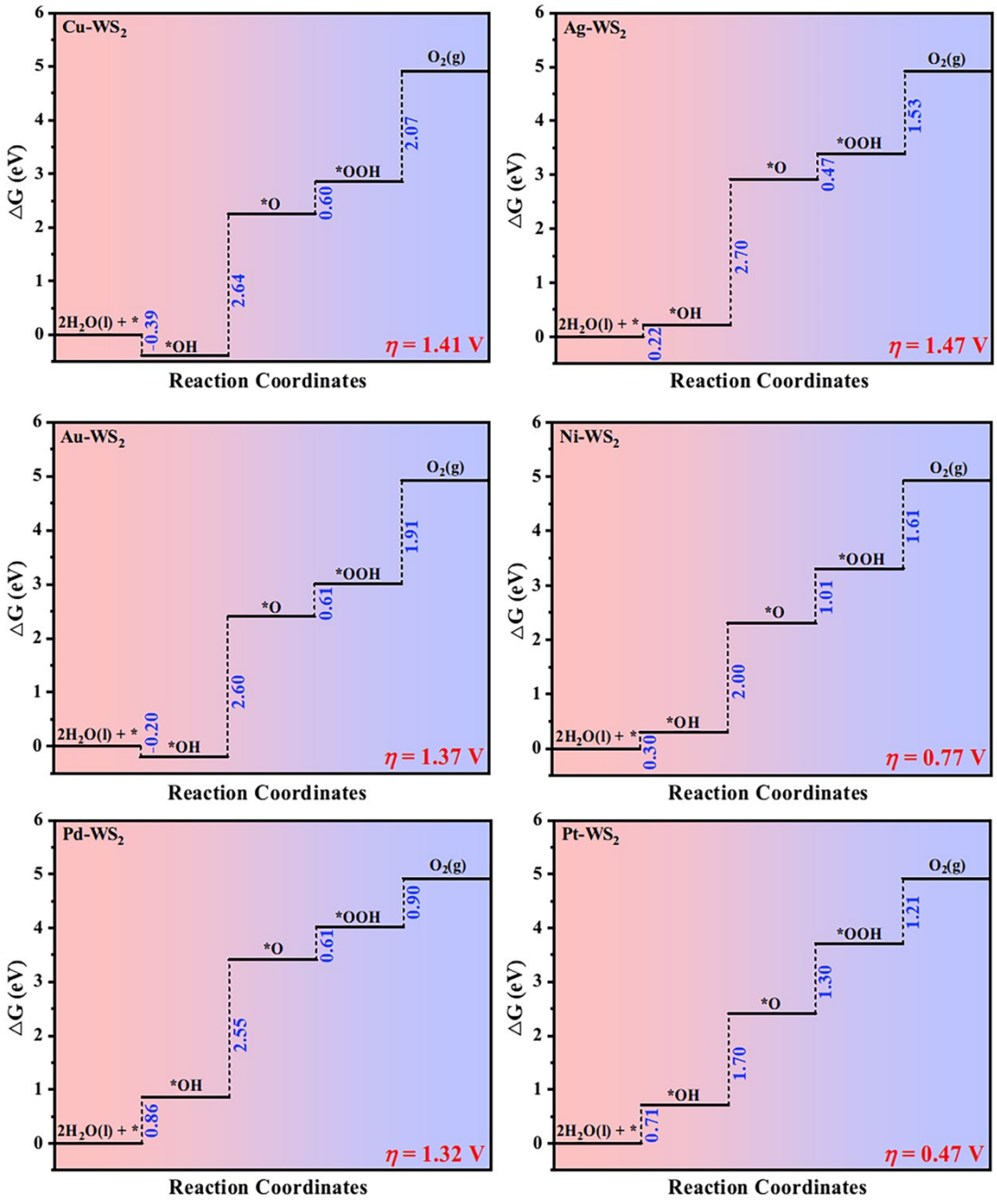


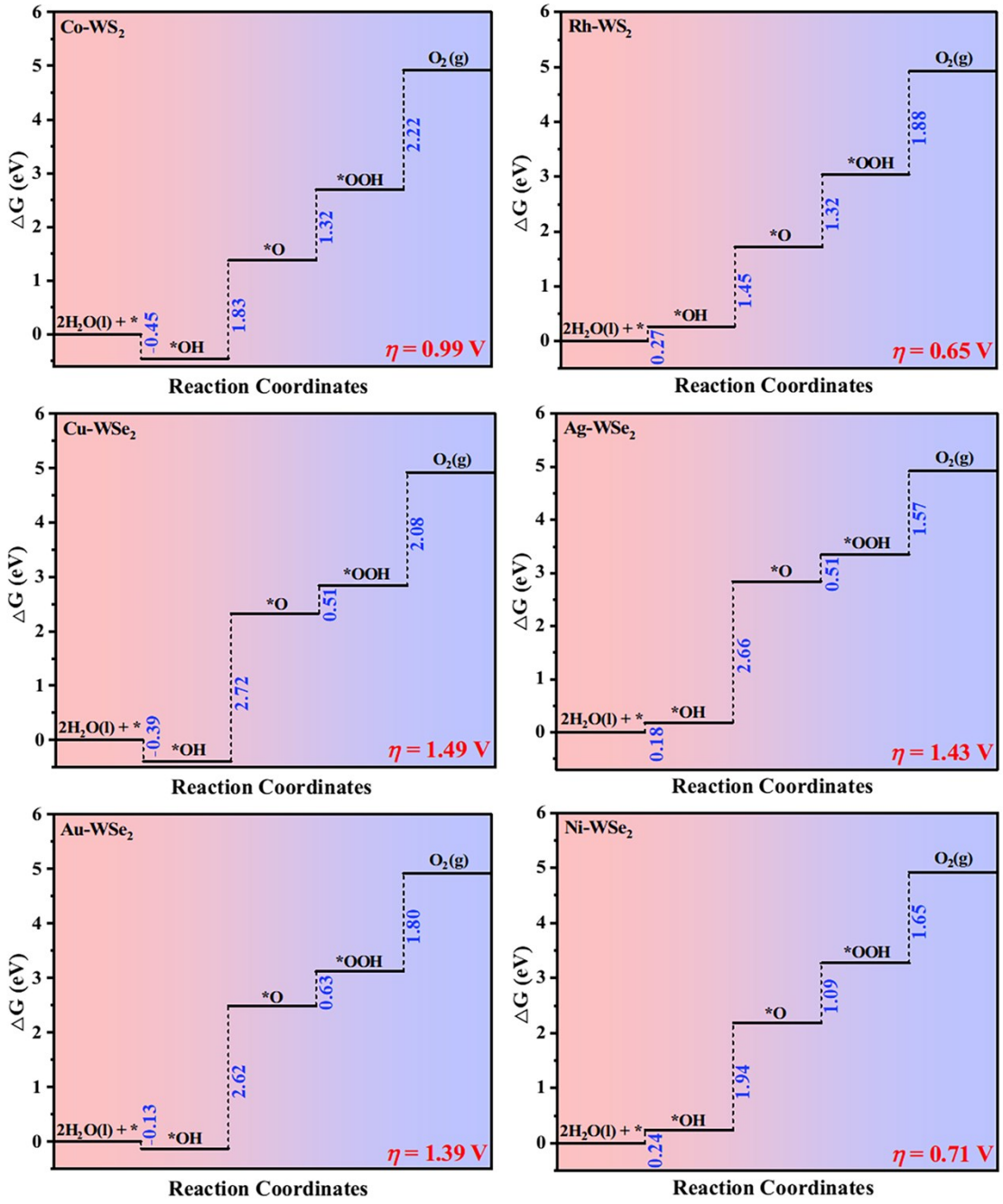
Fig.S3 Scaling relationship between the ε_d and E_{ad} of TM-$MoSe_2$.

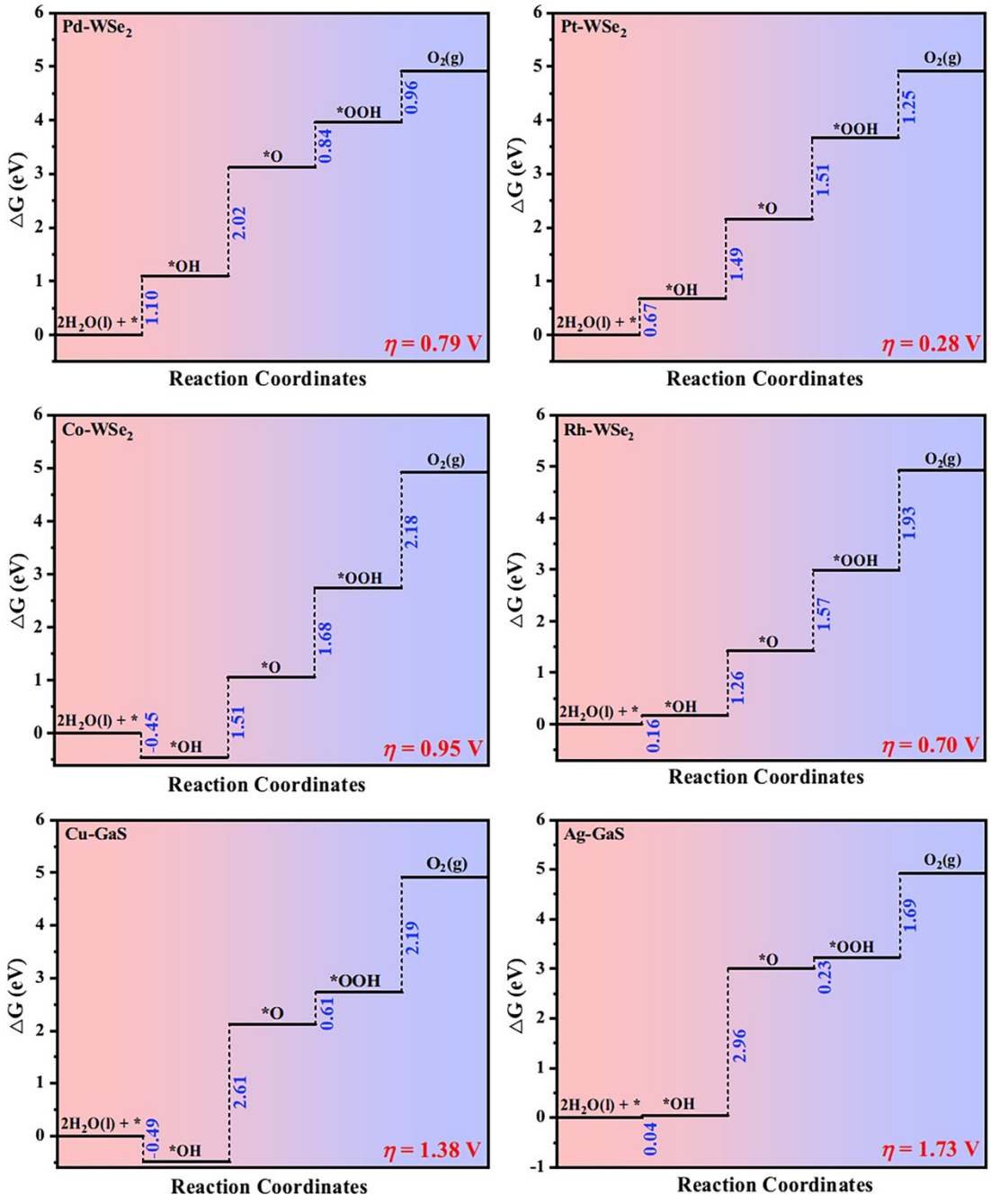


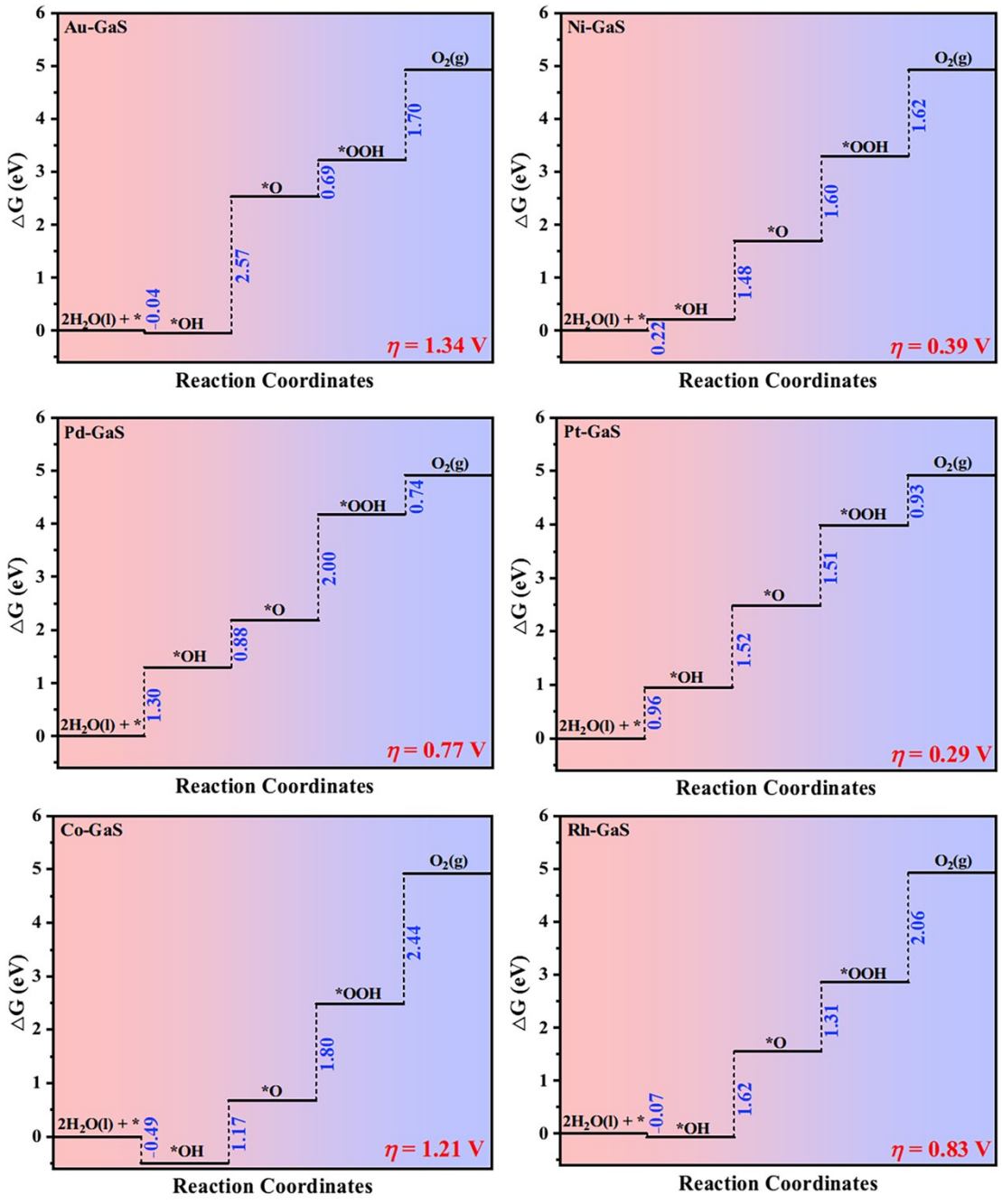

Pd-MoS2
2H2O(l) + *
*OH
*O
*OOH
O2(g)
1.25
2.16
0.64
0.87
η = 0.93 V
Pt-MoS2
0.78
1.61
1.37
1.16
η = 0.38 V
Co-MoS2
-0.40
1.83
1.58
1.91
η = 0.68 V
Rh-MoS2
0.33
1.38
1.38
1.83
η = 0.60 V
Cu-MoSe2
-0.11
2.67
0.51
1.85
η = 1.44 V
Ag-MoSe2
0.50
2.62
0.50
1.30
η = 1.39 V
ΔG (eV)
Reaction Coordinates

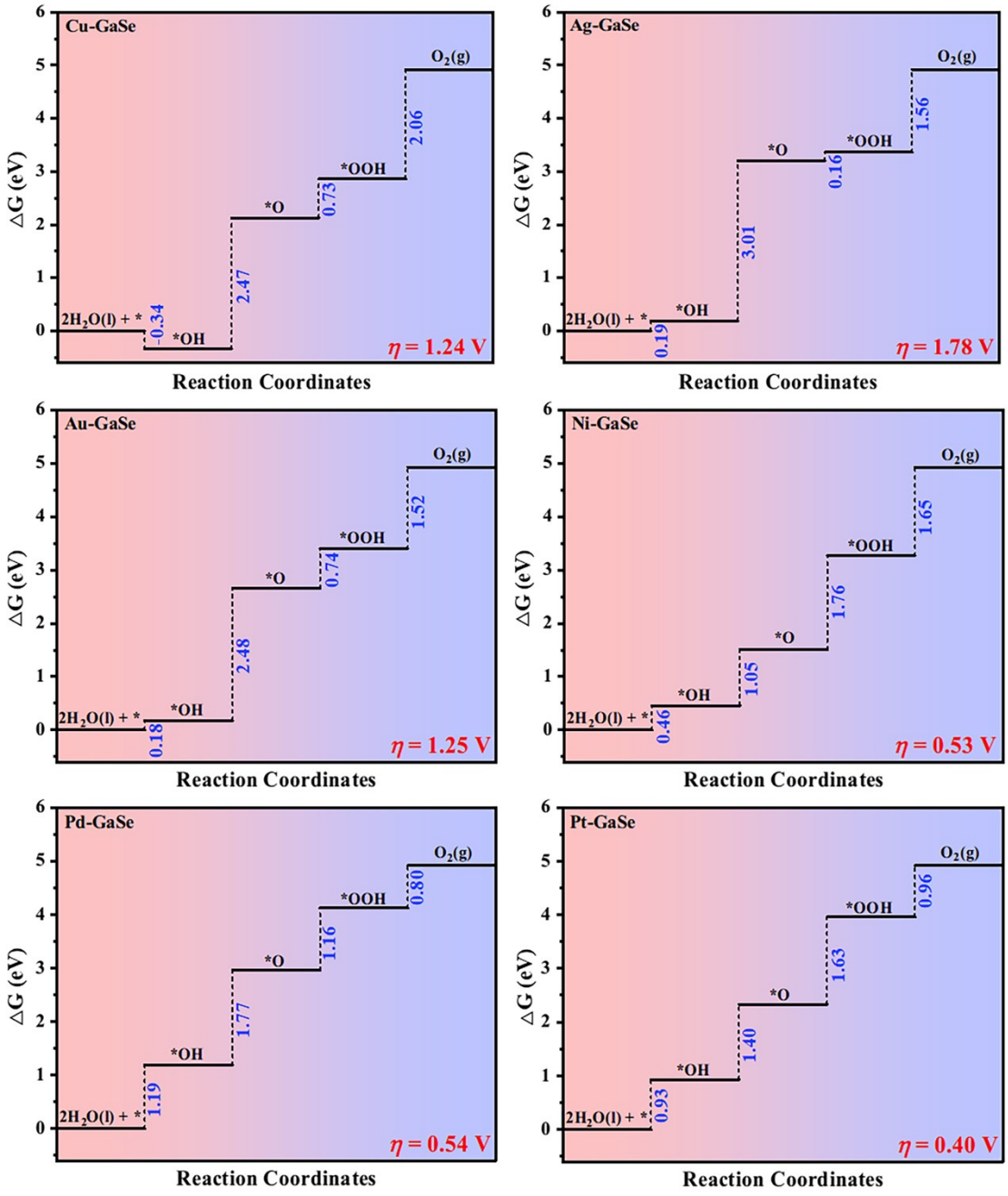

Au-MoSe2
Ni-MoSe2
Pd-MoSe2
Pt-MoSe2
Co-MoSe2
Rh-MoSe2
ΔG (eV)
Reaction Coordinates
2H2O(l) + *
*OH
*O
*OOH
O2(g)
0.21
2.58
0.62
1.51
η = 1.35 V
0.31
1.84
1.18
1.59
η = 0.61 V
1.17
1.95
0.89
0.91
η = 0.72 V
0.74
1.37
1.62
1.19
η = 0.39 V
-0.40
1.62
1.53
2.17
η = 0.95 V
0.28
1.04
1.72
1.88
η = 0.65 V

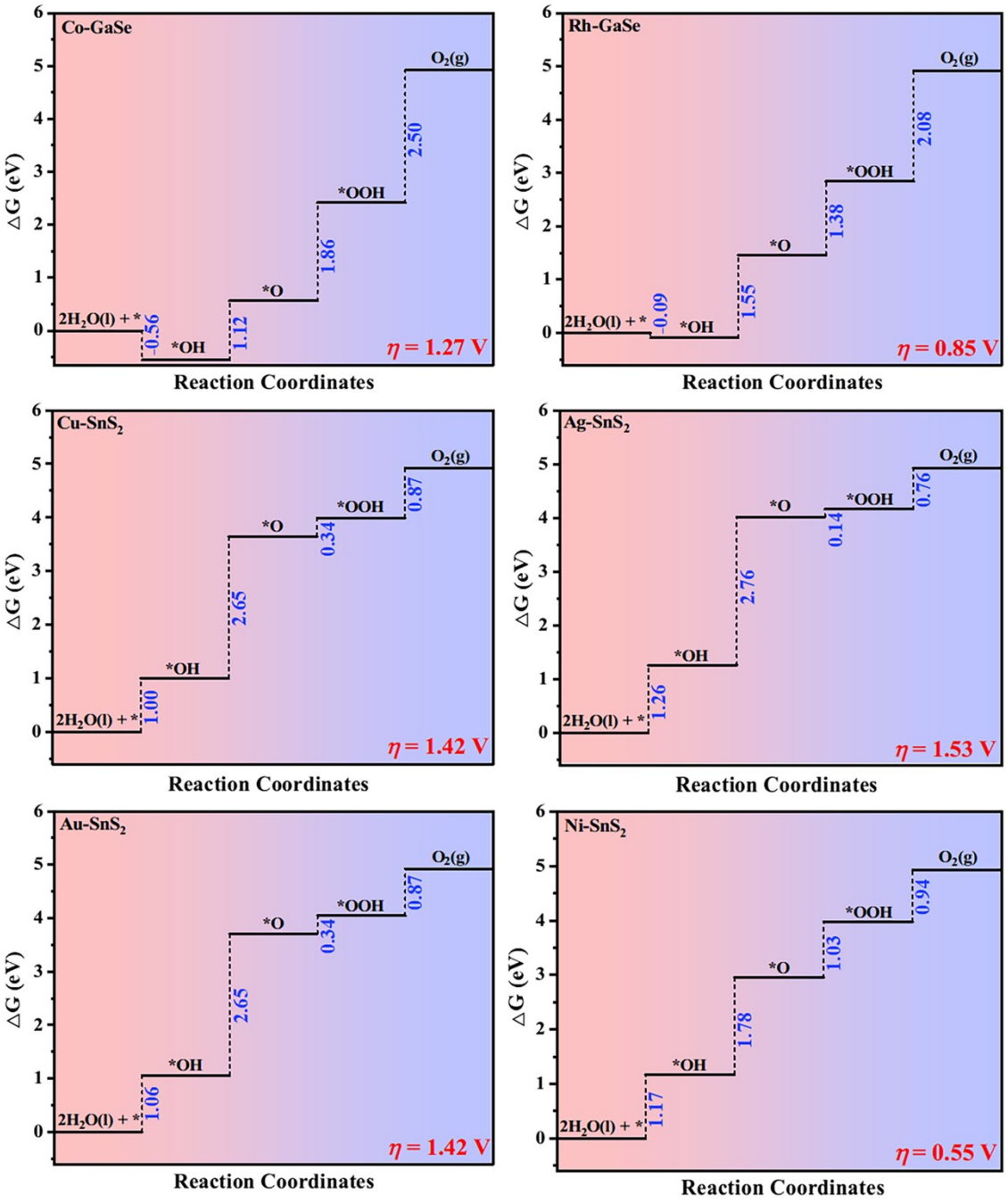

Cu-WS2
O2(g)
2H2O(l) + *
-0.39
*OH
2.64
*O
0.60
*OOH
2.07
η = 1.41 V
Ag-WS2
O2(g)
2H2O(l) + *
0.22
*OH
2.70
*O
0.47
*OOH
1.53
η = 1.47 V
Au-WS2
O2(g)
2H2O(l) + *
-0.20
*OH
2.60
*O
0.61
*OOH
1.91
η = 1.37 V
Ni-WS2
O2(g)
2H2O(l) + *
0.30
*OH
2.00
*O
1.01
*OOH
1.61
η = 0.77 V
Pd-WS2
O2(g)
2H2O(l) + *
0.86
*OH
2.55
*O
0.61
*OOH
0.90
η = 1.32 V
Pt-WS2
O2(g)
2H2O(l) + *
0.71
*OH
1.70
*O
1.30
*OOH
1.21
η = 0.47 V
ΔG (eV)
Reaction Coordinates

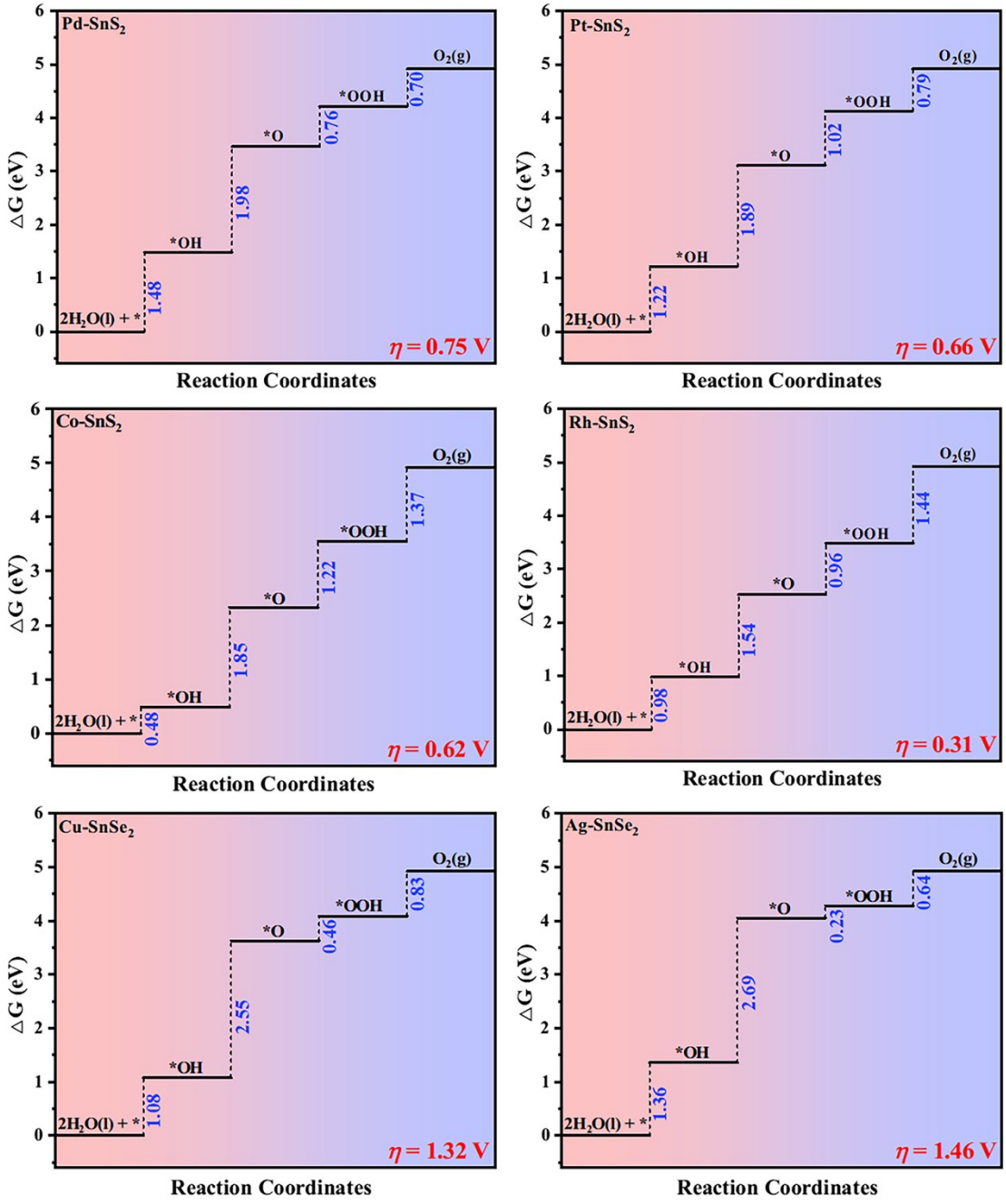

Co-WS2
O2(g)
2H2O(l) + *
-0.45
*OH
1.83
*O
1.32
*OOH
2.22
η = 0.99 V
Rh-WS2
O2(g)
2H2O(l) + *
0.27
*OH
1.45
*O
1.32
*OOH
1.88
η = 0.65 V
Cu-WSe2
O2(g)
2H2O(l) + *
-0.39
*OH
2.72
*O
0.51
*OOH
2.08
η = 1.49 V
Ag-WSe2
O2(g)
2H2O(l) + *
0.18
*OH
2.66
*O
0.51
*OOH
1.57
η = 1.43 V
Au-WSe2
O2(g)
2H2O(l) + *
-0.13
*OH
2.62
*O
0.63
*OOH
1.80
η = 1.39 V
Ni-WSe2
O2(g)
2H2O(l) + *
0.24
*OH
1.94
*O
1.09
*OOH
1.65
η = 0.71 V
ΔG (eV)
Reaction Coordinates

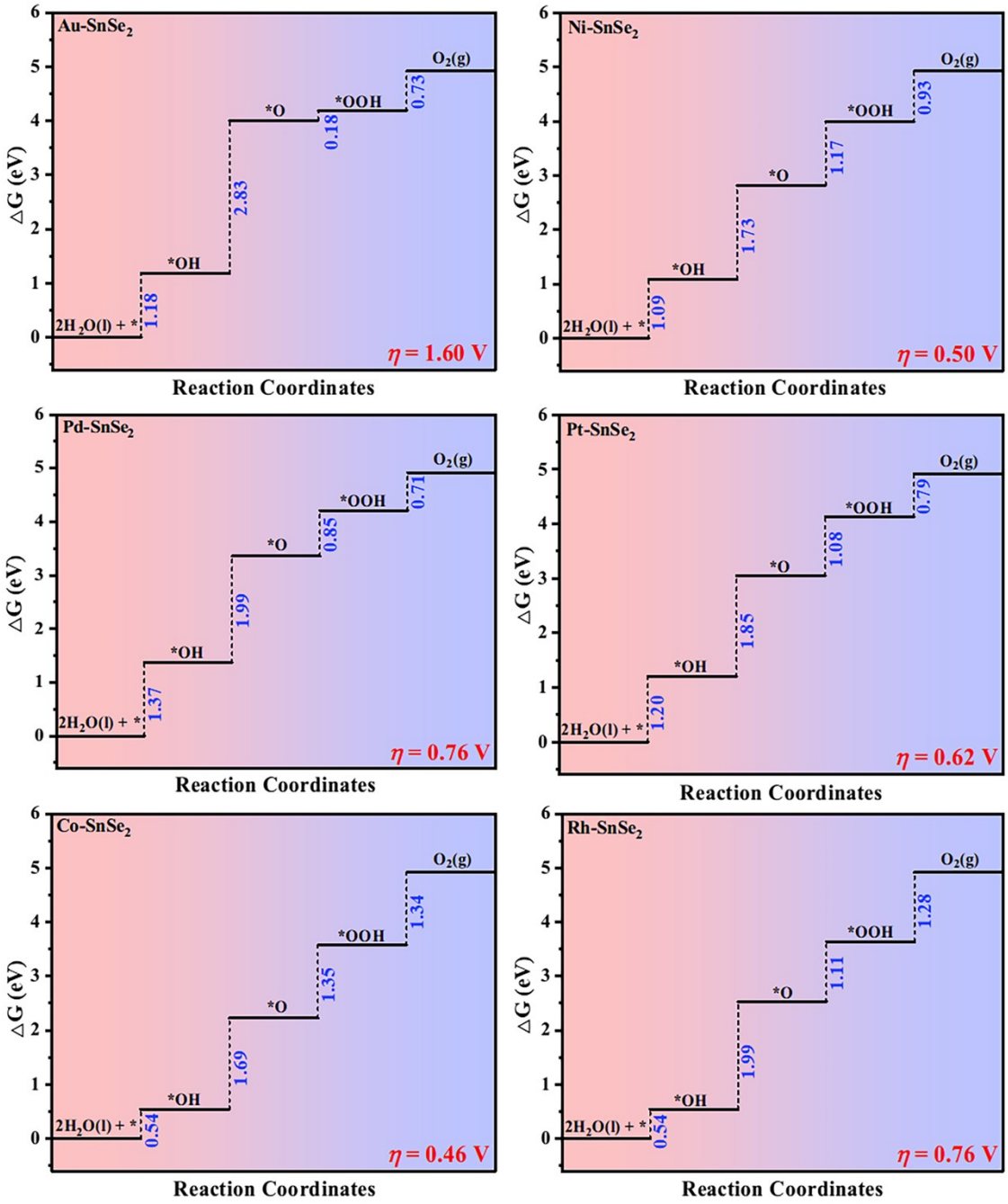

Pd-WSe2
2H2O(l) + *
*OH
*O
*OOH
O2(g)
1.10
2.02
0.84
0.96
η = 0.79 V
Pt-WSe2
0.67
1.49
1.51
1.25
η = 0.28 V
Co-WSe2
0.45
1.51
1.68
2.18
η = 0.95 V
Rh-WSe2
0.16
1.26
1.57
1.93
η = 0.70 V
Cu-GaS
0.49
2.61
0.61
2.19
η = 1.38 V
Ag-GaS
0.04
2.96
0.23
1.69
η = 1.73 V
ΔG (eV)
Reaction Coordinates

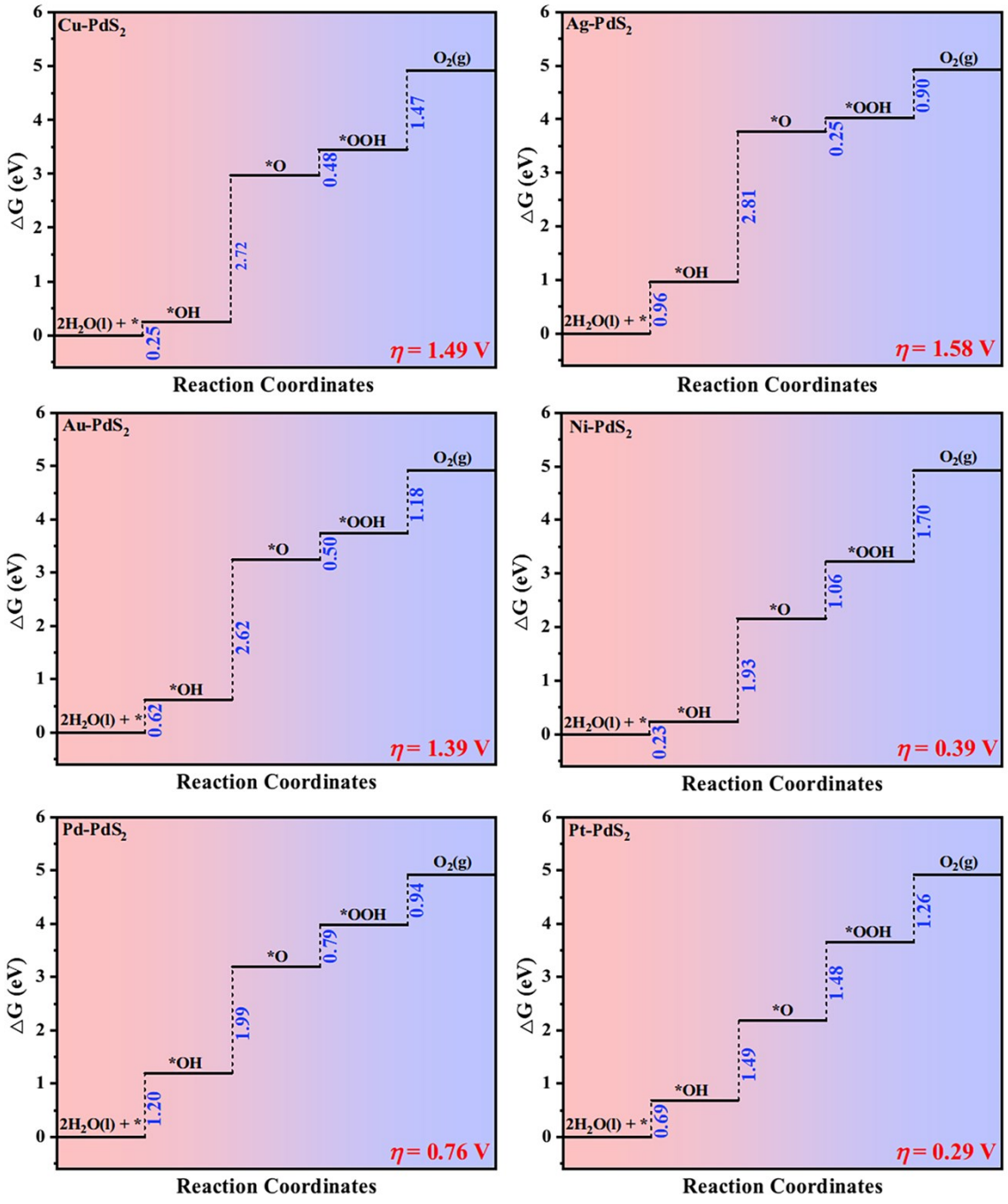

Au-GaS
$2H_2O(l) + *$
*OH
*O
*OOH
$O_2(g)$
-0.04
2.57
0.69
1.70
η = 1.34 V
Ni-GaS
0.22
1.48
1.60
1.62
η = 0.39 V
Pd-GaS
1.30
0.88
2.00
0.74
η = 0.77 V
Pt-GaS
0.96
1.52
1.51
0.93
η = 0.29 V
Co-GaS
-0.49
1.17
1.80
2.44
η = 1.21 V
Rh-GaS
-0.07
1.62
1.31
2.06
η = 0.83 V
△G (eV)
Reaction Coordinates

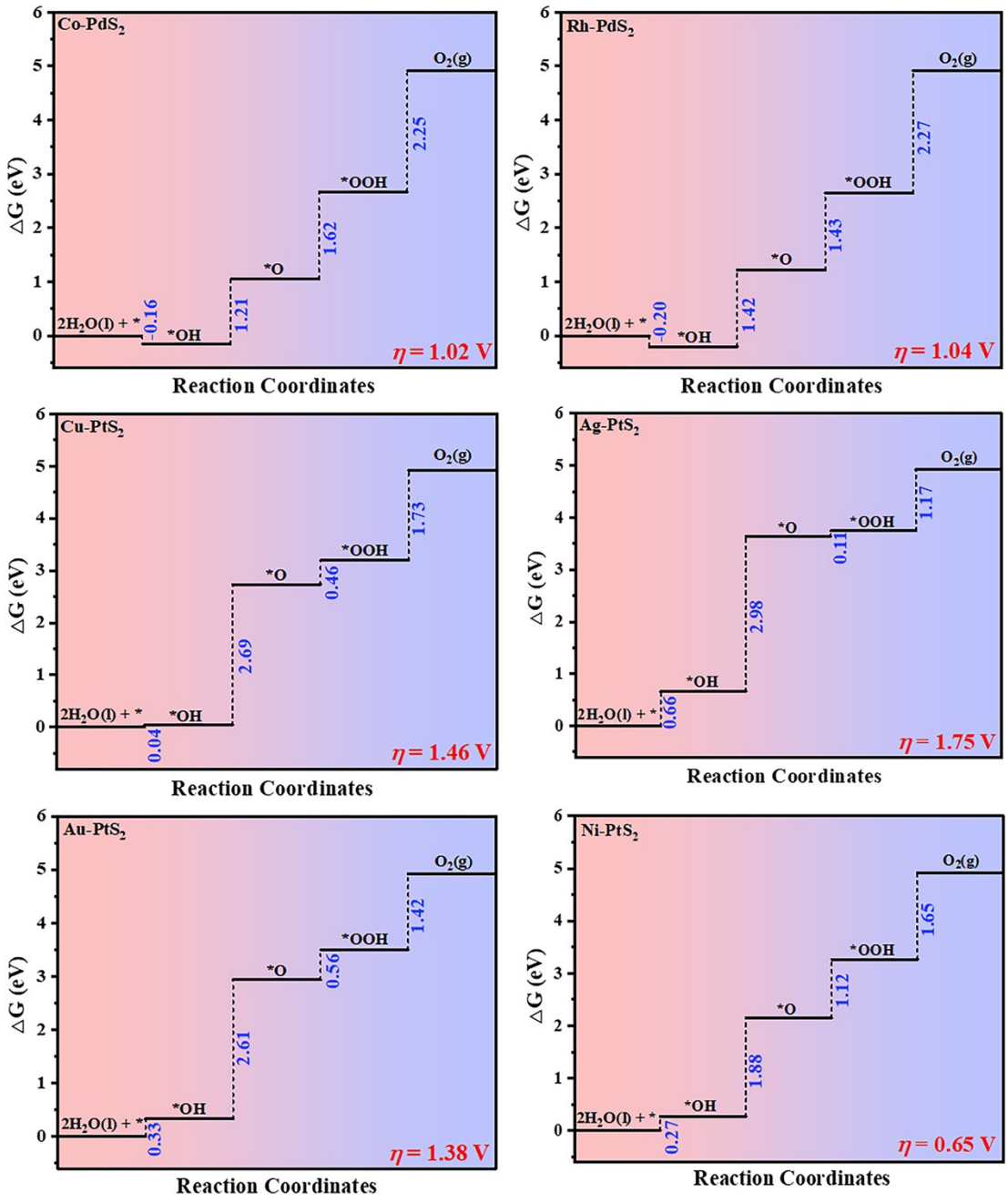

Cu-GaSe
$2H_2O(l) + *$
*OH
*O
*OOH
$O_2(g)$
0.34
2.47
0.73
2.06
η = 1.24 V
Ag-GaSe
$2H_2O(l) + *$
*OH
*O
*OOH
$O_2(g)$
0.19
3.01
0.16
1.56
η = 1.78 V
Au-GaSe
$2H_2O(l) + *$
*OH
*O
*OOH
$O_2(g)$
0.18
2.48
0.74
1.52
η = 1.25 V
Ni-GaSe
$2H_2O(l) + *$
*OH
*O
*OOH
$O_2(g)$
0.46
1.05
1.76
1.65
η = 0.53 V
Pd-GaSe
$2H_2O(l) + *$
*OH
*O
*OOH
$O_2(g)$
1.19
1.77
1.16
0.80
η = 0.54 V
Pt-GaSe
$2H_2O(l) + *$
*OH
*O
*OOH
$O_2(g)$
0.93
1.40
1.63
0.96
η = 0.40 V
△G (eV)
Reaction Coordinates


Co-GaSe
O_2(g)
2.50
*OOH
1.86
*O
1.12
*OH
-0.56
$2H_2O(l) + *$
η = 1.27 V
Rh-GaSe
O_2(g)
2.08
*OOH
1.38
*O
1.55
*OH
-0.09
$2H_2O(l) + *$
η = 0.85 V
Cu-SnS_2
O_2(g)
0.87
*OOH
0.34
*O
2.65
*OH
1.00
$2H_2O(l) + *$
η = 1.42 V
Ag-SnS_2
O_2(g)
0.76
*OOH
0.14
*O
2.76
*OH
1.26
$2H_2O(l) + *$
η = 1.53 V
Au-SnS_2
O_2(g)
0.87
*OOH
0.34
*O
2.65
*OH
1.06
$2H_2O(l) + *$
η = 1.42 V
Ni-SnS_2
O_2(g)
0.94
*OOH
1.03
*O
1.78
*OH
1.17
$2H_2O(l) + *$
η = 0.55 V
△G (eV)
Reaction Coordinates


Pd-SnS2
2H2O(l) + *
*OH
*O
*OOH
O2(g)
1.48
1.98
0.76
0.70
η = 0.75 V
Pt-SnS2
1.22
1.89
1.02
0.79
η = 0.66 V
Co-SnS2
0.48
1.85
1.22
1.37
η = 0.62 V
Rh-SnS2
0.98
1.54
0.96
1.44
η = 0.31 V
Cu-SnSe2
1.08
2.55
0.46
0.83
η = 1.32 V
Ag-SnSe2
1.36
2.69
0.23
0.64
η = 1.46 V
ΔG (eV)
Reaction Coordinates


Au-SnSe2
2H2O(l) + *
*OH
*O
*OOH
O2(g)
1.18
2.83
0.18
0.73
η = 1.60 V
Ni-SnSe2
1.09
1.73
1.17
0.93
η = 0.50 V
Pd-SnSe2
1.37
1.99
0.85
0.71
η = 0.76 V
Pt-SnSe2
1.20
1.85
1.08
0.79
η = 0.62 V
Co-SnSe2
0.54
1.69
1.35
1.34
η = 0.46 V
Rh-SnSe2
0.54
1.99
1.11
1.28
η = 0.76 V
ΔG (eV)
Reaction Coordinates


Cu-PdS2
O2(g)
*OOH
*O
*OH
2H2O(l) + *
0.25
2.72
0.48
1.47
η = 1.49 V
Ag-PdS2
0.96
2.81
0.25
0.90
η = 1.58 V
Au-PdS2
0.62
2.62
0.50
1.18
η = 1.39 V
Ni-PdS2
0.23
1.93
1.06
1.70
η = 0.39 V
Pd-PdS2
1.20
1.99
0.79
0.94
η = 0.76 V
Pt-PdS2
0.69
1.49
1.48
1.26
η = 0.29 V
ΔG (eV)
Reaction Coordinates


Co-PdS2
Rh-PdS2
Cu-PtS2
Ag-PtS2
Au-PtS2
Ni-PtS2
ΔG (eV)
Reaction Coordinates
2H2O(l) + *
*OH
*O
*OOH
O2(g)
-0.16
1.21
1.62
2.25
-0.20
1.42
1.43
2.27
0.04
2.69
0.46
1.73
0.66
2.98
0.11
1.17
0.33
2.61
0.56
1.42
0.27
1.88
1.12
1.65
η = 1.02 V
η = 1.04 V
η = 1.46 V
η = 1.75 V
η = 1.38 V
η = 0.65 V

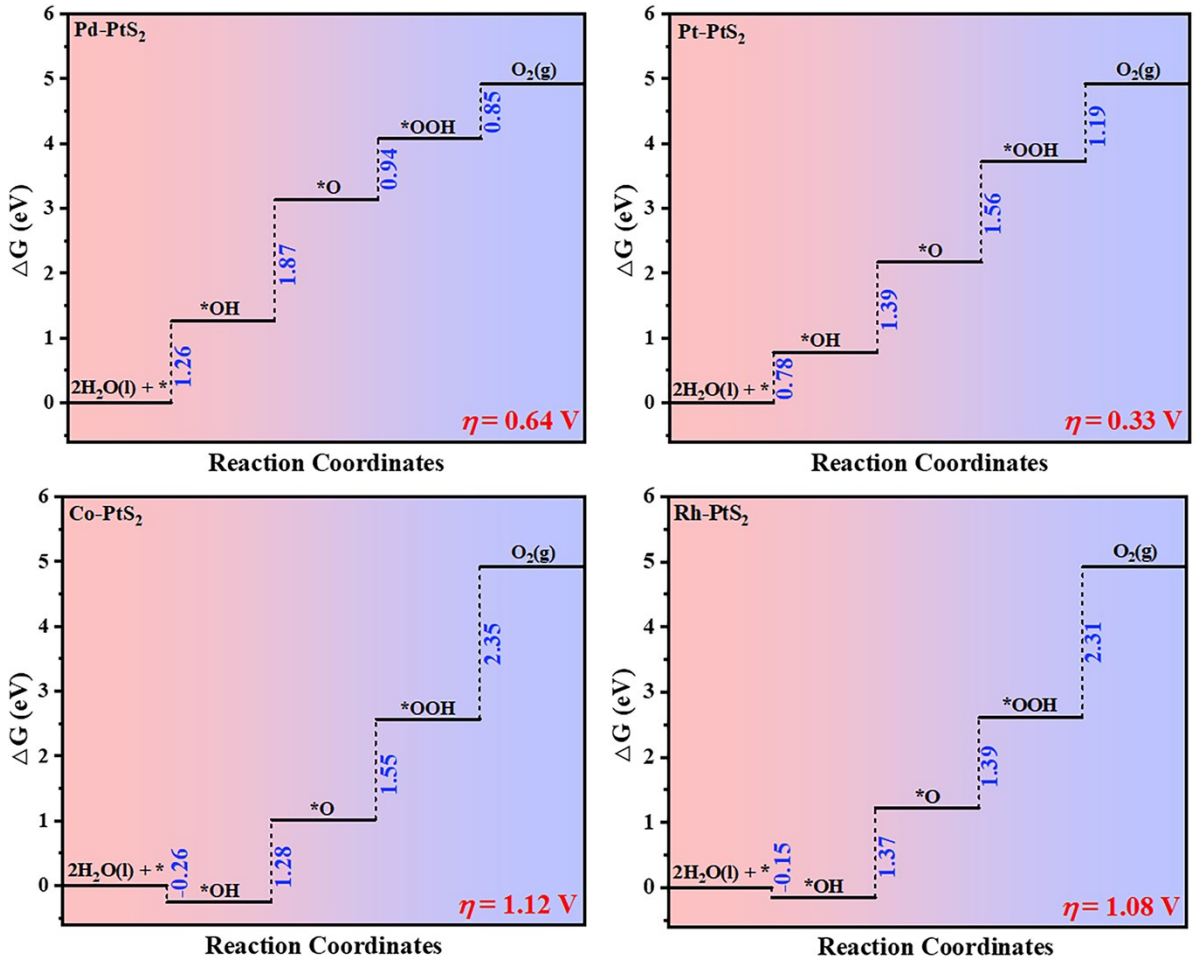


Fig. S4 Free energy diagrams for various SACs.

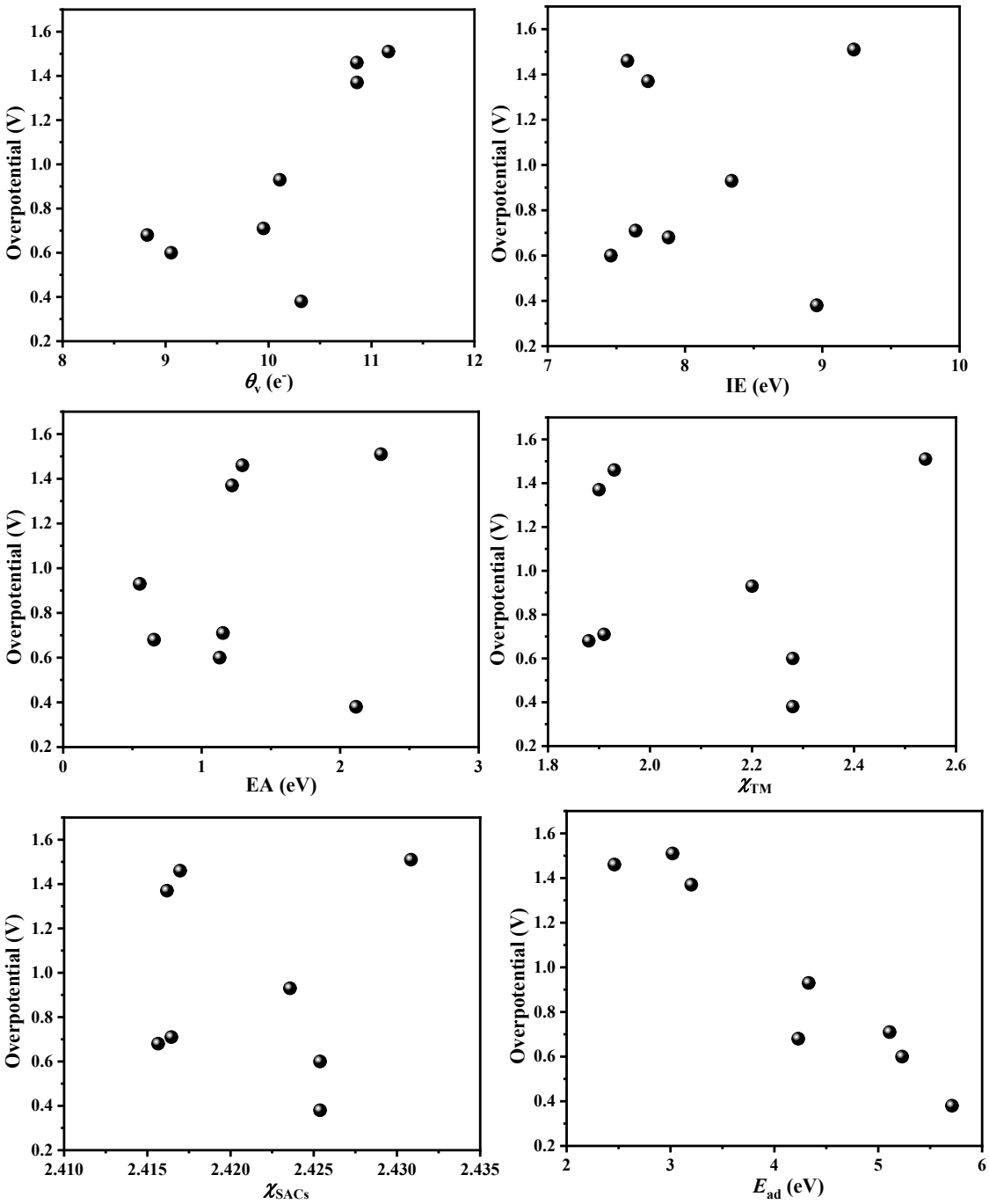


Fig. S5 Scaling relationship between the θ_v, IE, EA, χ_{TM}, χ_{SACs}, E_{ad} and the OER overpotentials of TM-MoS_2.

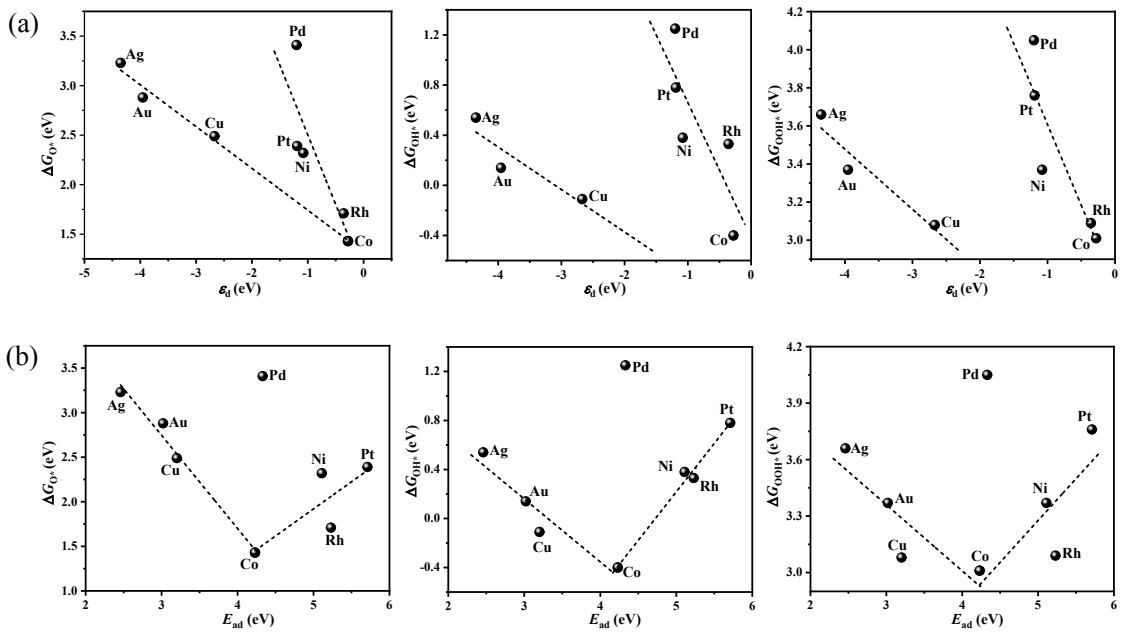


Fig. S6 Scaling relationship between (a) the *d*-band center; (b) the E_{ad} of TM atoms and $\Delta G_{O*}/\Delta G_{OH*}/\Delta G_{OOH*}$ for TM-MoS_2.

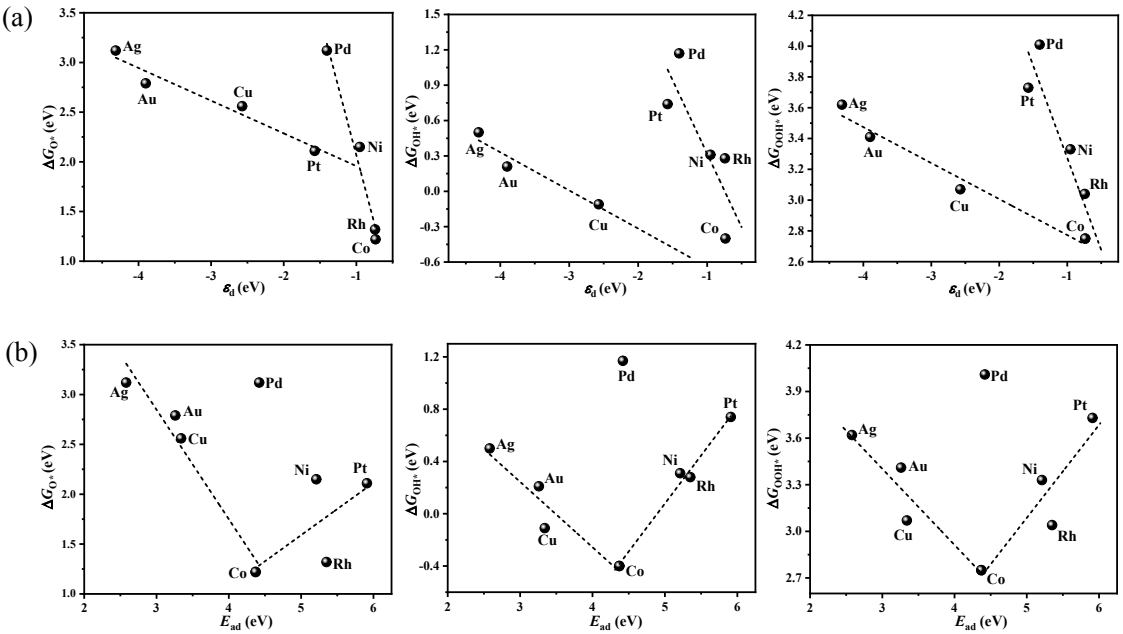


Fig. S7 Scaling relationship between (a) the *d*-band center; (b) the E_{ad} of TM atoms and $\Delta G_{O*}/\Delta G_{OH*}/\Delta G_{OOH*}$ for TM-$MoSe_2$.

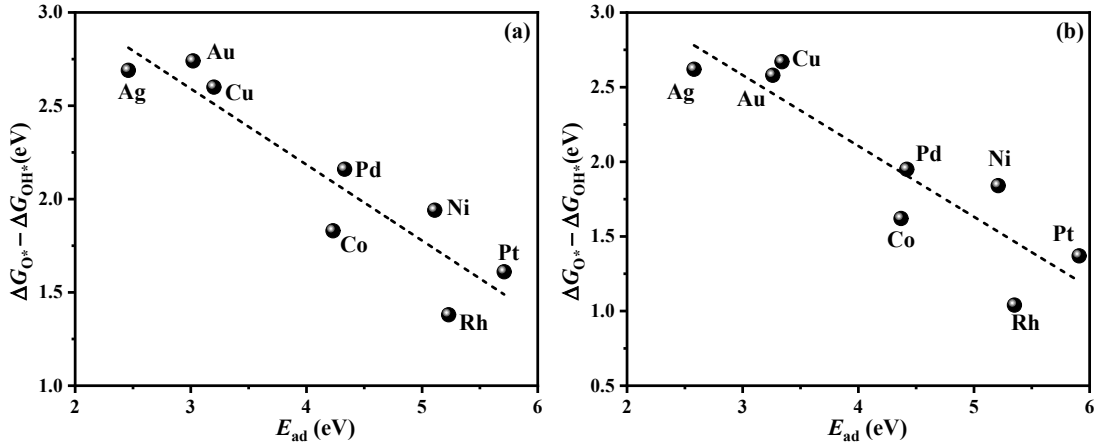


Fig. S8 Scaling relationship between the E_{ad} and ΔG_{O^*} - ΔG_{OH^*} of (a) MoS_2 and (b) $MoSe_2$.

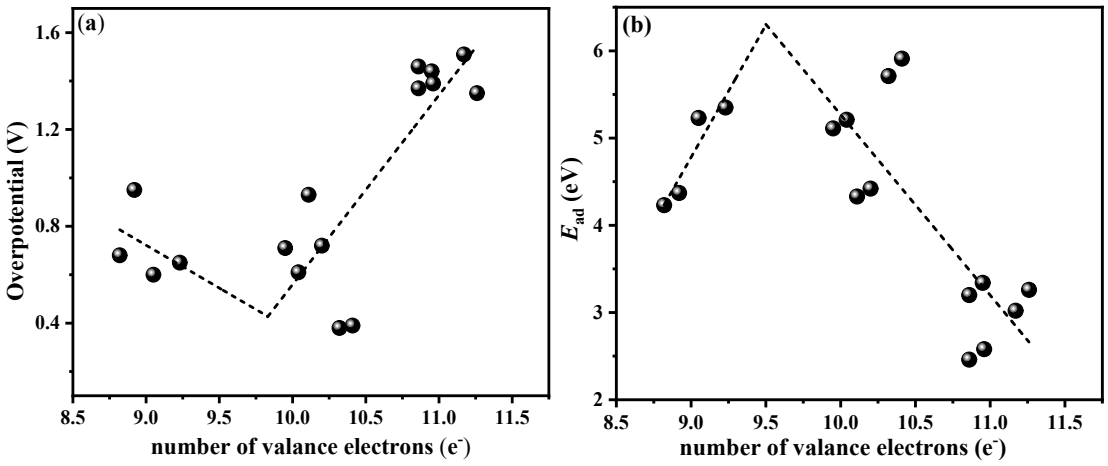


Fig. S9 Scaling relationship between the θ_v of active center and (a) the overpotential of the reaction; (b) E_{ad} of the TMs.

Table S1. Calculated Lattice Parameters (Å) and Bond Lengths (Å) of Various Monolayer Materials. (A Represents the Metal Atoms and X Represents the S/Se Atoms)

substrates	$a = b$		d_{A-X}	
	this work	previous works	this work	previous works
MoS_2	3.18[S2]	3.16[S3]	2.41[S2]	2.41[S4]
$MoSe_2$	3.29[S2]	3.29[S3]	2.53[S2]	2.54[S5]
WS_2	3.17[S2]	3.15[S3]	2.41[S2]	2.40[S6]
WSe_2	3.29[S2]	3.29[S3]	2.54[S2]	2.53[S6]
GaS	3.62[S2]	3.64[S7]	2.36[S2]	2.34[S8]
GaSe	3.80[S2]	3.82[S7]	2.49[S2]	2.47[S8]
SnS_2	3.67	3.65[S9]	2.59	2.63[S10]
$SnSe_2$	3.85	3.81[S11]	2.74	2.73[S12]
PdS_2	3.52	3.53[S13]	2.39	2.40[S13]
PtS_2	3.54	3.54[S13]	2.39	2.39[S14]

Table S2. Adsorption energies of various SACs. The Unit for Energy is eV.

	Cu	Ag	Au	Ni	Pd	Pt	Co	Rh
MoS_2	3.20	2.46	3.02	5.11	4.33	5.71	4.2	5.23
$MoSe_2$	3.24	2.58	3.26	5.21	4.42	5.91	4.37	5.35
WS_2	2.95	2.26	2.92	5.09	4.46	5.96	4.17	5.37
WSe_2	3.10	2.36	3.12	5.20	4.52	6.12	4.34	5.48
GaS	2.68	2.02	3.21	5.03	4.86	6.68	3.78	5.50
GaSe	2.99	2.36	3.76	5.19	5.01	6.88	4.01	5.80
SnS_2	3.15	1.56	2.08	5.33	3.64	5.24	4.80	5.45
$SnSe_2$	3.49	2.08	2.73	5.49	3.97	5.67	5.04	5.92
PdS_2	3.11	2.56	2.96	4.67	3.95	5.09	3.85	4.56
PtS_2	3.34	2.63	3.16	5.27	4.46	5.81	4.42	5.16

Table S3. The Zero-point Energy (E_{ZPE}) of Various OER Intermediates on TM-MoS_2. The Unit for Energy is eV.

	O*	OH*	OOH*
ref[S1]	0.07	0.36	0.39
Cu-MoS_2	0.04	0.32	0.43
Ag-MoS_2	0.03	0.31	0.41
Au-MoS_2	0.05	0.31	0.42
Ni-MoS_2	0.05	0.33	0.42
Pd-MoS_2	0.06	0.32	0.42
Pt-MoS_2	0.07	0.33	0.43
Co-MoS_2	0.06	0.33	0.41
Rh-MoS_2	0.07	0.34	0.44

Table S4. The Solvation Energies Corrections (G_{solv}), Zero-point Energy (E_{ZPE}) and Entropy (S) of Gas or Liquid Phase Molecules and Various OER Intermediates. The Unit for Energy is eV.

Species	G_{solv}	E_{ZPE}	TS
H_2O (l)	0.00	0.56	0.67
H_2 (g)	0.00	0.27	0.41
O*	0.00	0.07	0.00
OH*	-0.30	0.36	0.00
OOH*	-0.30	0.39	0.00

The data are obtained from the ref[S1]

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