

Single transition metal atoms anchored on WSe₂/WS₂ heterostructures as efficient bifunctional electrocatalysts for OER/ORR reactions

Kai Zhang^{a†}, Jianjun Fang^{a†}, Chenkun Li^a, Tongmeng Xing^a, Qiuyan

Xiang^a, Xianfang Yue^b, Breno R. L. Galvão^{a,c,*} and Jing Li^{a†}

^a*Shandong Key Laboratory of Medical Physics and Image Processing,*

Shandong Engineering Research Center for Medical Physical Imaging Technology,

School of Physics and Physical Engineering, Qufu Normal University, Qufu 273165, Shandong, China

^b*Department of Physics and Information Engineering, Jining University, Qufu 273155, China and*

^c*Centro Federal de Educação Tecnológica de Minas Gerais, CEFET-MG,*

Av. Amazonas 5253, 30421-169 Belo Horizonte, Minas Gerais, Brazil

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* Corresponding author: brenogalvao@gmail.com

† Corresponding author: jli@qfnu.edu.cn

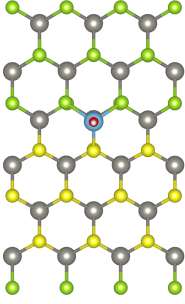
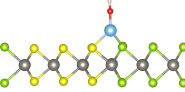
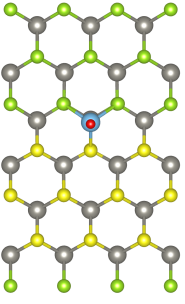
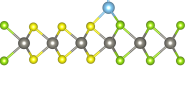
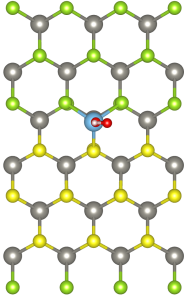
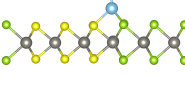
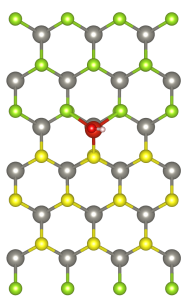
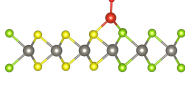
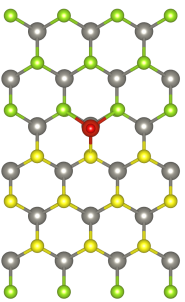
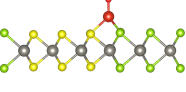
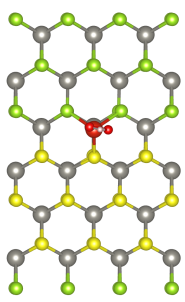
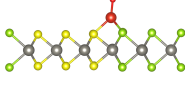
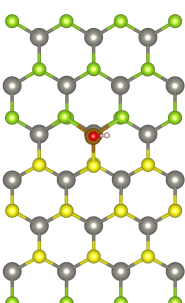
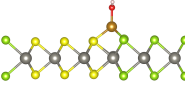
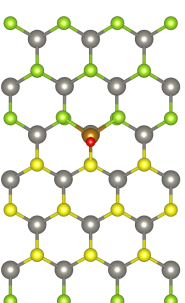
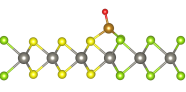
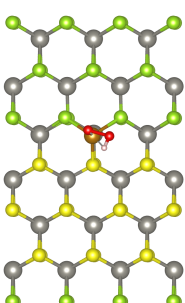
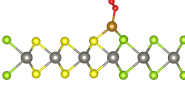
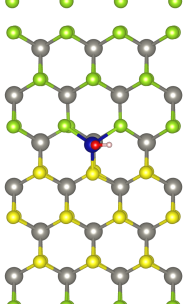
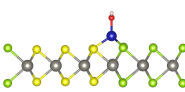
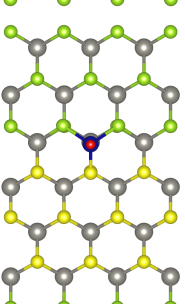
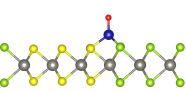
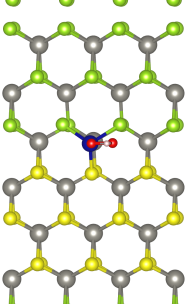
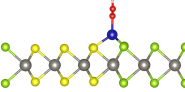
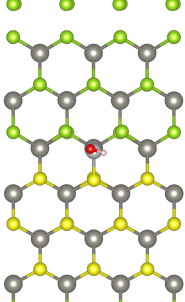
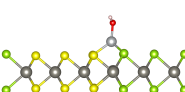
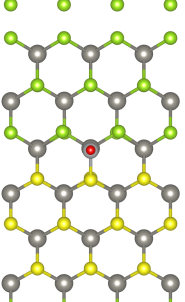
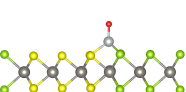
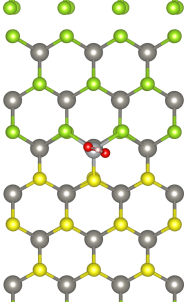
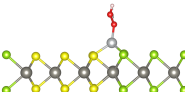









TABLE S1. Calculated binding energies of various TM single atoms at adsorption sites on the WSe₂/WS₂ heterostructure. The most stable sites are marked in bold. All values are in eV.

Systems	T _{Se} site	T _{Se-S} site	T _S site	H _{Se} site	H _{Se-S} site	H _S site
Ti@WSe ₂ /WS ₂	-1.77	-2.20	-1.93	-1.35	-2.20	-1.98
V@WSe ₂ /WS ₂	-1.64	-2.09	-1.77	-1.04	-2.09	-1.56
Cr@WSe ₂ /WS ₂	-0.69	-0.90	-0.77	-0.60	-0.71	-0.72
Mn@WSe ₂ /WS ₂	-0.39	-0.71	-0.51	-0.27	-0.40	-0.57
Fe@WSe ₂ /WS ₂	-1.18	-1.92	-1.69	-0.79	-1.48	-1.46
Co@WSe ₂ /WS ₂	-2.25	-2.69	-2.46	-2.01	-2.23	-2.36
Ni@WSe ₂ /WS ₂	-3.13	-3.53	-3.29	-2.68	-3.02	-3.15
Cu@WSe ₂ /WS ₂	-0.91	-0.97	-0.97	-0.84	-0.92	-0.90
Zr@WSe ₂ /WS ₂	-2.00	-2.43	-2.15	-1.56	-2.43	-2.40
Nb@WSe ₂ /WS ₂	-1.76	-2.23	-1.94	-1.21	-2.23	-1.55
Mo@WSe ₂ /WS ₂	-0.56	-0.89	-0.61	-0.51	-0.58	-0.58
Ru@WSe ₂ /WS ₂	-2.76	-3.15	-2.93	-1.88	-2.06	-2.39
Rh@WSe ₂ /WS ₂	-2.80	-3.08	-2.92	-2.11	-3.08	-2.30
Pd@WSe ₂ /WS ₂	-1.78	-1.99	-1.85	-1.55	-1.99	-1.62
Ag@WSe ₂ /WS ₂	-0.36	-0.33	-0.36	-0.40	-0.40	-0.37
Ta@WSe ₂ /WS ₂	-1.90	-2.34	-2.07	-1.27	-2.34	-1.52
W@WSe ₂ /WS ₂	-1.12	-1.50	-1.26	-0.86	-1.03	-1.07
Re@WSe ₂ /WS ₂	-0.56	-1.21	-0.97	-0.17	-1.21	-0.73
Os@WSe ₂ /WS ₂	-2.16	-2.54	-2.34	-0.87	-2.54	-2.03
Ir@WSe ₂ /WS ₂	-2.66	-2.94	-2.80	-1.64	-2.94	-1.86
Pt@WSe ₂ /WS ₂	-2.48	-2.68	-2.56	-2.01	-2.68	-2.09
Au@WSe ₂ /WS ₂	-0.54	-0.64	0.58	-0.64	-0.58	-0.53

TABLE S2. The bond lengths between TM atoms and neighboring Se/S atoms (d_{TM-Se_1} , d_{TM-Se_2} and d_{TM-S} in Å) and the charge transfer between TM atoms and neighboring Se/S atoms (Q_{TM} , Q_{Se_1} , Q_{Se_2} and Q_S in e).

Systems	d_{TM-Se_1}	d_{TM-Se_2}	d_{TM-S}	Q_{TM}	Q_{Se_1}	Q_{Se_2}	Q_S
Ti@WSe ₂ /WS ₂	2.50	2.50	2.32	-1.02	1.04	1.01	1.00
V@WSe ₂ /WS ₂	2.44	2.44	2.29	-0.90	0.87	0.84	0.86
Fe@WSe ₂ /WS ₂	2.26	2.26	2.12	-0.58	0.78	0.76	0.89
Co@WSe ₂ /WS ₂	2.25	2.25	2.07	-0.42	0.87	0.85	0.79
Ni@WSe ₂ /WS ₂	2.24	2.24	2.09	-0.35	0.92	0.90	0.75
Zr@WSe ₂ /WS ₂	2.61	2.61	2.45	-1.03	0.97	0.93	0.82
Ru@WSe ₂ /WS ₂	2.37	2.37	2.24	-0.51	0.79	0.77	0.75
Rh@WSe ₂ /WS ₂	2.38	2.38	2.23	-0.42	0.78	0.76	0.75
Pd@WSe ₂ /WS ₂	2.46	2.46	2.30	-0.19	0.51	0.48	0.64
Ir@WSe ₂ /WS ₂	2.37	2.37	2.22	-0.38	0.58	0.56	0.63
Pt@WSe ₂ /WS ₂	2.43	2.43	2.26	-0.38	0.75	0.73	0.37

TABLE S3. The optimized structures of OH*, O* and OOH* intermediates on the different TM@WSe₂/WS₂.

TM	OH*		O*		OOH*	
	Top view	Side view	Top view	Side view	Top view	Side view
Ti						
						
						
						
V						
						
						
Fe						
Co						
Ni						

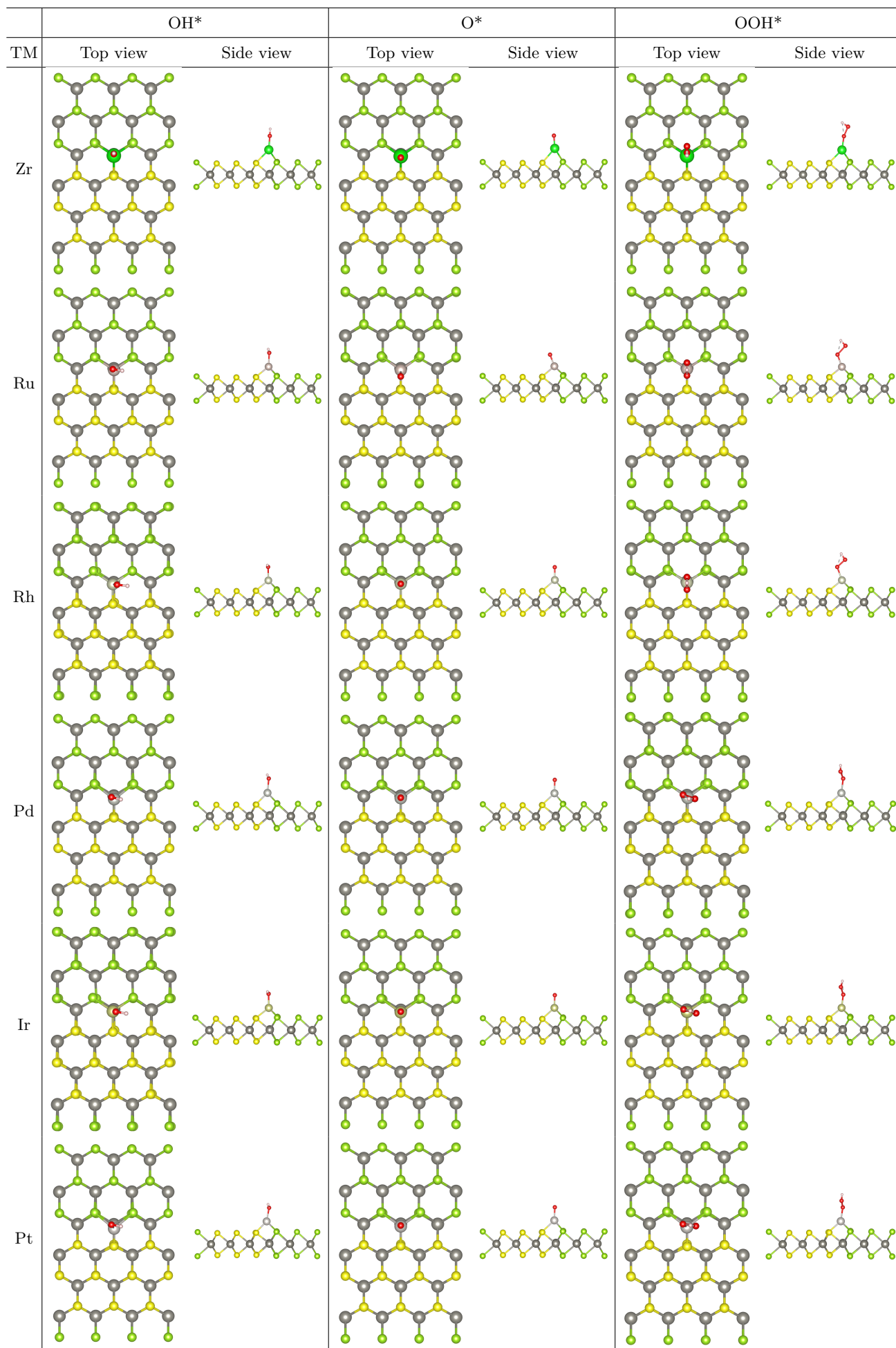


TABLE S4. The adsorption energies of *OH , *O and *OOH $\Delta G_{^*OH}$, $\Delta G_{^*O}$ and $\Delta G_{^*OOH}$.

Systems	$\Delta G_{^*OH}$ (eV)	$\Delta G_{^*O}$ (eV)	$\Delta G_{^*OOH}$ (eV)
Ti@WSe ₂ /WS ₂	-2.19	-2.04	1.41
V@WSe ₂ /WS ₂	-1.73	-1.43	1.95
Fe@WSe ₂ /WS ₂	-0.61	0.59	2.69
Co@WSe ₂ /WS ₂	-0.38	0.76	2.95
Ni@WSe ₂ /WS ₂	0.45	1.83	3.54
Zr@WSe ₂ /WS ₂	-2.42	-2.30	1.20
Ru@WSe ₂ /WS ₂	-0.20	0.09	2.81
Rh@WSe ₂ /WS ₂	0.38	1.34	3.39
Pd@WSe ₂ /WS ₂	1.20	2.84	4.18
Ir@WSe ₂ /WS ₂	-0.17	0.37	2.85
Pt@WSe ₂ /WS ₂	0.49	1.70	3.61

TABLE S5. The four reaction steps ΔG_1 , ΔG_2 , ΔG_3 and ΔG_4 of the TM@WSe₂/WS₂ system and the corresponding OER and ORR overpotentials.

Systems	ΔG_1 (eV)	ΔG_2 (eV)	ΔG_3 (eV)	ΔG_4 (eV)	η_{OER} (V)	η_{ORR} (V)
Ti@WSe ₂ /WS ₂	-2.19	0.15	3.45	3.51	2.28	3.42
V@WSe ₂ /WS ₂	-1.37	-0.06	3.38	2.97	2.15	2.60
Fe@WSe ₂ /WS ₂	-0.61	1.20	2.10	2.23	1.00	1.84
Co@WSe ₂ /WS ₂	-0.38	1.14	2.19	1.97	0.96	1.61
Ni@WSe ₂ /WS ₂	0.45	1.39	1.71	1.38	0.48	0.78
Zr@WSe ₂ /WS ₂	-2.42	0.12	3.51	3.72	2.49	3.65
Ru@WSe ₂ /WS ₂	-0.20	0.29	2.71	2.11	1.48	1.43
Rh@WSe ₂ /WS ₂	0.38	0.96	2.05	1.53	0.82	0.85
Pd@WSe ₂ /WS ₂	1.20	1.64	1.34	0.74	0.41	0.49
Ir@WSe ₂ /WS ₂	-0.17	0.55	2.47	2.07	1.24	1.40
Pt@WSe ₂ /WS ₂	0.49	1.20	1.91	1.31	0.68	0.74

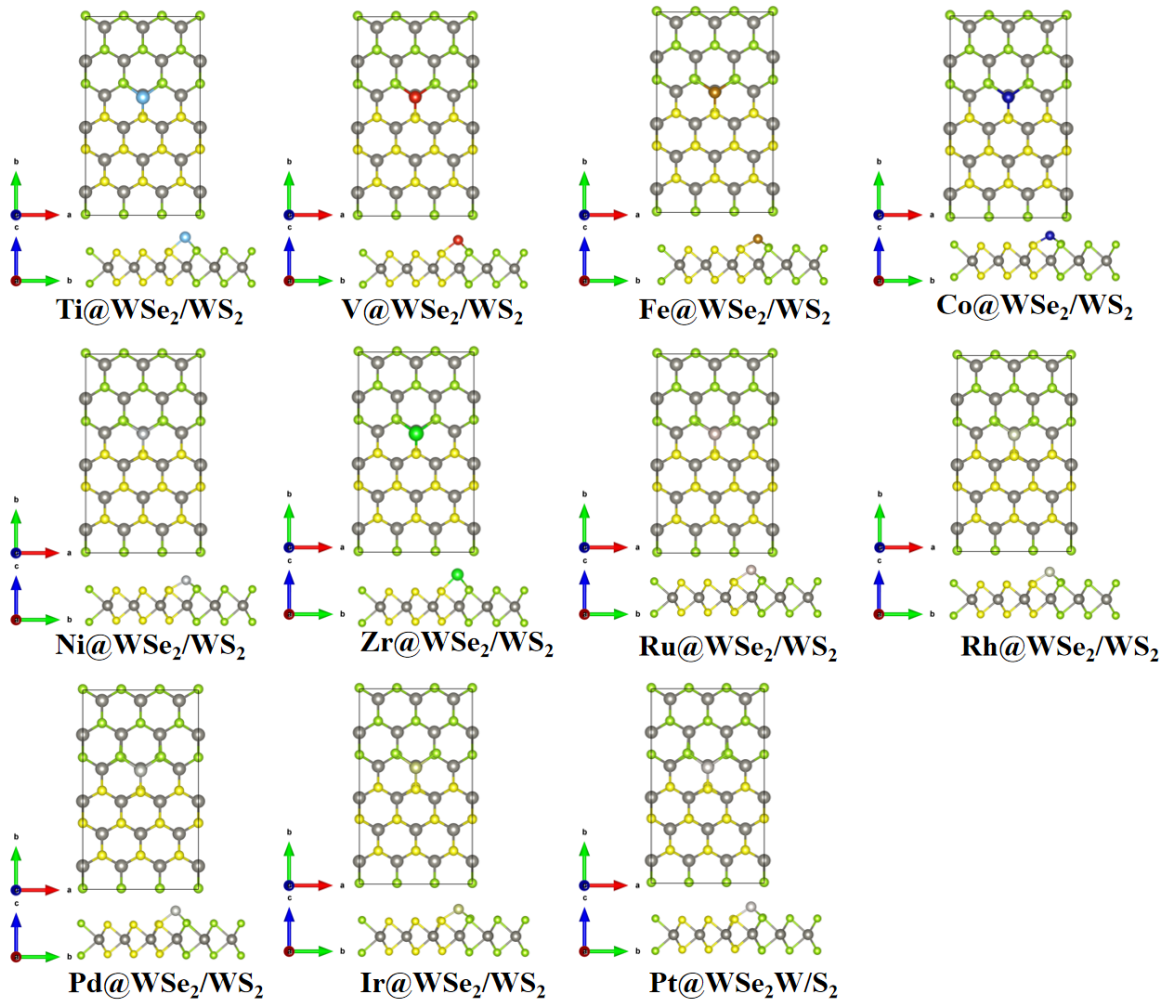


FIG. S1. The top and side views of the TM@WSe₂/WS₂ structures.

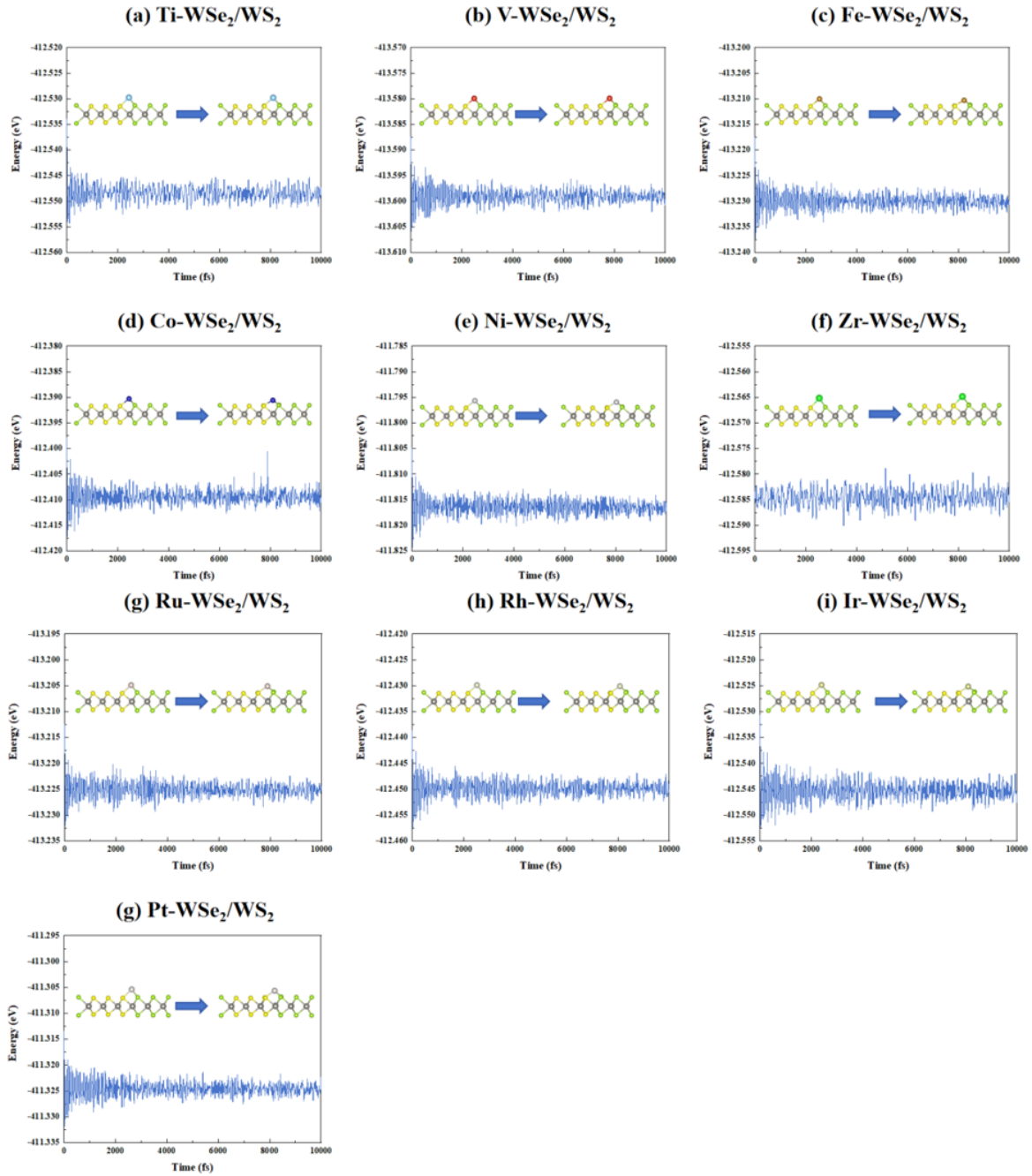


FIG. S2. Variations of the energies for Ti, V, Fe, Co, Ni, Zr, Ru, Rh, Ir, and Pt@WSe₂/WS₂ in a 10000 fs AIMD simulation at 300 K. The inset shows the atomic structure at the beginning and end of the AIMD simulation.

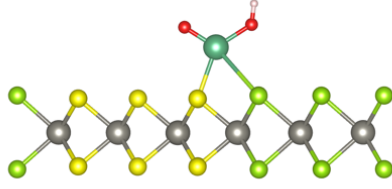
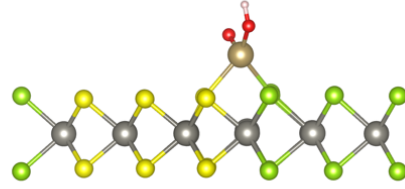
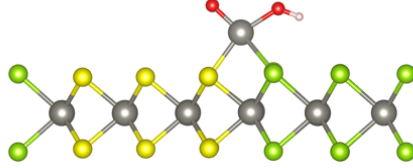
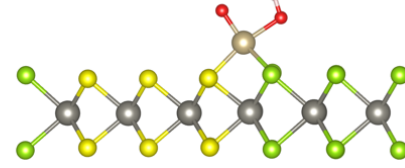
(a) Nb-WSe₂/WS₂(b) Ta-WSe₂/WS₂(c) W-WSe₂/WS₂(d) Os-WSe₂/WS₂

FIG. S3. Optimized structures of *OOH adsorbed on (a) Nb@WSe₂/WS₂, (b) Ta@WSe₂/WS₂, (c) W@WSe₂/WS₂, and (d) Os@WSe₂/WS₂ (stable *OOH adsorption geometries could not be obtained).

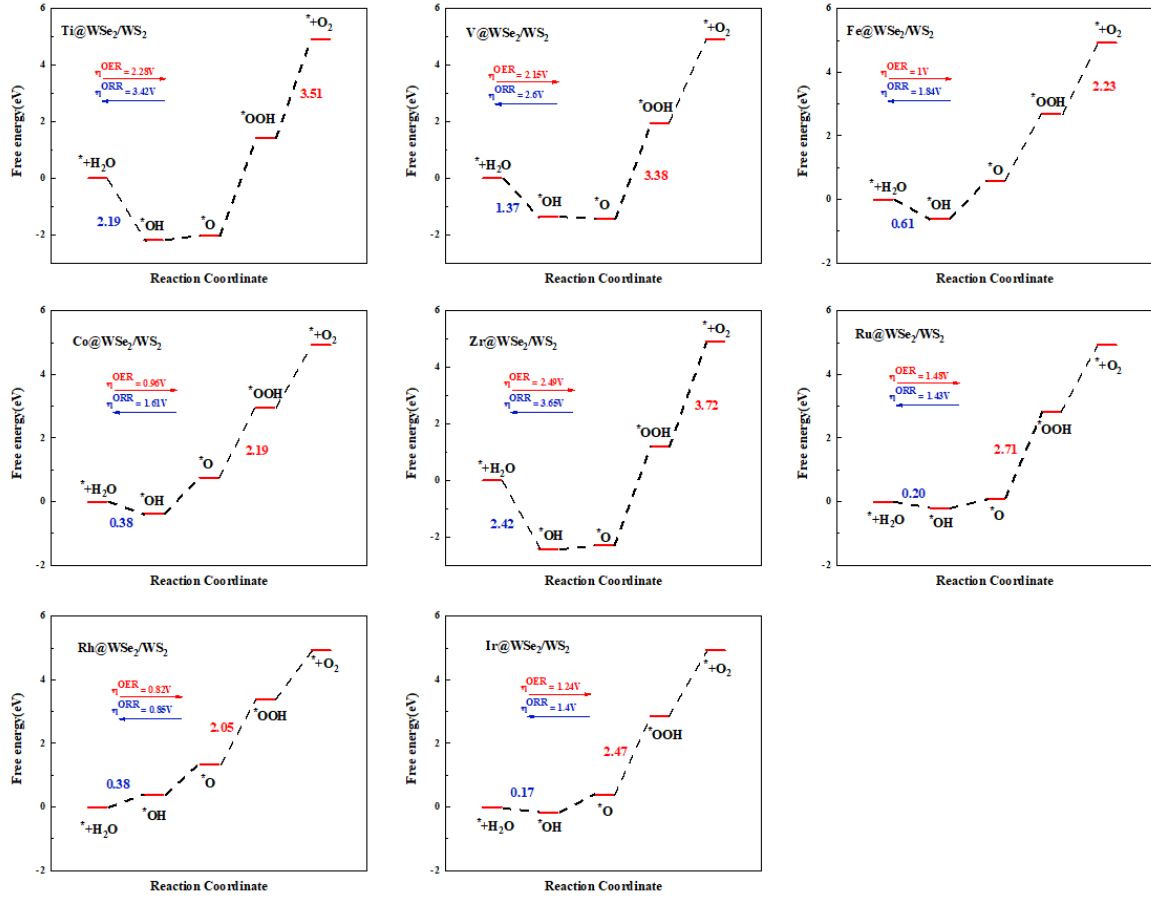


FIG. S4. Free energy diagrams for OER/ORR on TM@WSe₂/WS₂ (TM= Ti, V, Fe, Co, Zr, Ru, Rh, and Ir)

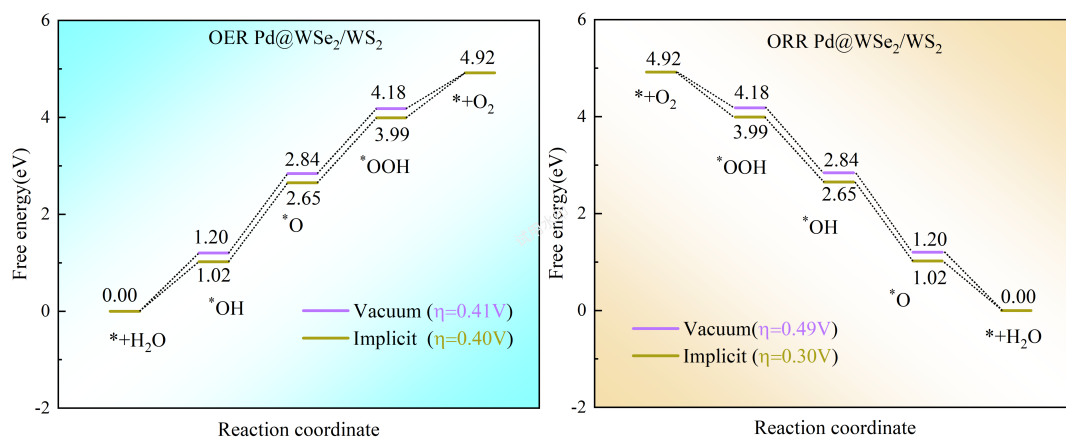


FIG. S5. -COHP between the TM (TM= Ti, V, Fe, Co, Zr, Ru, Rh, and Ir) and OH intermediates, where the Fermi energy level is set to zero.

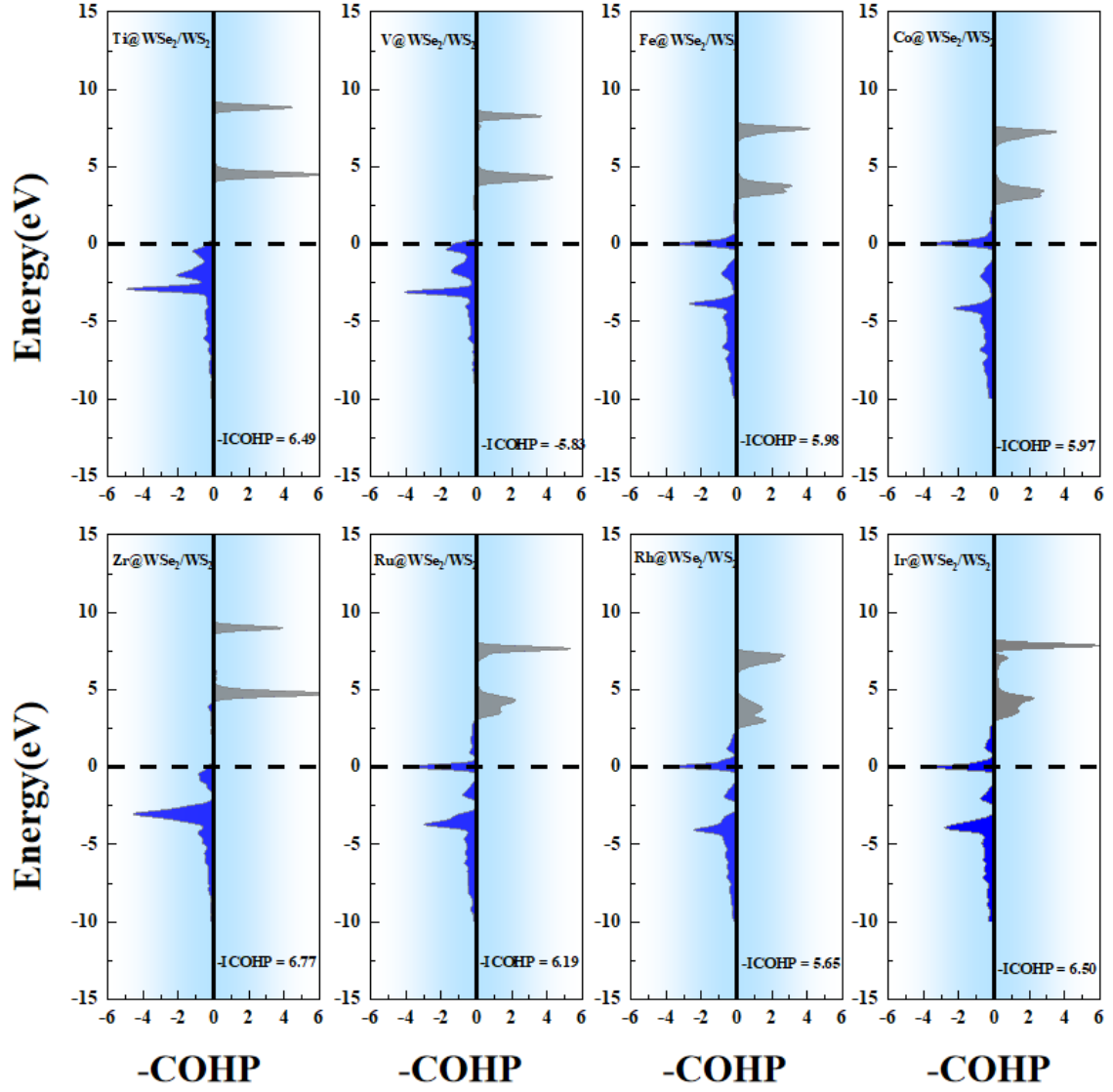


FIG. S6. The project density of states (PDOS) of d-orbital of transitional-metal. The red line denoted the d-band center of the metal atom.