**Supporting Information**

# Machine Learning-Aided Understanding of Structure-Activity Relation: A Case Study of $MoS_2$ Supported Metal-Nonmetal Pair for Hydrogen Evolution Reaction

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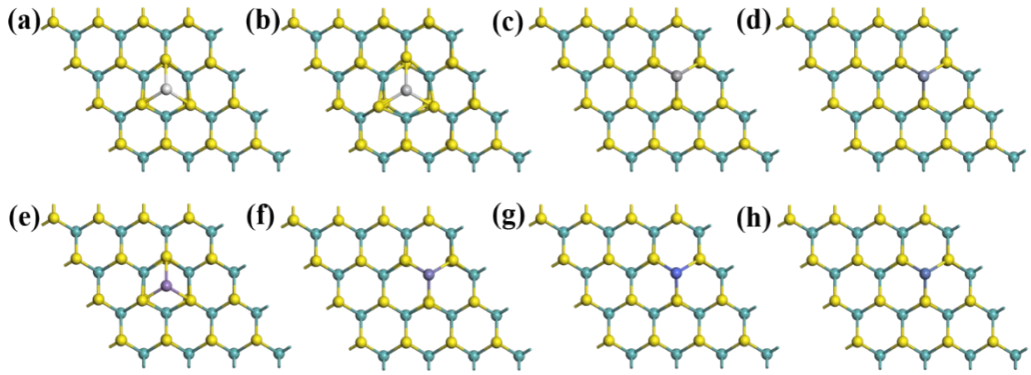


**Fig. S1** (a) Sc, (b) Ti, (c) V, (d) Cr, (e) Mn, (f) Fe, (g) Co, and (h) Ni supported on $MoS_2$ supercell structure optimization diagram.

| Sc simples | $BSc@MoS_2$ | $CSc@MoS_2$ | $NSc@MoS_2$ | $OSc@MoS_2$ | $PSc@MoS_2$ | $SeSc@MoS_2$ | $TeSc@MoS_2$ | $SSc@MoS_2$ |
|---|---|---|---|---|---|---|---|---|
| Top view | | | | | | | | |
| **Ti simples** | $BTi@MoS_2$ | $CTi@MoS_2$ | $NTi@MoS_2$ | $OTi@MoS_2$ | $PTi@MoS_2$ | $SeTi@MoS_2$ | $TeTi@MoS_2$ | $STi@MoS_2$ |
| Top view | | | | | | | | |
| **V simples** | $BV@MoS_2$ | $CV@MoS_2$ | $NV@MoS_2$ | $OV@MoS_2$ | $PV@MoS_2$ | $SeV@MoS_2$ | $TeV@MoS_2$ | $SV@MoS_2$ |
| Top view | | | | | | | | |
| **Cr simples** | $BCr@MoS_2$ | $CCr@MoS_2$ | $NCr@MoS_2$ | $OCr@MoS_2$ | $PCr@MoS_2$ | $SeCr@MoS_2$ | $TeCr@MoS_2$ | $SCr@MoS_2$ |
| Top view | | | | | | | | |
| **Mn simples** | $BMn@MoS_2$ | $CMn@MoS_2$ | $NMn@MoS_2$ | $OMn@MoS_2$ | $PMn@MoS_2$ | $SeMn@MoS_2$ | $TeMn@MoS_2$ | $SMn@MoS_2$ |
| Top view | | | | | | | | |
| **Fe simples** | $BFe@MoS_2$ | $CFe@MoS_2$ | $NFe@MoS_2$ | $OFe@MoS_2$ | $PFe@MoS_2$ | $SeFe@MoS_2$ | $TeFe@MoS_2$ | $SFe@MoS_2$ |
| Top view | | | | | | | | |
| **Co simples** | $BCo@MoS_2$ | $CCo@MoS_2$ | $NCo@MoS_2$ | $OCo@MoS_2$ | $PCo@MoS_2$ | $SeCo@MoS_2$ | $TeCo@MoS_2$ | $SCo@MoS_2$ |
| Top view | | | | | | | | |
| **Ni simples** | $BNi@MoS_2$ | $CNi@MoS_2$ | $NNi@MoS_2$ | $ONi@MoS_2$ | $PNi@MoS_2$ | $SeNi@MoS_2$ | $TeNi@MoS_2$ | $SNi@MoS_2$ |
| Top view | | | | | | | | |

**Fig. S2** Structure optimization of $XTM@MoS_2$ supercell.

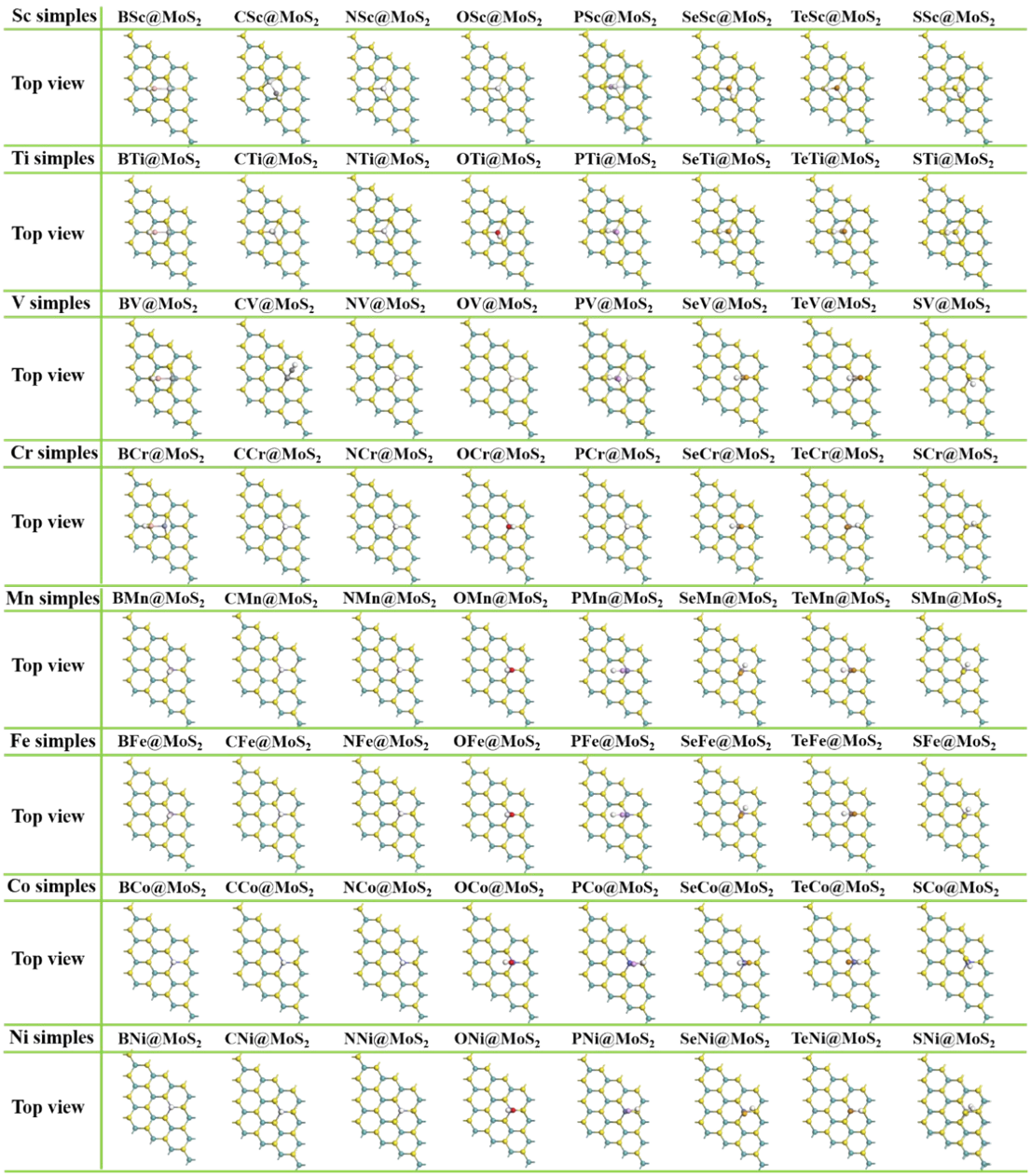


**Fig. S3** The optimized structure of XTM@$MoS_2$ supercell after adsorbing H.

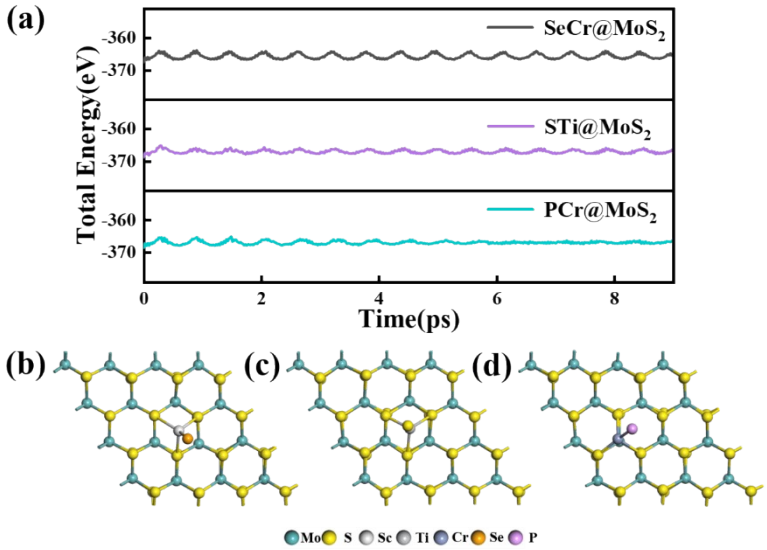


**Fig. S4** (a) The variations of energy during the 9 ps AIMD simulation. (b-d) The final structures of SeSc@$MoS_2$, STi@$MoS_2$, and PCr@$MoS_2$ at 300 K after the simulation.

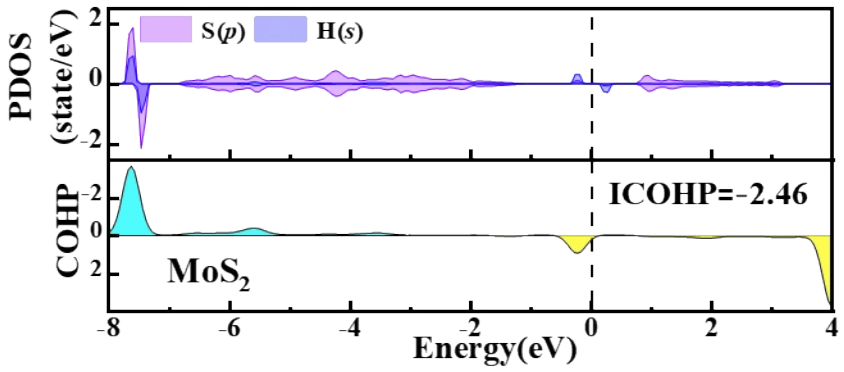


**Fig. S5** The Partial density of states (PDOS), crystal orbital Hamilton populations (COHPs), and integrated COHP (ICOHP) for H adsorption on $MoS_2$.

**Table S1** The binding energies ($E_{b\text{-}TM}$s) of these TM@$MoS_2$s (TM=Sc-Ni).

| SYS | $T_S$ (eV) | $T_M$ (eV) | $H_S$ (eV) | SYS | $T_S$ (eV) | $T_M$ (eV) | $H_S$ (eV) |
|---|---|---|---|---|---|---|---|
| **Sc** | -2.316 | -2.350 | **-2.584** | **Mn** | -0.346 | -1.193 | **-1.220** |
| **Ti** | | -2.799 | **-2.859** | **Fe** | -0.724 | -2.351 | **-2.255** |
| **V** | | **-2.527** | -2.038 | **Co** | -1.435 | **-3.078** | -2.759 |
| **Cr** | -0.607 | **-1.204** | -0.857 | **Ni** | -2.092 | **-3.778** | -3.467 |