Support Information

Double Transition Metal Carbides MXenes (D-MXenes) as Promising Electrocatalysts for Hydrogen Reduction Reaction: *Ab initio* Calculations

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The surface Pourbaix diagram of the Mo_2NbC_2 is constructed by plotting the thermodynamically most stable surface state under relevant U_{SHE} and pH, as shown in Figure S1. We have been investigated the Pourbaix diagram of four different MXenes with adsorbates: partially terminated 1/8 ML OH*, fully terminated 1 ML OH*, a mixed termination consisting of 1/2 ML OH* and 1/2 ML O*, and fully terminated 1 ML O*. The U_{SHE} is the applied voltage on the electrode referenced to the standard hydrogen electrode, and theoretically is defined in solution [pH = 0, $p(H_2)$ = 1 bar]. Bare Mo_2NbC_2 is found to be stable at potentials lower than -1.13 V in an acidic solution (pH = 0).

With increasing U_{SHE}, more OH* are adsorbed on the Mo_2NbC_2 surface. U_{SHE} of the Mo_2NbC_2 surface with full OH* (l ML) termination [$Mo_2NbC_2(OH)_2$] reaches to

−0.92 V. When the U_{SHE} continually increases, water oxidation begins, leading to increasing coverage of hydroxyl and oxygen on the MXenes surface. All terminated OH* will be oxidized if the U_{SHE} continually increases, and then the stable O* terminated Mo_2NbC_2 MXenes ($Mo_2NbC_2O_2$) are formed. At the acidic condition (pH = 0), the lowest potential values for the fully O* terminated Mo_2NbC_2 (1 ML O*) are −0.435 V.

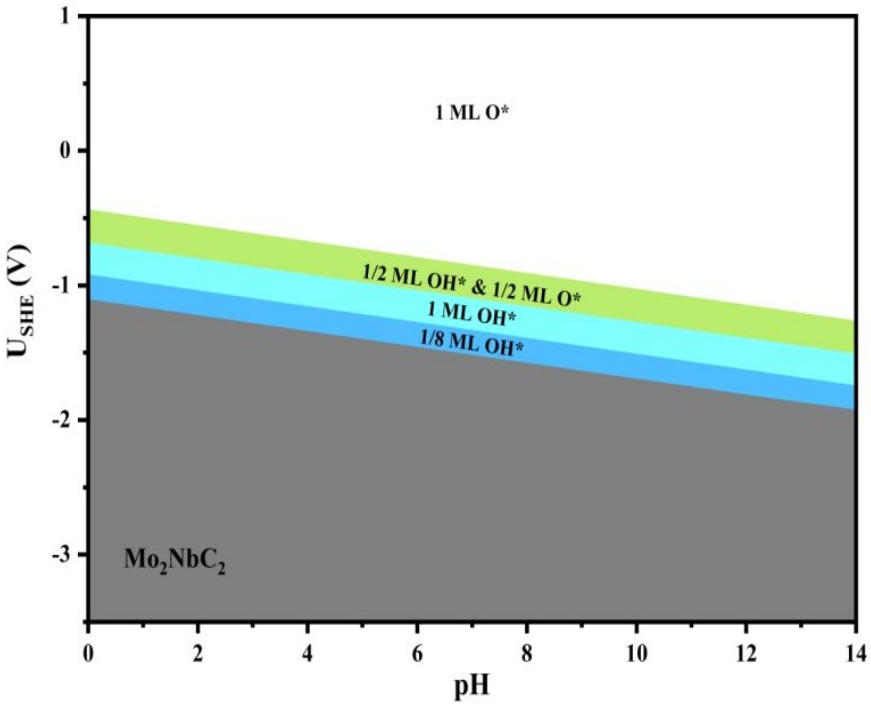

Figure S1. Calculated surface Pourbaix diagrams of Mo_2NbC_2 showing the most stable terminations on the surface at specific potentials and pH values.

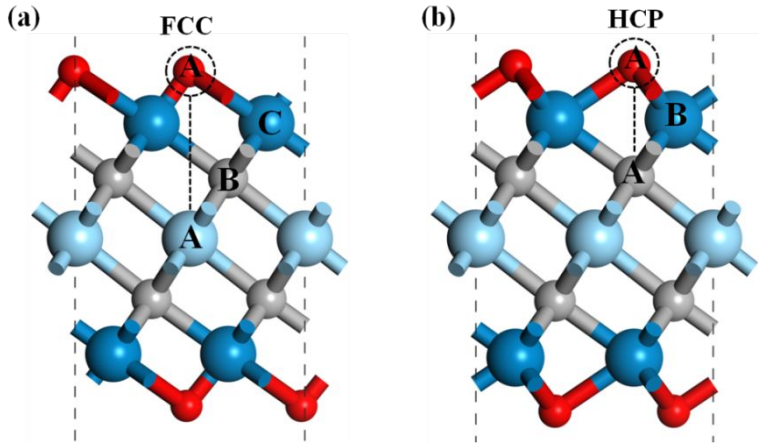

Figure S2. Side view of (a) FCC and (b) HCP sites for O-terminal double MXenes ($M'_2M''C_2O_2$).