# Supplementary Information

## First-principles study of 2H-$Mo_2C$-based MXene under biaxial strain implemented as Li-battery anode.

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**Table S1** Comparison of adsorption energy ($E_{ad}$), bond population of Mo-Li (for $Mo_2C$) or O-Li (for $Mo_2CO_2$), and adsorption height (H) between an adsorbed Li-ion at S3 site and an adsorbed Li layer.

| Configurations of Li-ion | | $E_{ad}$ (eV) | | | Bond population | | | H (Å) | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | -4% | 0% | 4% | -4% | 0% | 4% | -4% | 0% | 4% |
| **$Mo_2C$** | Li-ion@S3 | -0.88 | -0.96 | -1.31 | -0.42 | -0.41 | -0.40 | 2.280 | 2.262 | 2.217 |
| | Li layer | -0.49 | -0.50 | -0.68 | -0.72 | -0.61 | -0.56 | 2.285 | 2.261 | 2.216 |
| **$Mo_2CO_2$** | Li-ion@S3 | -3.15 | -3.13 | -3.08 | -0.13 | -0.14 | -0.15 | 1.151 | 1.131 | 1.089 |
| | Li layer | -0.87 | -0.95 | -1.07 | -0.12 | -0.11 | -0.10 | 1.057 | 0.952 | 0.819 |

To address the opposite trend of strain dependent $E_{ed}$ of these two MXenes, the different interactions between Mo-Li for $Mo_2C$ and O-Li for $Mo_2CO_2$ are considered. According to Table 1S, the more extended adsorption height (H) of an adsorbed Li-ion on $Mo_2C$ reflects weaker interactions than its counterpart. This is also supported by less negative adsorption energy and a more negative bond population (corresponding to higher antibonding levels). Interestingly, we have observed that the bond population is less negative (-0.42 to -0.40) for $Mo_2C$ from -4% to 4%; meanwhile, it is more negative (-0.13 to -0.15) for $Mo_2CO_2$. The stronger repulsion of enhancing antibonding of $Mo_2CO_2$ (also increase energy of the system) might be a reason why the trend of an adsorbed Li-ion on $Mo_2CO_2$ is opposite to that of $Mo_2C$.

Moreover, one can observe that the tendency of $E_{ad}$ of $Mo_2CO_2$ when considering adsorbed Li layer, flips to be the same trend with that of $Mo_2C$. According to Table 1S, the H of an adsorbed Li layer (H=1.131 Å at S3 and ε=0%) is significantly lower than that of an adsorbed Li-ion (H=0.952 Å at ε=0%). This indicates that it has stronger interaction compared with another. To clarify the change of tendency of adsorption energy when decoration the Li layer on $Mo_2CO_2$, the reduction of H respecting that of an adsorbed Li-ion to be 0.094 Å, 0.179 Å, and 0.270 Å for strains of -4%, 0%, and 4%, respectively. It also corresponds to the reduction of antibonding (also decreasing energy of the system) of $Mo_2CO_2$ from compression to tension. Therefore, this finding supports that the $E_{ad}$ of the adsorbed Li layers increases (more negative) with the arial expansion of $Mo_2CO_2$ monolayer.

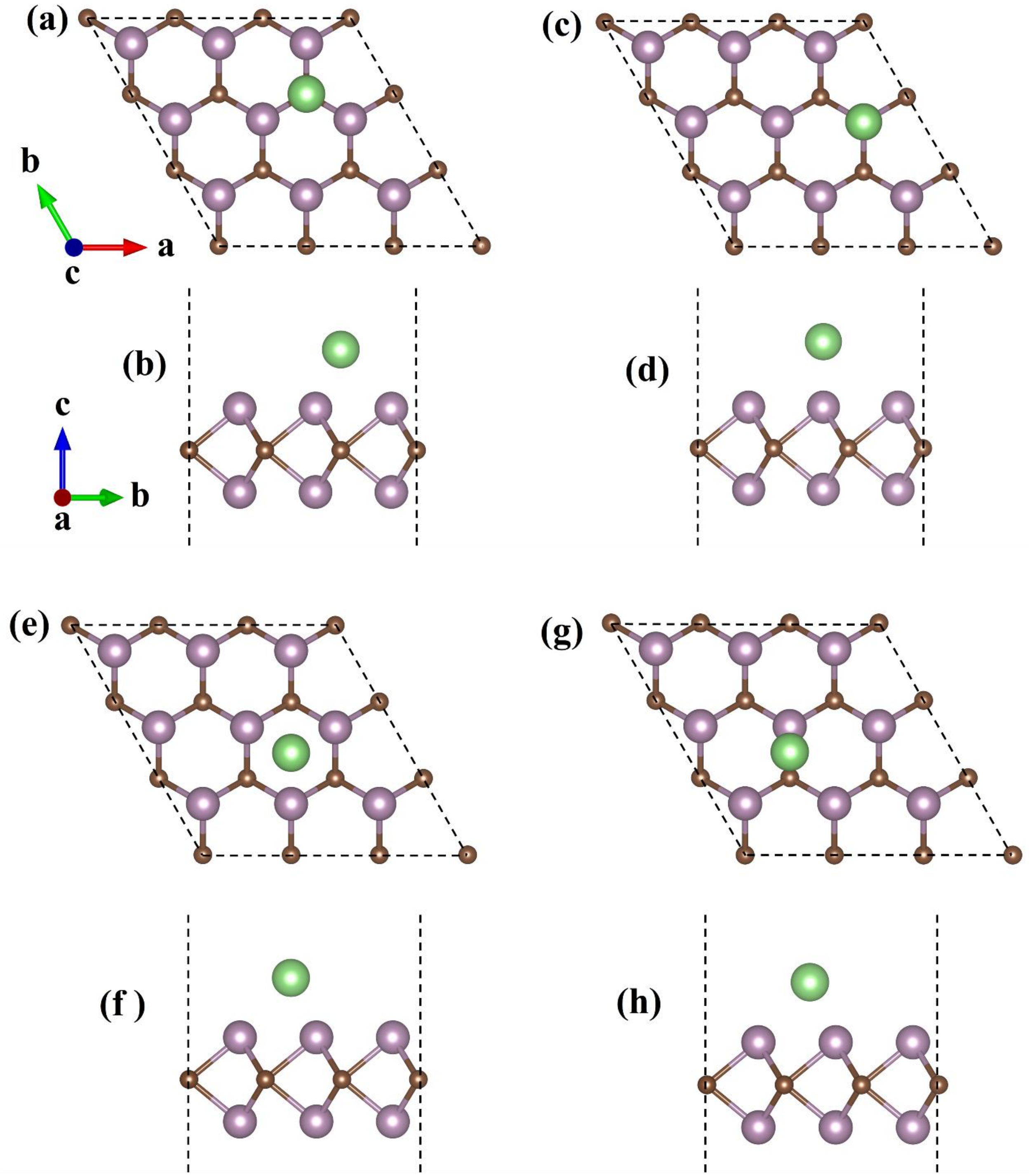

**Fig. 1S** Top and side views of the Li positions on $Mo_2C$ at the different sites consisting of S1 (a, b), S2 (c, d), S3 (e, f), and S4 (g, h). The brown, purple, and green balls represent C, Mo, and Li atoms, respectively.

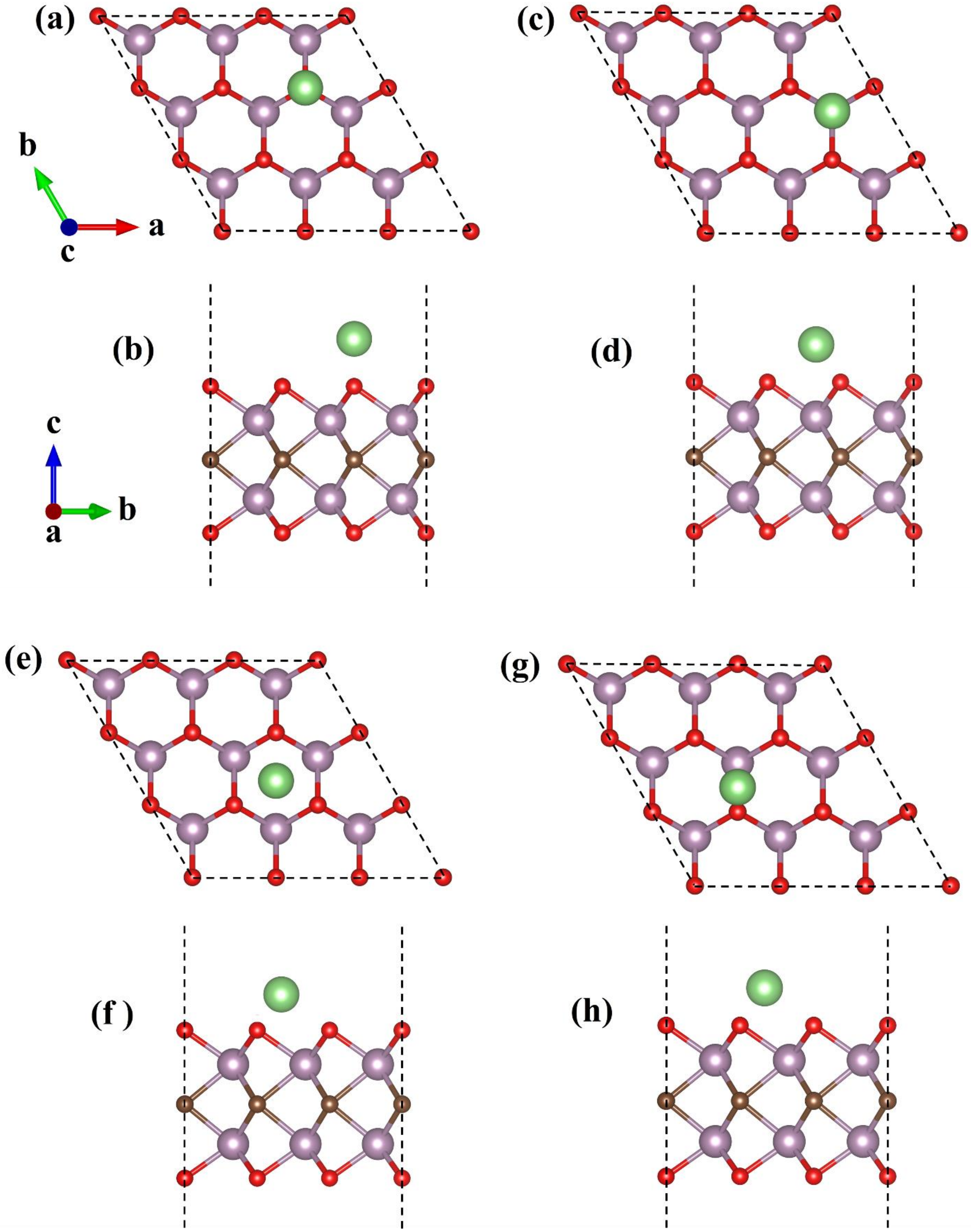

**Fig. 2S** Top and side views of the Li positions on $Mo_2CO_2$ at the different sites consisting of S1 (a, b), S2 (c, d), S3 (e, f), and S4 (g, h). The brown, purple, red and green balls represent C, Mo, O, and Li atoms, respectively.

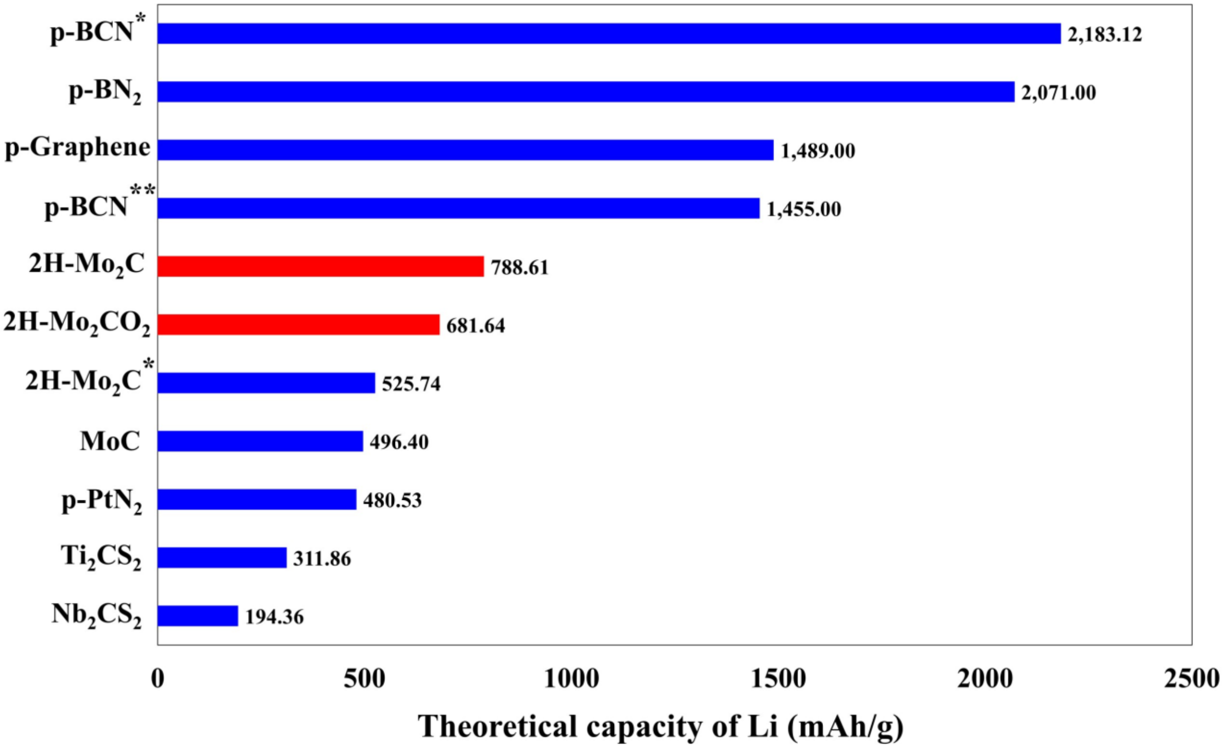

**Fig. 3S** The theoretical capacities of some promising anode materials for Li-ion batteries with corresponding references including p-BNC*[1], p-BN$_2$[2], p-Graphene[3], p-BNC**[4], 2H-Mo$_2$C*[5], p-PtN$_2$[6], MoC[7], Ti$_2$CS$_2$[8], and Nb$_2$CS$_2$[9].

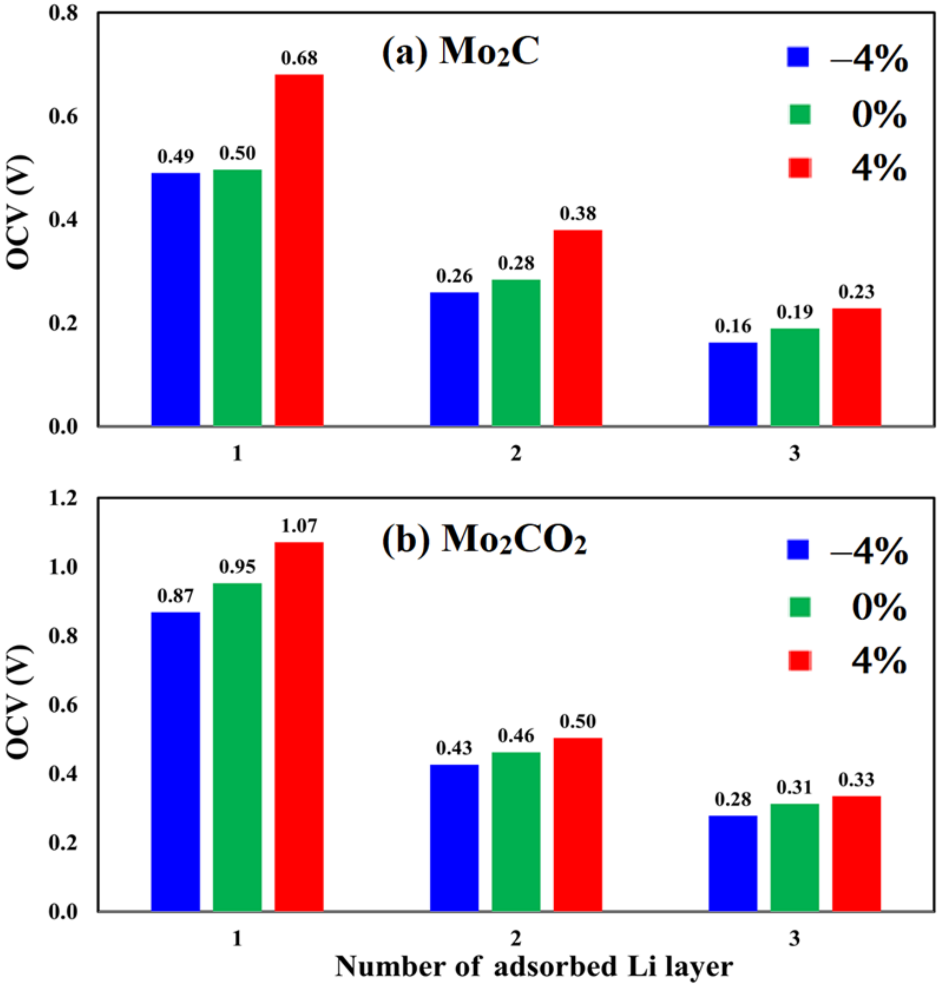

**Fig. 4S** The OCV of number of adsorbed Li layers on (a) $Mo_2C$ and (b) $Mo_2CO_2$ at certain biaxial strains at -4%, 0%, and 4%.

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