# Electronic Supplementary Information

# Bipolar magnetic semiconductors among the intermediate states during the conversion from $Sc_2C(OH)_2$ to $Sc_2CO_2$ MXene

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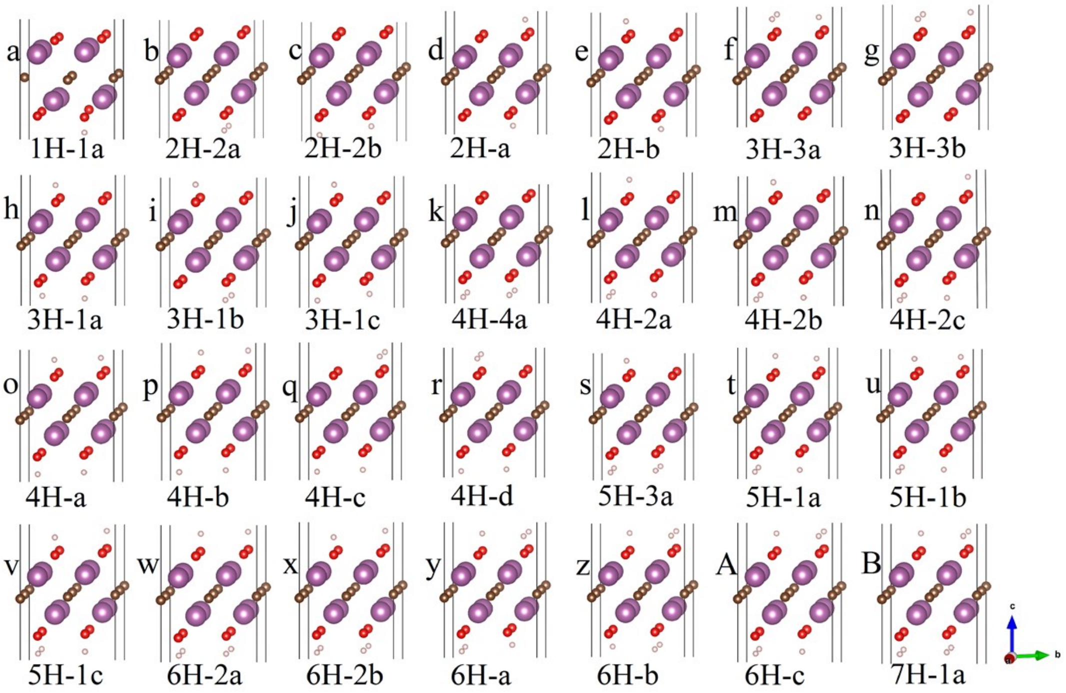


**Figure S1** All the possible initial configurations for intermediate states by removing hydrogen atoms from the $Sc_2C(OH)_2$ MXene based on $2\times2\times1$ supercells.

**Figure S2** All the possible initial configurations for intermediate states by adding hydrogen atoms on the $Sc_2CO_2$ MXene based on 2×2×1 supercells.

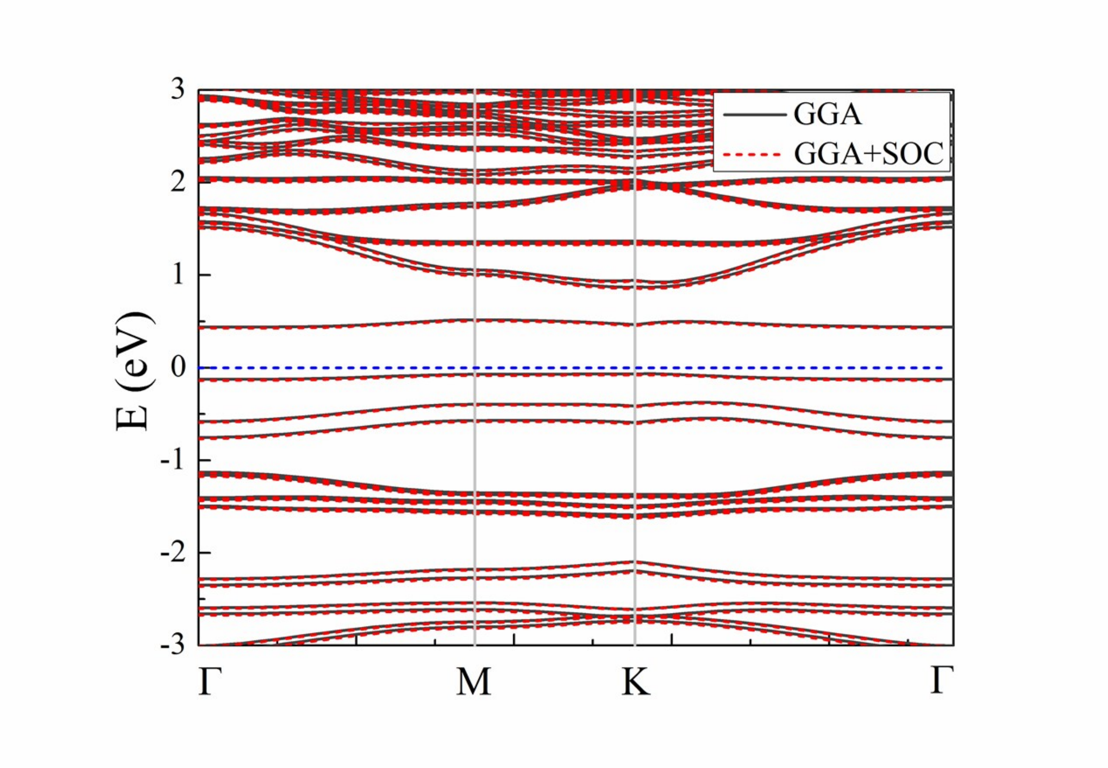


**Figure S3** The electronic energy band of the 1H-1a state with/without considering SOC effect based on the GGA functional.

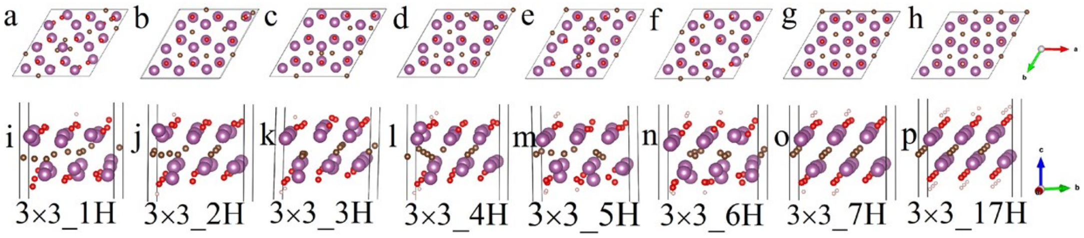


**Figure S4** The structures of those rearranged intermediate states with their hydrogen number from one to six, combined with the 3×3_7H and 3×3_17H states based on 3 ×3×1 supercells. The first and second rows present the top-views and side-views of these configurations, respectively.

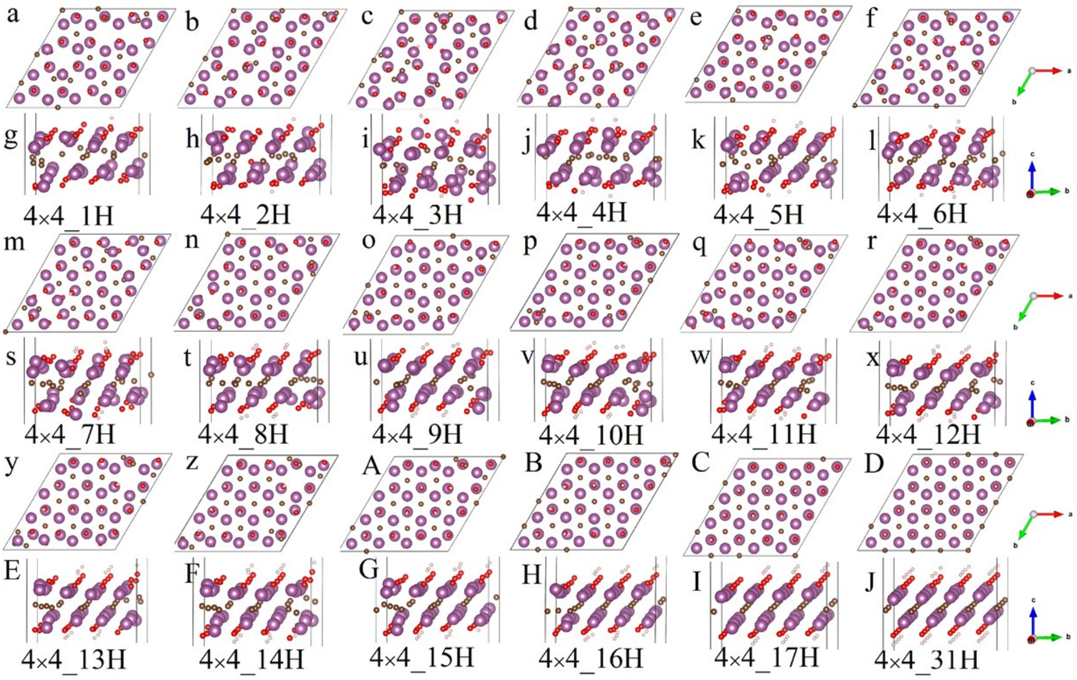


**Figure S5** The structures of those rearranged intermediate states with their hydrogen number from one to sixteen, combined with the 4×4_17H and 4×4_31H states based on 4×4×1 supercells. The odd and even rows present the top-views and side-views of these configurations, respectively.

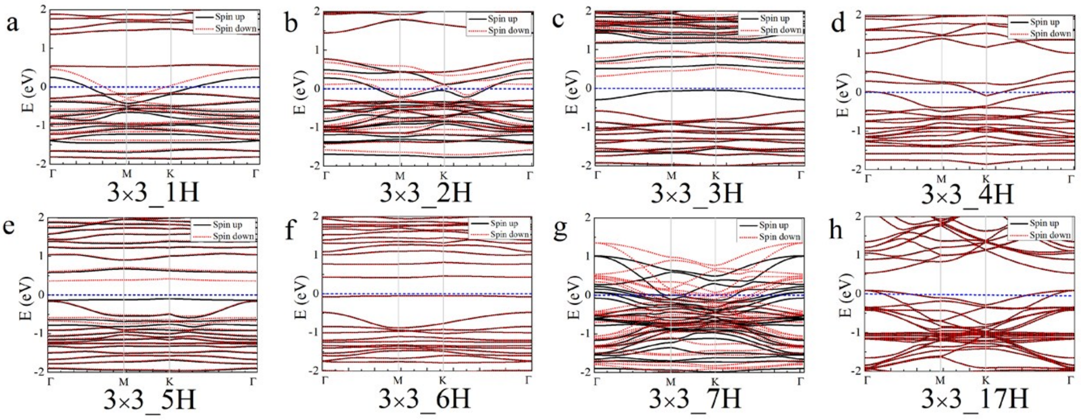
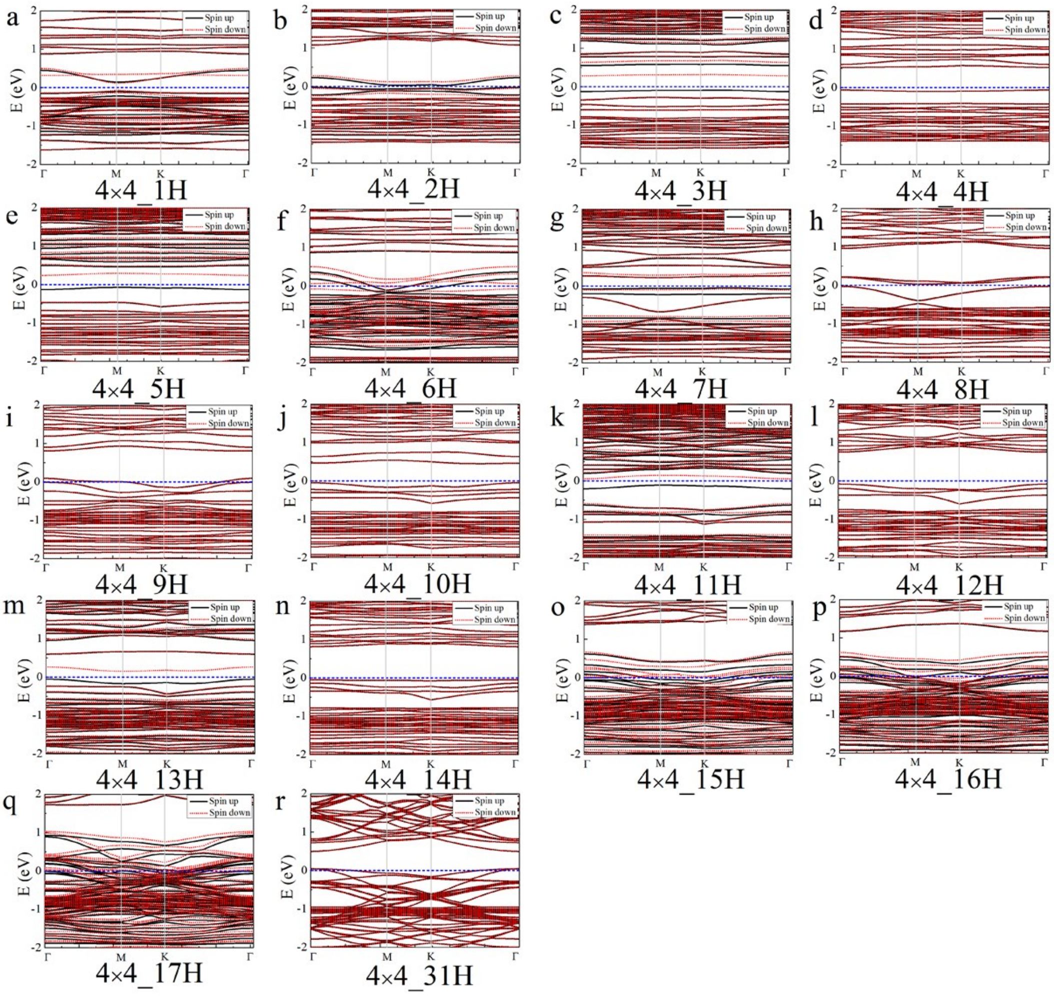


**Figure S6** The electronic energy bands of the those rearranged intermediate states with their hydrogen number from one to six, combined with the 3×3_7H and 3×3_17H states based on 3×3×1 supercells.



4×4_1H
4×4_2H
4×4_3H
4×4_4H
4×4_5H
4×4_6H
4×4_7H
4×4_8H
4×4_9H
4×4_10H
4×4_11H
4×4_12H
4×4_13H
4×4_14H
4×4_15H
4×4_16H
4×4_17H
4×4_31H

**Figure S7** The electronic energy bands of those rearranged intermediate states with their hydrogen number from one to sixteen, combined with the 4×4_17H and 4×4_31H states based on 4×4×1 supercells.

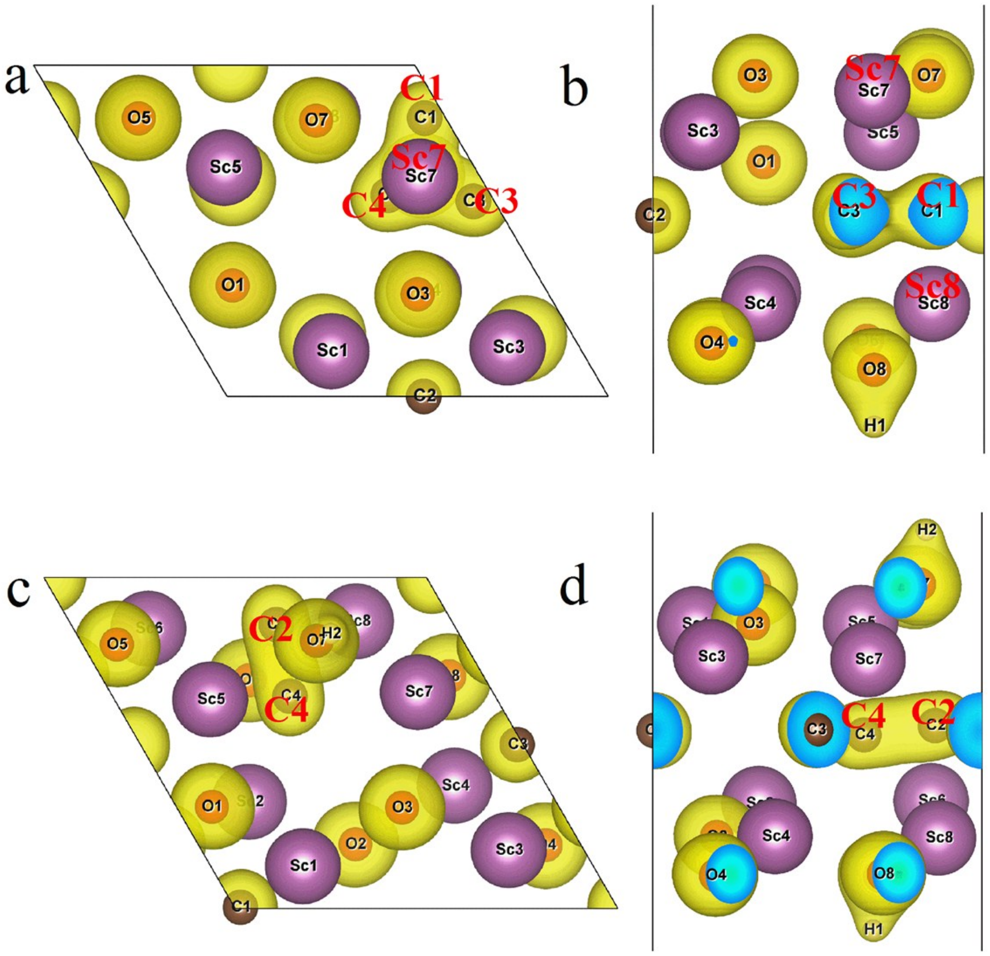


**Figure S8** The electron density distributions for 1H-1a and 2H-a states. (a) and (b) are the top-view and side-view for the 1H-1a state; (c) and (d) are the top-view and side-view for 2H-a state, respectively.

**Table S1** The atomic charges for the 1H-1a and 2H-a states

| **Atom** | **Atomic Charge** | |
|---|---|---|
| | **1H-1a** | **2H-a** |
| C1 | -0.819 | -1.726 |
| C2 | -1.781 | -0.854 |
| C3 | -0.852 | -1.726 |
| C4 | **-0.858** | **-0.752** |
| Sc1 | 1.642 | 1.627 |
| Sc2 | 1.692 | 1.638 |
| Sc3 | 1.621 | 1.638 |
| Sc4 | 1.645 | 1.627 |
| **Sc5** | 1.646 | **1.645** |
| Sc6 | 1.612 | 1.643 |
| **Sc7** | **1.607** | 1.643 |
| Sc8 | 1.638 | 1.645 |
| O1 | -1.270 | -1.121 |

| | | |
|---|---|---|
| O2 | -1.136 | -1.188 |
| O3 | -1.148 | -1.188 |
| O4 | -1.132 | -1.121 |
| O5 | -1.131 | -1.121 |
| O6 | -1.134 | -1.259 |
| O7 | -1.152 | -1.260 |
| O8 | -1.279 | -1.121 |
| H1 | 0.614 | 0.589 |
| H2 | - | 0.590 |

## Discussions on atomic charges

From Figure S8 and Table S1, we find that three carbon atoms labeled as C1, C3 and C4 move together in the 1H-1a state, and strong p-d hybridization is formed between these carbon atoms and neighboring Sc7 and Sc8 atoms as discussed in our paper. However, in the 2H-a state, a strong covalent bond is formed between the C2 and C4 atoms according to the shape of the electron density distribution and the lower atomic charge of C4 (denoted in bold). Consequently, the p-d hybridization is weaker in 2H-a state, which is also reflected by the difference in atomic charges of scandium atoms such as Sc5 in 2H-a vs. Sc7 in 1H-1a denoted in bold in the table. Furthermore, through investigating the electron density distributions and atomic charges of all the intermediate states based on $3\times3\times1$ supercells, we also determine that the rearranged configurations with odd hydrogen atoms generally present stronger p-d hybridizations compared to those with even hydrogen atoms. The reasons can be understood as follows: the central carbon atoms are prone to slightly move to one side in the configurations with odd hydrogen atoms due to the non-uniform interactions between two sides, thus form strong local hybridization with their nearest scandium atoms. However, in those states with even hydrogen atoms, the numbers of hydrogen atoms on opposite sides are equivalent, and thus the carbon atoms are readily to form much stronger covalent bond to each other in the central layers.

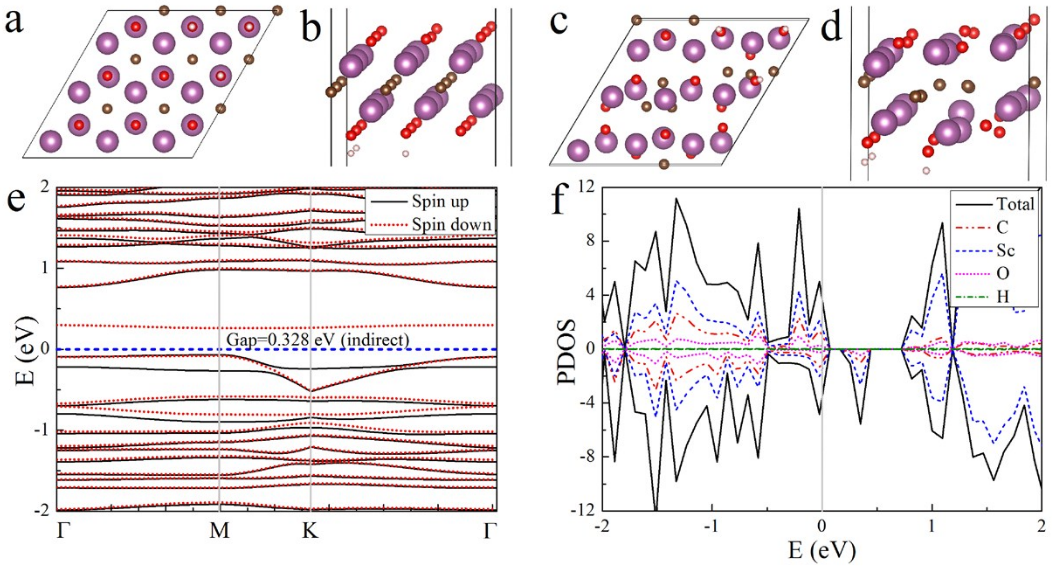


**Figure S9** (a) and (b) are respectively the top-view and side-view of the metastable configuration which with three hydrogen atoms at one side based on the $3\times3\times1$ supercell. (c) and (d) present the optimized configuration. (e) and (f) are the electronic energy band and projected density of states (PDOS), respectively.

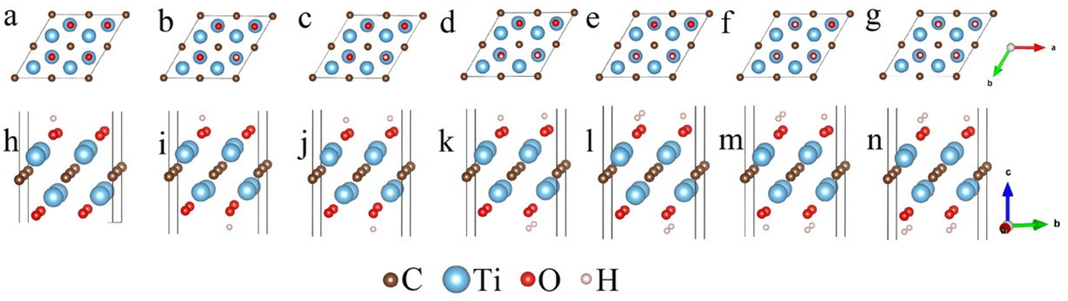


**Figure S10** The stable intermediate states during the structure conversion from $Ti_2C(OH)_2$ to $Ti_2CO_2$ MXene on the basis of $2\times2\times1$ supercells.

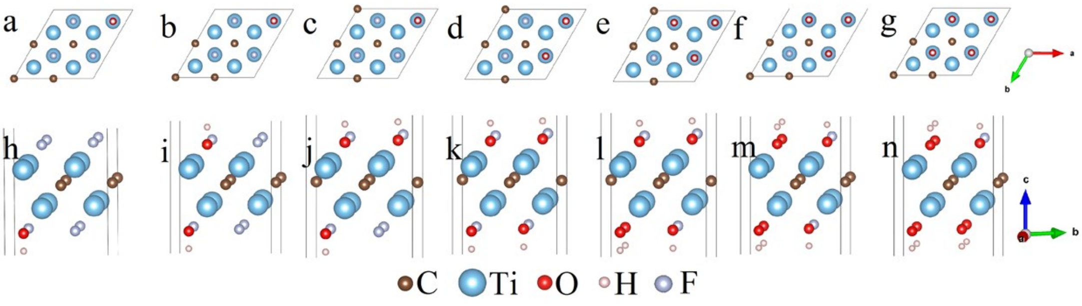


**Figure S11** The stable intermediate states during the structure conversion from $Ti_2CF_2$ to $Ti_2C(OH)_2$ MXene on the basis of $2\times2\times1$ supercells.