Enhancing hydrogen evolution reaction on O-terminated Ti_3C_2 MXene via dual non-metal doping: a first-principles study

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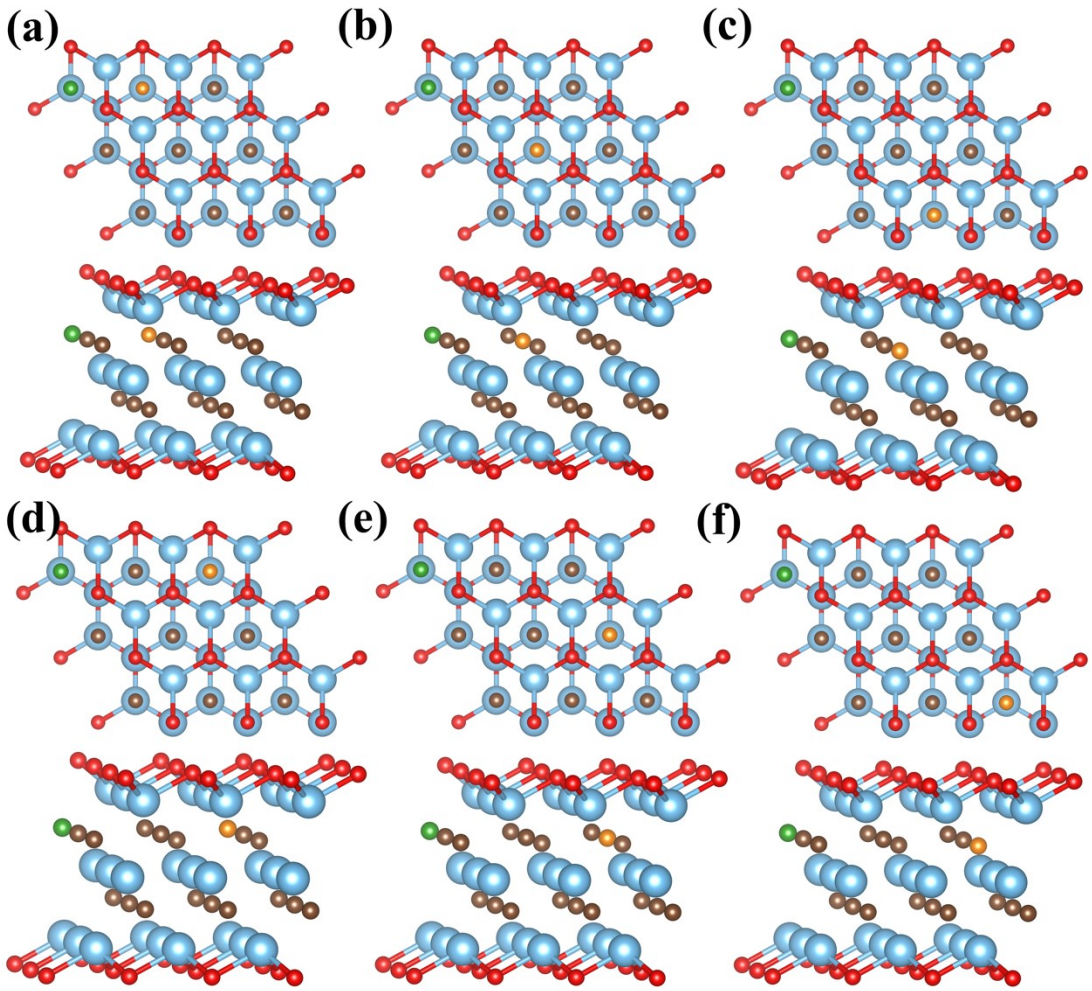

Figure S1. Top and side views of all possible doping configurations structures of intra-$Ti_3C_2O_2$.

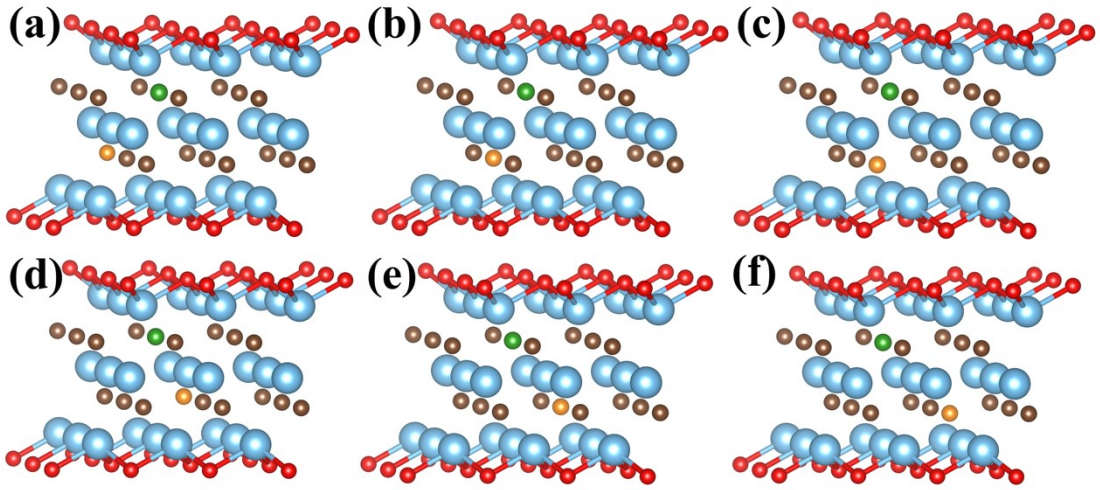

Figure S2. Top and side views of all possible doping configurations structures of inter-$Ti_3C_2O_2$.

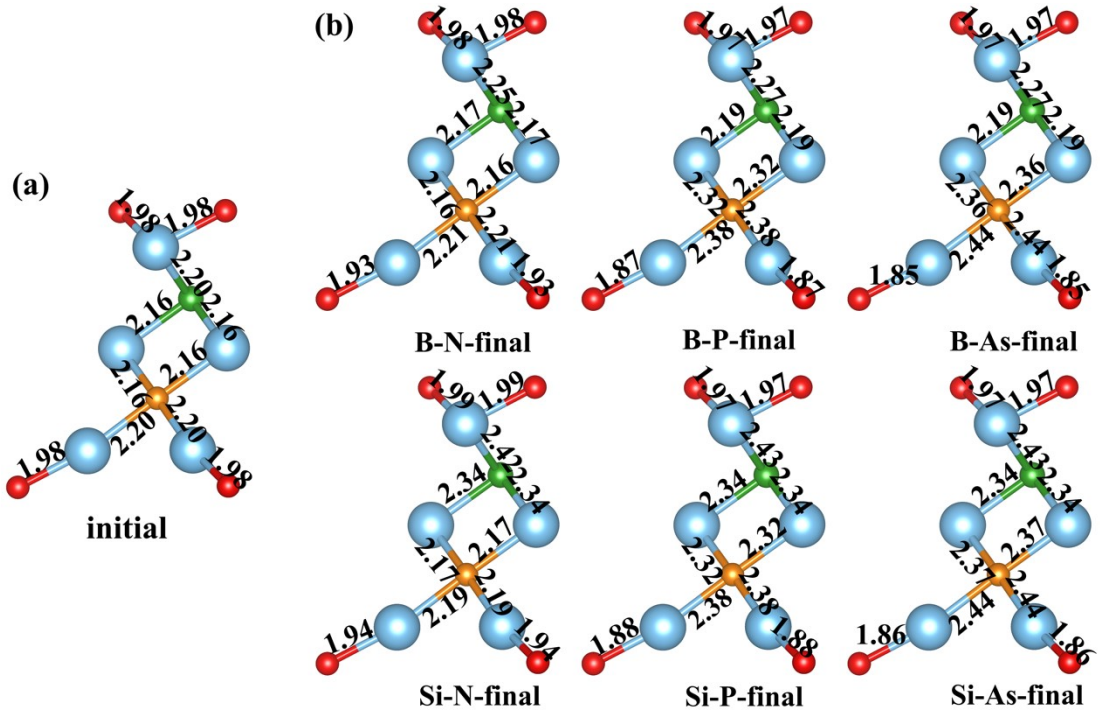

Figure S3. The changes in the bond lengths of surrounding atoms (a) before and (b) after the optimization of inter-layer doping for the X-Y pairs.

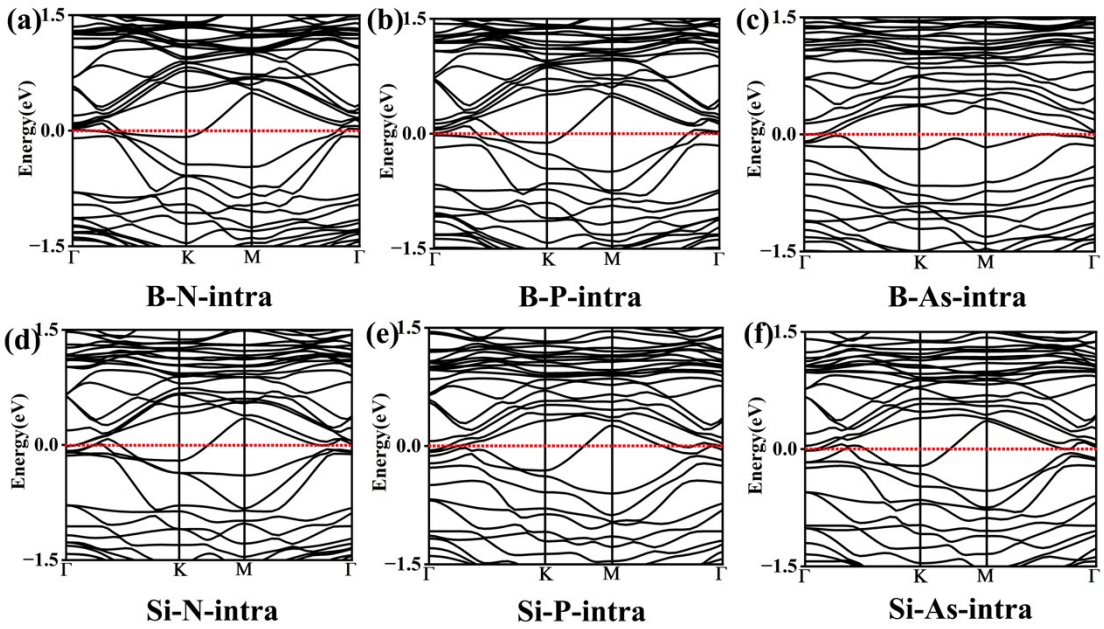

Figure S4. The band structure of (a-c) B-N/B-P/B-As-intra doped $Ti_3C_2O_2$ and (d-f) Si-N/Si-P/Si-As-intra doped $Ti_3C_2O_2$ as obtained from the *p*(3×3) unit cell.

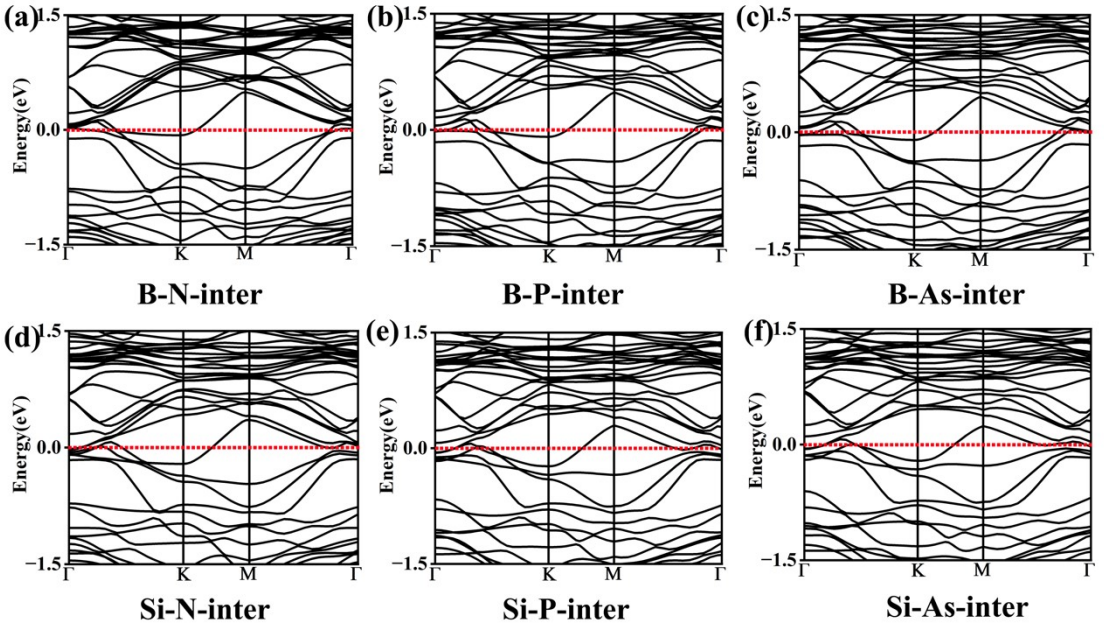

Figure S5. The band structure of (a-c) B-N/B-P/B-As-inter doped $Ti_3C_2O_2$ and (d-f) Si-N/Si-P/Si-As-inter doped $Ti_3C_2O_2$ as obtained from the *p*(3×3) unit cell.

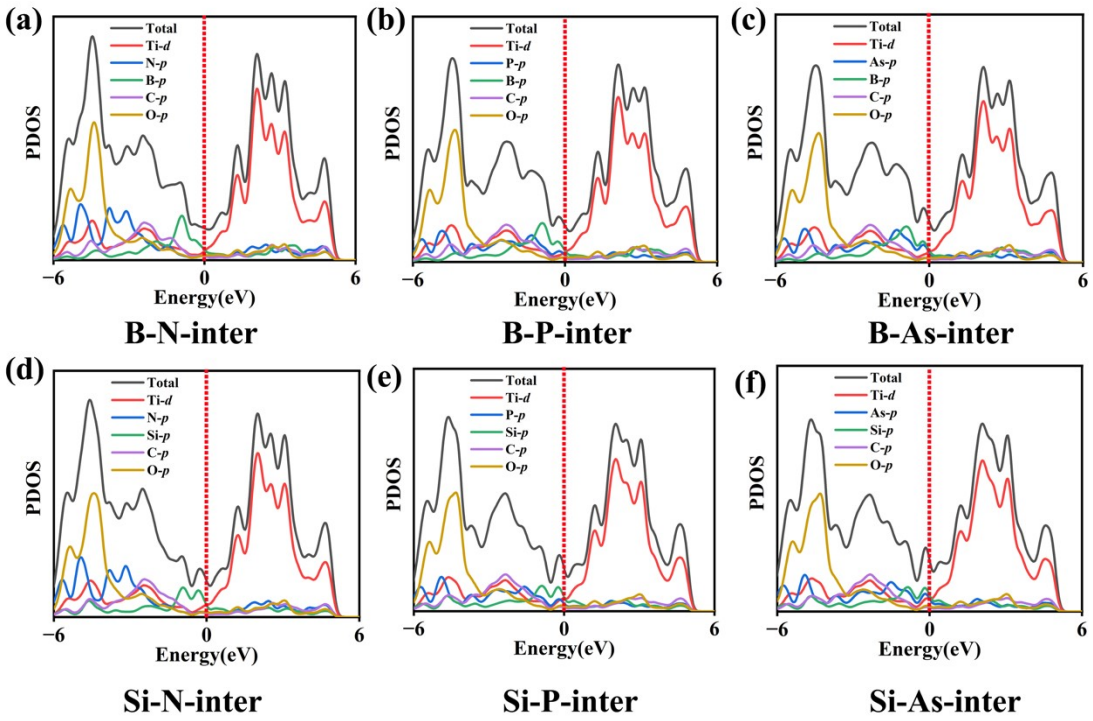

Figure S6. The projected density of states (PDOS) of (a-c) B-N/B-P/B-As-inter doped $Ti_3C_2O_2$ and (d-f) Si-N/Si-P/Si-As-inter doped $Ti_3C_2O_2$ as obtained from the *p*(3×3) unit cell.

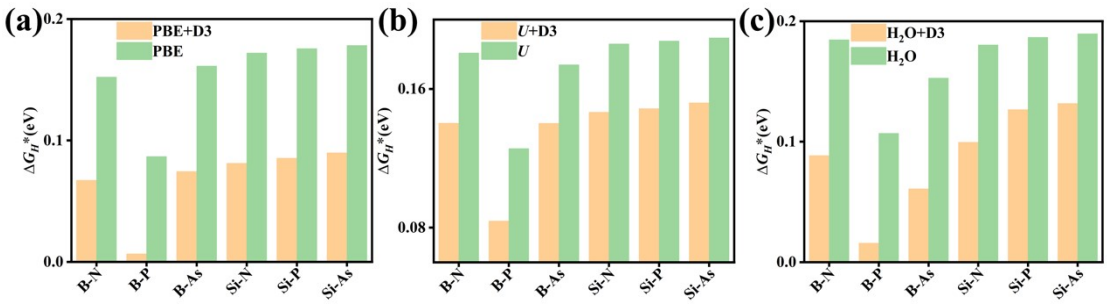

Figure S7. (a) The effect of D3 correction and no D3 correction under vacuum environment, (b) The effect of D3 correction and no D3 correction under Hubbard *U* conditions, (c) The effect of D3 correction and no D3 correction under H_2O solvent conditions.

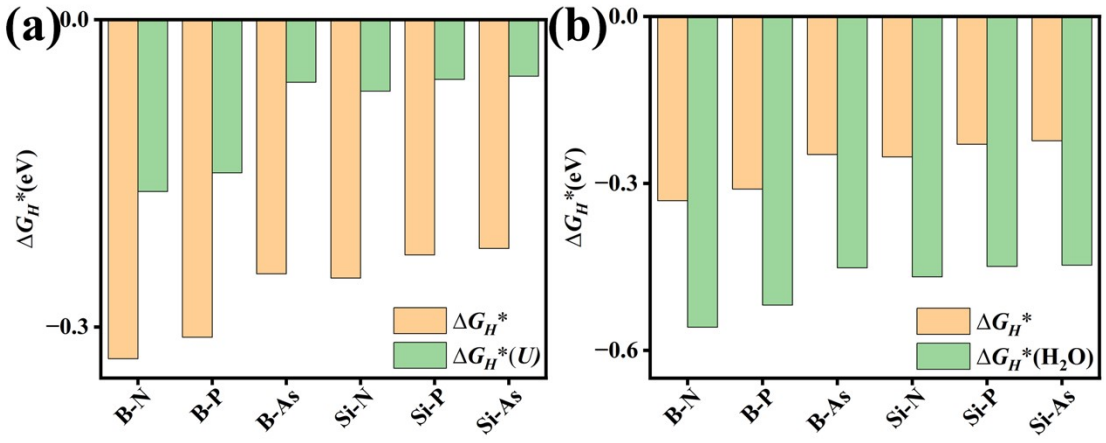

Figure S8. (a) the Effect of *Hubbard U*. (b) the Effect of H_2O solvent.

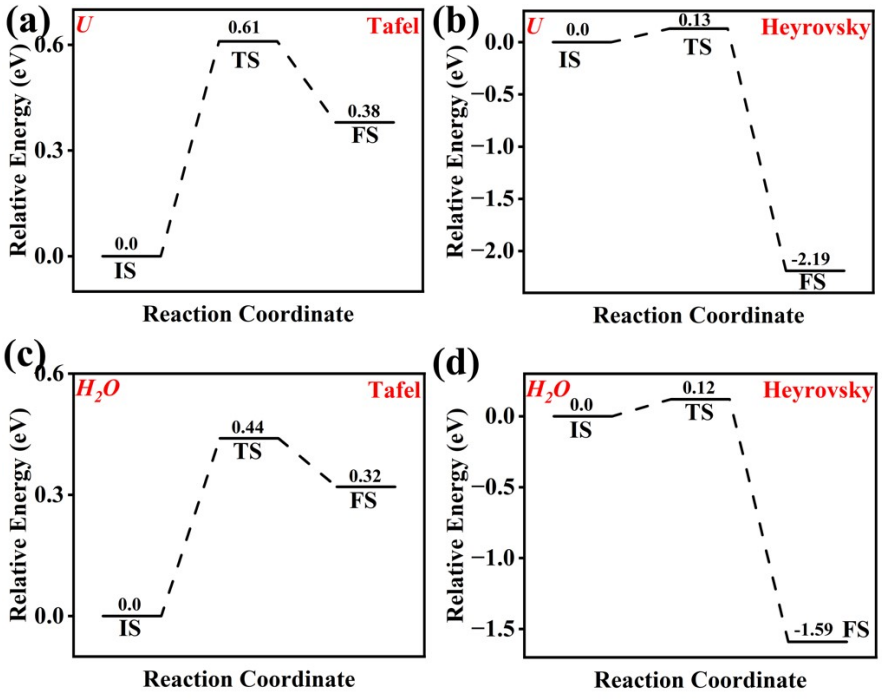

Figure S9. HER Tafel and Heyrovsky steps on Si-As-doped: (a) Tafel step with Hubbard *U* correction; (b) Heyrovsky step with Hubbard *U* correction; (c) Tafel step in aqueous solvent; (d) Heyrovsky step in aqueous solvent.

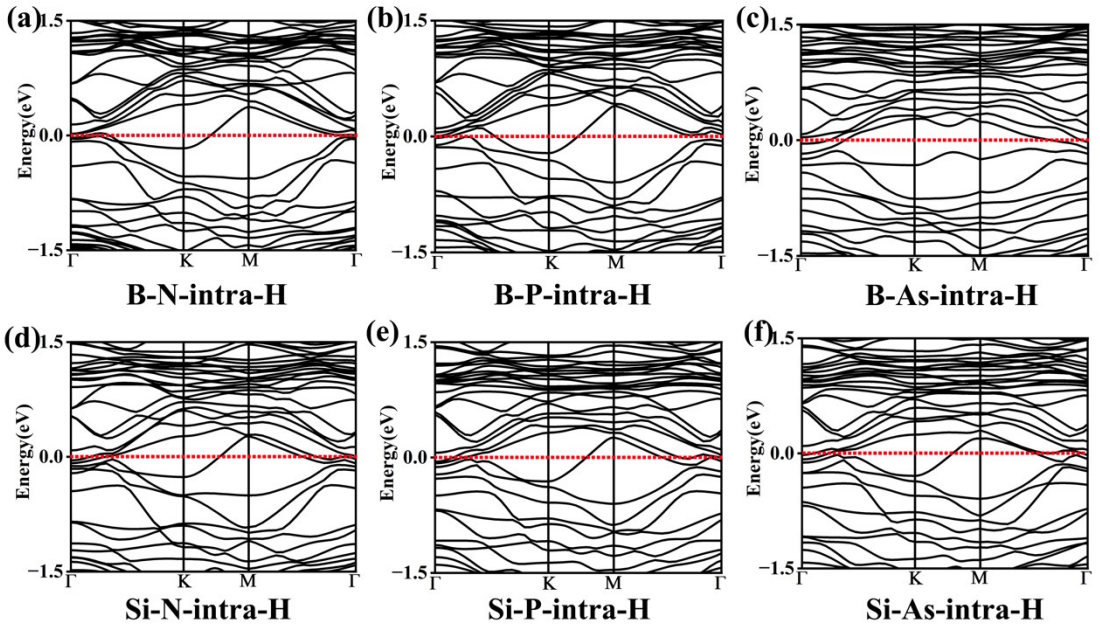

Figure S10. The band structure of H adsorbed on (a-c) B-N/B-P/B-As-intra doped $Ti_3C_2O_2$ and (d-f) Si-N/Si-P/Si-As-intra doped $Ti_3C_2O_2$.

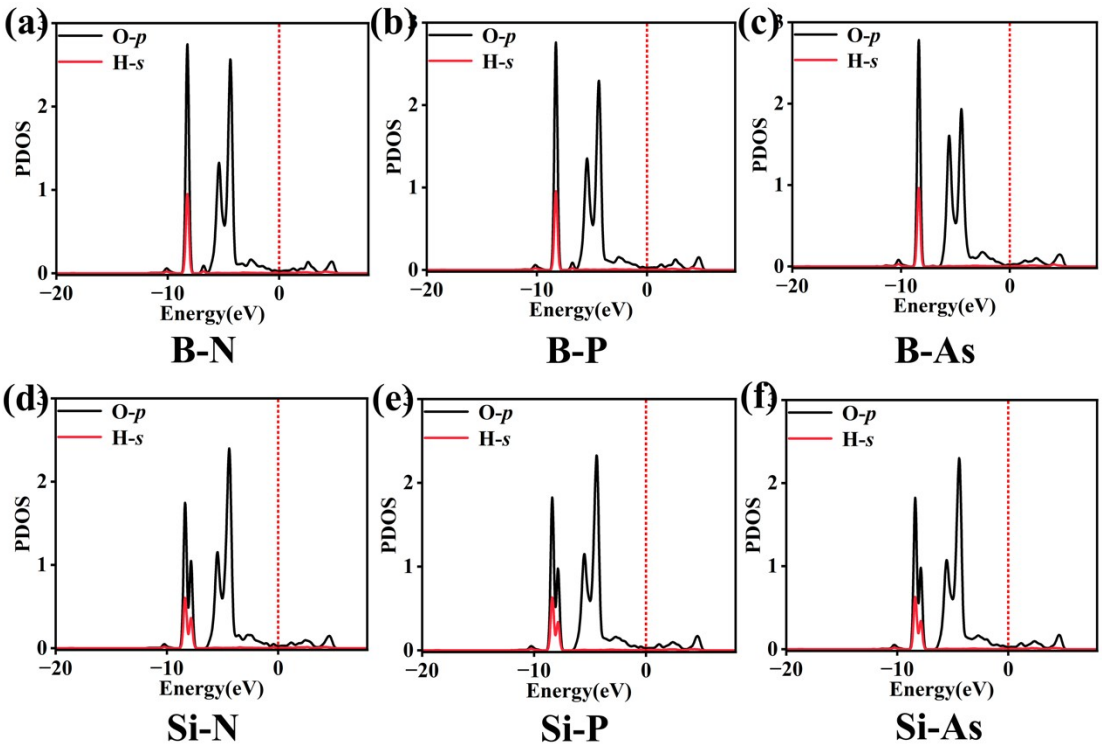

Figure S11. The PDOS diagrams of the O (2*p*) orbitals (bonded to H) and the H(1*s*) orbitals for (a-c) B-N/B-P/B-As-intra doped $Ti_3C_2O_2$ and (d-f) Si-N/Si-P/Si-As-intra doped $Ti_3C_2O_2$.

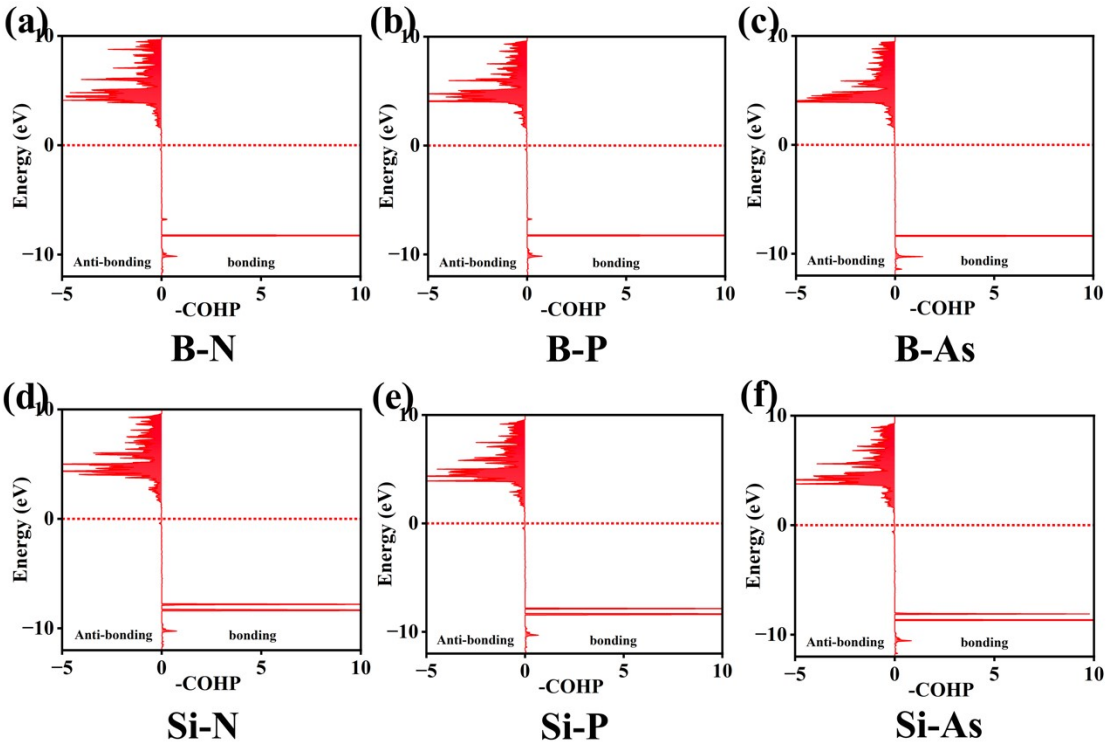

Figure S12. Crystal Orbital Hamilton Population (COHP) analysis of the interaction between H and O atom in (a-c) B-N/B-P/B-As-intra doped $Ti_3C_2O_2$ and (d-f) Si-N/Si-P/Si-As-intra doped $Ti_3C_2O_2$, the red dashed line indicates the Fermi level.

Table S1. The calculated ΔE_H, ΔE_{ZPE}-$T\Delta S$, ΔG_H^* of hydrogen adsorption on the top surface of the intra-doped $Ti_3C_2O_2$ structure.

mode	*X-Y-dop*	ΔE_H	ΔE_{ZPE}-$T\Delta S$	ΔG_H^*
intra	B-N	-0.565	0.234	-0.331
	B-P	-0.540	0.230	-0.310
	B-As	-0.486	0.238	-0.248
	Si-N	-0.497	0.245	-0.252
	Si-P	-0.4729	0.2441	-0.229
	Si-As	-0.4712	0.2448	-0.226

The VASP geometry files of starting geometry to carry out geometry optimization for pristine, intra-doped $Ti_3C_2O_2$ with two non-metallic atoms with best ΔG_H^*, including cell vectors, number of atoms, and direct, fractional coordinates:

pristine-$Ti_3C_2O_2$

1.0

9.1704998016	0.0000000000	0.0000000000
-4.5852499008	7.9418857936	0.0000000000
0.0000000000	0.0000000000	19.7441005707

Ti	C	O
27	18	18

Direct

0.958390034	0.980070017	0.421180021
0.291839999	0.980229966	0.421189971
0.625179985	0.980070017	0.421109984
0.958500045	0.313510020	0.421220015
0.291669996	0.313499993	0.421180021
0.625020029	0.313399995	0.421220015
0.958390071	0.646900028	0.421229965
0.291830043	0.647000056	0.421109984
0.624890072	0.646709999	0.421109984
0.180619992	0.091280002	0.293949990
0.513970006	0.091269998	0.293920019
0.847310001	0.091339998	0.293920019
0.180640006	0.424650009	0.293920019
0.513989962	0.424589999	0.293929993
0.847289978	0.424609992	0.293949990
0.180559981	0.757909999	0.293929993
0.513960032	0.757950047	0.293879977
0.847249990	0.757920026	0.293929993
0.736160003	0.869080039	0.171519993
0.402830027	0.869070013	0.171519993
0.069490012	0.869070013	0.171529991
0.736200006	0.535770035	0.171540002
0.402850011	0.535760009	0.171519993
0.069500025	0.535720021	0.171529991
0.736140021	0.202419999	0.171529991
0.402859985	0.202419999	0.171529991
0.069509993	0.202429996	0.171529991
0.736180029	0.869019999	0.355089989
0.402850019	0.869030025	0.355089989
0.069480002	0.869030025	0.355089989
0.736159997	0.535680034	0.355109985

0.402850002	0.535690001	0.355089989
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0.736199971	0.202350007	0.355100011
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0.069550004	0.202390009	0.355080014
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0.291709971	0.980170045	0.225289992
0.625049969	0.980189979	0.225309988
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0.291750028	0.313530013	0.225289992
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0.958409989	0.646850014	0.225319987
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0.625060039	0.646860041	0.225309988
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0.847289977	0.424530018	0.467599969
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0.513880003	0.424440017	0.467610016
0.847390022	0.091210002	0.467599969
0.513999973	0.091229996	0.467579972
0.847270030	0.757839992	0.467579972
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0.846639993	0.422560019	0.132350003
0.513309990	0.755900014	0.132340004
0.179979982	0.755990015	0.132360001
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0.513340006	0.089259997	0.132330006
0.846610011	0.755869993	0.132330006
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B, N-intra-$Ti_3C_2O_2$

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9.1704998016	0.0000000000	0.0000000000
-4.5852499008	7.9418857936	0.0000000000
0.0000000000	0.0000000000	19.7441005707

Ti	C	N	B	O
27	16	1	1	18

Direct

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0.291640030	0.979359975	0.424730005

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0.291660019	0.312680016	0.424740004
0.625000039	0.312689983	0.424740004
0.958320050	0.646009983	0.424730005
0.291660030	0.646030037	0.424740004
0.624989992	0.646009983	0.424740004
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0.513909993	0.091130005	0.295280003
0.847240005	0.091140002	0.295280003
0.180580013	0.424480004	0.295270004
0.513909999	0.424470007	0.295280003
0.847240011	0.424480004	0.295270004
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0.847240001	0.757800005	0.295280003
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0.402689989	0.868960018	0.165810003
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0.736010080	0.535610027	0.165810003
0.402680014	0.535619993	0.165810003
0.069350017	0.535610027	0.165810003
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0.069349993	0.202279999	0.165810003
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0.402799998	0.535450018	0.358430005
0.069460009	0.535450018	0.358430005
0.736130000	0.202120005	0.358430005
0.402799994	0.202120005	0.358430005
0.069470002	0.202120005	0.358430005
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0.291640063	0.980040057	0.232149997
0.624970069	0.980040057	0.232149997
0.958299994	0.313370005	0.232149997
0.291640021	0.313370005	0.232149997
0.624970047	0.313380002	0.232149997
0.958309980	0.646709999	0.232149997
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0.402799994	0.868780016	0.358430005
0.069470014	0.868780016	0.358430005
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B, P-intra-$Ti_3C_2O_2$

1.0

9.1704998016	0.0000000000	0.0000000000
-4.5852499008	7.9418857936	0.0000000000
0.0000000000	0.0000000000	19.7441005707

Ti	C	P	B	O
27	16	1	1	18

Direct

0.958329939	0.979359975	0.424740004
0.291640030	0.979359975	0.424730005
0.624989997	0.979330014	0.424740004
0.958309973	0.312689983	0.424740004
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0.958320050	0.646009983	0.424730005
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B, As-intra-$Ti_3C_2O_2$

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0.0000000000 0.0000000000 19.7441005707
Ti C As B O
27 16 1 1 18
Direct
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0.847150040 0.091069995 0.120109993
0.513819997 0.091080006 0.120119991
0.847150058 0.757740024 0.120109993
0.180480013 0.424409996 0.120109993

Si, N-intra-$Ti_3C_2O_2$

1.0
9.1704998016 0.0000000000 0.0000000000
-4.5852499008 7.9418857936 0.0000000000
0.0000000000 0.0000000000 19.7441005707
Ti C Si N O
27 16 1 1 18
Direct
0.958329939 0.979359975 0.424740004

0.291640030	0.979359975	0.424730005
0.624989997	0.979330014	0.424740004
0.958309973	0.312689983	0.424740004
0.291660019	0.312680016	0.424740004
0.625000039	0.312689983	0.424740004
0.958320050	0.646009983	0.424730005
0.291660030	0.646030037	0.424740004
0.624989992	0.646009983	0.424740004
0.180580013	0.091149999	0.295270004
0.513909993	0.091130005	0.295280003
0.847240005	0.091140002	0.295280003
0.180580013	0.424480004	0.295270004
0.513909999	0.424470007	0.295280003
0.847240011	0.424480004	0.295270004
0.180580016	0.757800005	0.295280003
0.513910017	0.757810031	0.295270004
0.847240001	0.757800005	0.295280003
0.736030017	0.868940024	0.165810003
0.402689989	0.868960018	0.165810003
0.069360044	0.868950051	0.165800005
0.736010080	0.535610027	0.165810003
0.402680014	0.535619993	0.165810003
0.069350017	0.535610027	0.165810003
0.736020000	0.202279999	0.165810003
0.402680011	0.202279999	0.165810003
0.069349993	0.202279999	0.165810003
0.736129991	0.868789982	0.358430005
0.402799994	0.868780016	0.358430005
0.736130003	0.535450018	0.358430005
0.402799998	0.535450018	0.358430005
0.069460009	0.535450018	0.358430005
0.402799994	0.202120005	0.358430005
0.069470002	0.202120005	0.358430005
0.958300022	0.980040057	0.232149997
0.291640063	0.980040057	0.232149997
0.624970069	0.980040057	0.232149997
0.958299994	0.313370005	0.232149997
0.291640021	0.313370005	0.232149997
0.624970047	0.313380002	0.232149997
0.958309980	0.646709999	0.232149997
0.291640037	0.646709999	0.232149997
0.624970043	0.646709999	0.232149997
0.069470014	0.868780016	0.358430005
0.736130000	0.202120005	0.358430005

0.180550002	0.090840001	0.470440014
0.847210001	0.424169983	0.470440014
0.513879992	0.757499981	0.470450013
0.180549999	0.757499981	0.470440014
0.513879996	0.424169983	0.470440014
0.847219996	0.090840001	0.470440014
0.513880007	0.090840001	0.470450013
0.847219981	0.757499981	0.470440014
0.180549990	0.424169983	0.470450013
0.180480007	0.091069995	0.120109993
0.847150047	0.424409996	0.120119991
0.513819974	0.757740024	0.120109993
0.180489995	0.757740024	0.120119991
0.513819989	0.424409996	0.120119991
0.847150040	0.091069995	0.120109993
0.513819997	0.091080006	0.120119991
0.847150058	0.757740024	0.120109993
0.180480013	0.424409996	0.120109993

Si, P-intra-$Ti_3C_2O_2$

1.0

9.1704998016	0.0000000000	0.0000000000
-4.5852499008	7.9418857936	0.0000000000
0.0000000000	0.0000000000	19.7441005707

Ti	C	Si	P	O
27	16	1	1	18

Direct

0.958329939	0.979359975	0.424740004
0.291640030	0.979359975	0.424730005
0.624989997	0.979330014	0.424740004
0.958309973	0.312689983	0.424740004
0.291660019	0.312680016	0.424740004
0.625000039	0.312689983	0.424740004
0.958320050	0.646009983	0.424730005
0.291660030	0.646030037	0.424740004
0.624989992	0.646009983	0.424740004
0.180580013	0.091149999	0.295270004
0.513909993	0.091130005	0.295280003
0.847240005	0.091140002	0.295280003
0.180580013	0.424480004	0.295270004
0.513909999	0.424470007	0.295280003
0.847240011	0.424480004	0.295270004
0.180580016	0.757800005	0.295280003

0.513910017	0.757810031	0.295270004
0.847240001	0.757800005	0.295280003
0.736030017	0.868940024	0.165810003
0.402689989	0.868960018	0.165810003
0.069360044	0.868950051	0.165800005
0.736010080	0.535610027	0.165810003
0.402680014	0.535619993	0.165810003
0.069350017	0.535610027	0.165810003
0.736020000	0.202279999	0.165810003
0.402680011	0.202279999	0.165810003
0.069349993	0.202279999	0.165810003
0.736129991	0.868789982	0.358430005
0.402799994	0.868780016	0.358430005
0.736130003	0.535450018	0.358430005
0.402799998	0.535450018	0.358430005
0.069460009	0.535450018	0.358430005
0.402799994	0.202120005	0.358430005
0.069470002	0.202120005	0.358430005
0.958300022	0.980040057	0.232149997
0.291640063	0.980040057	0.232149997
0.624970069	0.980040057	0.232149997
0.958299994	0.313370005	0.232149997
0.291640021	0.313370005	0.232149997
0.624970047	0.313380002	0.232149997
0.958309980	0.646709999	0.232149997
0.291640037	0.646709999	0.232149997
0.624970043	0.646709999	0.232149997
0.069470014	0.868780016	0.358430005
0.736130000	0.202120005	0.358430005
0.180550002	0.090840001	0.470440014
0.847210001	0.424169983	0.470440014
0.513879992	0.757499981	0.470450013
0.180549999	0.757499981	0.470440014
0.513879996	0.424169983	0.470440014
0.847219996	0.090840001	0.470440014
0.513880007	0.090840001	0.470450013
0.847219981	0.757499981	0.470440014
0.180549990	0.424169983	0.470450013
0.180480007	0.091069995	0.120109993
0.847150047	0.424409996	0.120119991
0.513819974	0.757740024	0.120109993
0.180489995	0.757740024	0.120119991
0.513819989	0.424409996	0.120119991
0.847150040	0.091069995	0.120109993

0.513819997 0.091080006 0.120119991
0.847150058 0.757740024 0.120109993
0.180480013 0.424409996 0.120109993

Si, As-intra-$Ti_3C_2O_2$

1.0
9.1704998016 0.0000000000 0.0000000000
-4.5852499008 7.9418857936 0.0000000000
0.0000000000 0.0000000000 19.7441005707
Ti C Si As O
27 16 1 1 18
Direct
0.958329939 0.979359975 0.424740004
0.291640030 0.979359975 0.424730005
0.624989997 0.979330014 0.424740004
0.958309973 0.312689983 0.424740004
0.291660019 0.312680016 0.424740004
0.625000039 0.312689983 0.424740004
0.958320050 0.646009983 0.424730005
0.291660030 0.646030037 0.424740004
0.624989992 0.646009983 0.424740004
0.180580013 0.091149999 0.295270004
0.513909993 0.091130005 0.295280003
0.847240005 0.091140002 0.295280003
0.180580013 0.424480004 0.295270004
0.513909999 0.424470007 0.295280003
0.847240011 0.424480004 0.295270004
0.180580016 0.757800005 0.295280003
0.513910017 0.757810031 0.295270004
0.847240001 0.757800005 0.295280003
0.736030017 0.868940024 0.165810003
0.402689989 0.868960018 0.165810003
0.069360044 0.868950051 0.165800005
0.736010080 0.535610027 0.165810003
0.402680014 0.535619993 0.165810003
0.069350017 0.535610027 0.165810003
0.736020000 0.202279999 0.165810003
0.402680011 0.202279999 0.165810003
0.069349993 0.202279999 0.165810003
0.736129991 0.868789982 0.358430005
0.402799994 0.868780016 0.358430005
0.736130003 0.535450018 0.358430005
0.402799998 0.535450018 0.358430005

0.069460009	0.535450018	0.358430005
0.402799994	0.202120005	0.358430005
0.069470002	0.202120005	0.358430005
0.958300022	0.980040057	0.232149997
0.291640063	0.980040057	0.232149997
0.624970069	0.980040057	0.232149997
0.958299994	0.313370005	0.232149997
0.291640021	0.313370005	0.232149997
0.624970047	0.313380002	0.232149997
0.958309980	0.646709999	0.232149997
0.291640037	0.646709999	0.232149997
0.624970043	0.646709999	0.232149997
0.069470014	0.868780016	0.358430005
0.736130000	0.202120005	0.358430005
0.180550002	0.090840001	0.470440014
0.847210001	0.424169983	0.470440014
0.513879992	0.757499981	0.470450013
0.180549999	0.757499981	0.470440014
0.513879996	0.424169983	0.470440014
0.847219996	0.090840001	0.470440014
0.513880007	0.090840001	0.470450013
0.847219981	0.757499981	0.470440014
0.180549990	0.424169983	0.470450013
0.180480007	0.091069995	0.120109993
0.847150047	0.424409996	0.120119991
0.513819974	0.757740024	0.120109993
0.180489995	0.757740024	0.120119991
0.513819989	0.424409996	0.120119991
0.847150040	0.091069995	0.120109993
0.513819997	0.091080006	0.120119991
0.847150058	0.757740024	0.120109993
0.180480013	0.424409996	0.120109993