Supporting Information

Universal Descriptor for Large-Scale Screening of High-Performance MXene-Based Materials for Energy Storage and Conversion

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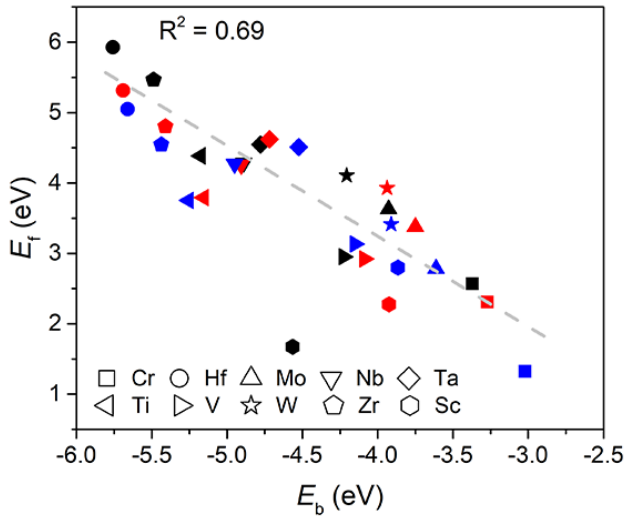

Figure S1. Linear relationship between oxygen binding energy (E_b) and oxygen vacancy formation energy (E_f). Black, red and blue symbols represent M_2CO_2, $M_3C_2O_2$ and $M_4C_3O_2$, respectively. Gray dashed line represents linear fitting.

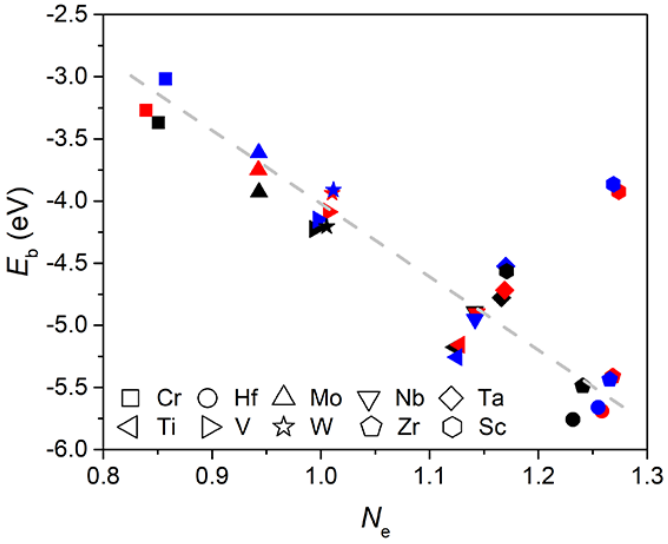

Figure S2. Linear relationship between the number of electrons gained by oxygen atom (N_e) and oxygen binding energy (E_b). Black, red and blue symbols represent M_2CO_2, $M_3C_2O_2$ and $M_4C_3O_2$, respectively. Gray dashed line represents linear fitting, except for $Sc_{n+1}C_nO_2$ (n=1, 2, 3).

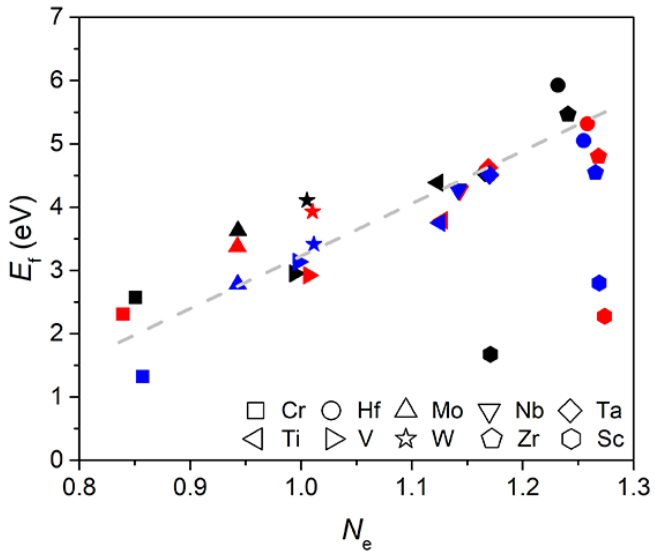

Figure S3. Linear relationship between the number of electrons gained by oxygen atom (N_e) and oxygen formation energy (E_f). Black, red and blue symbols represent M_2CO_2, $M_3C_2O_2$ and $M_4C_3O_2$, respectively. Gray dashed line represents linear fitting, except for $Sc_{n+1}C_nO_2$ (n=1, 2, 3).

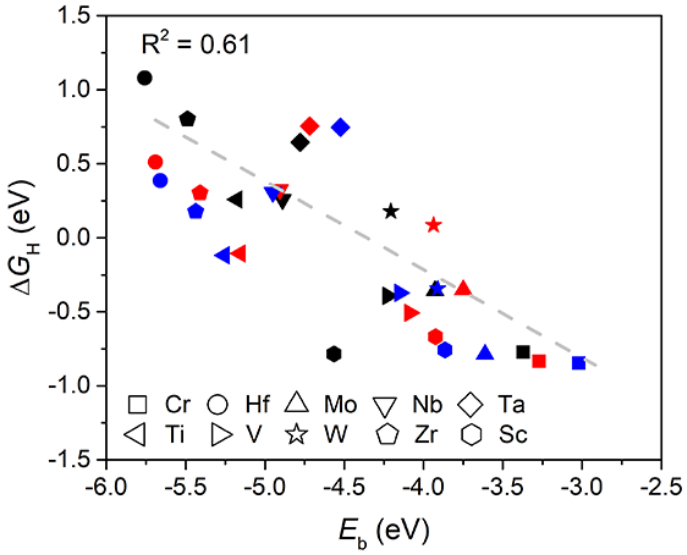

Figure S4. Linear relationship between oxygen binding energy (E_b) and free energy of hydrogen adsorption (ΔG_H). Black, red and blue symbols represent M_2CO_2, $M_3C_2O_2$ and $M_4C_3O_2$, respectively. Gray dashed line represents linear fitting.

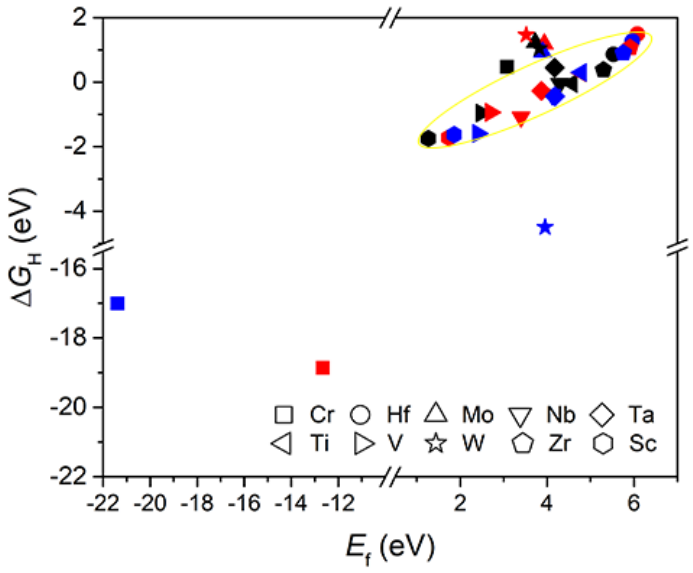

Figure S5. Relationship between oxygen formation energy (E_f) and free energy of hydrogen adsorption (ΔG_H) of $M_{n+1}N_nO_2$. Black, red and blue symbols represent M_2NO_2, $M_3N_2O_2$ and $M_4N_3O_2$, respectively. Yellow circle includes the data with strong linear relationships.

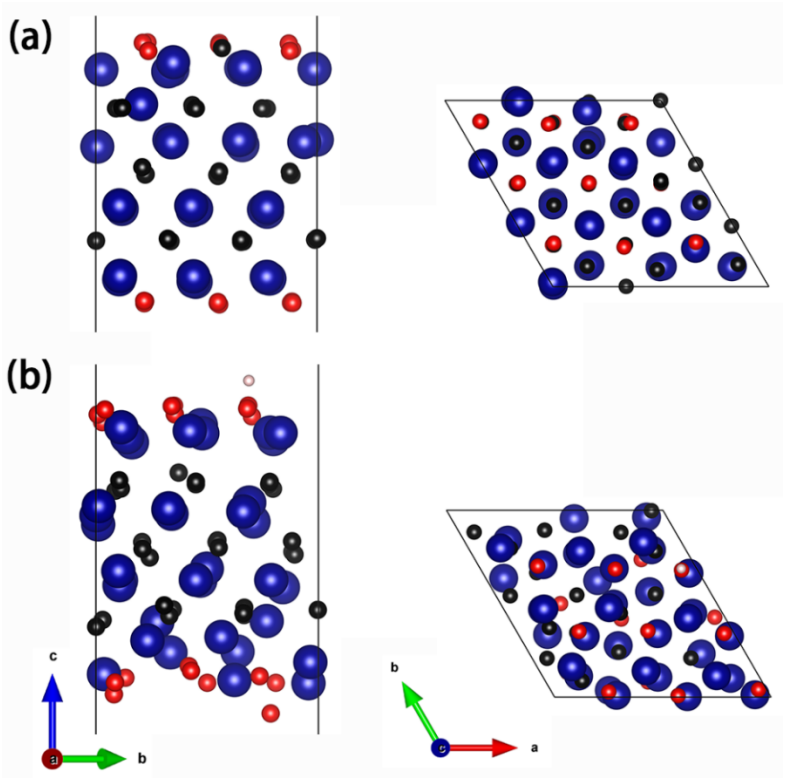

Figure S6. Optimized structures of (a) $Cr_4N_3O_2$ with oxygen vacancy, (b) hydrogen adsorption on $Cr_4N_3O_2$. Black, blue, red, white spheres represent nitrogen, chromium, oxygen and hydrogen atoms, respectively. From this figure, it is clear that serious distortion happens in the substrate when oxygen vacancy is introduced in, and hydrogen adsorbs on the substrates.

Table S1. O-H bond length of hydrogen adsorption on various oxygen-terminated MXenes substrates.

Substrates	O-H bond length (Å)	Substrates	O-H bond length (Å)	Substrates	O-H bond length (Å)
Sc_2CO_2	0.978	$Sc_3C_2O_2$	0.976	$Sc_4C_3O_2$	0.976
Cr_2CO_2	0.980	$Cr_3C_2O_2$	0.980	$Cr_4C_3O_2$	0.981
Hf_2CO_2	0.975	$Hf_3C_2O_2$	0.976	$Hf_4C_3O_2$	0.976
Mo_2CO_2	0.980	$Mo_3C_2O_2$	0.980	$Mo_4C_3O_2$	0.980
Nb_2CO_2	0.975	$Nb_3C_2O_2$	0.975	$Nb_4C_3O_2$	0.975
Ta_2CO_2	0.976	$Ta_3C_2O_2$	0.976	$Ta_4C_3O_2$	0.978
Ti_2CO_2	0.975	$Ti_3C_2O_2$	0.975	$Ti_4C_3O_2$	0.976
V_2CO_2	0.975	$V_3C_2O_2$	0.975	$V_4C_3O_2$	0.975
W_2CO_2	0.979	$W_3C_2O_2$	0.981	$W_4C_3O_2$	0.981
Zr_2CO_2	0.974	$Zr_3C_2O_2$	0.975	$Zr_4C_3O_2$	0.975
Sc_2NO_2	0.974	$Sc_3N_2O_2$	0.973	$Sc_4N_3O_2$	0.974
Cr_2NO_2	0.979	$Cr_3N_2O_2$	0.976	$Cr_4N_3O_2$	0.977
Hf_2NO_2	0.976	$Hf_3N_2O_2$	0.976	$Hf_4N_3O_2$	0.976
Mo_2NO_2	0.984	$Mo_3N_2O_2$	0.979	$Mo_3N_2O_2$	0.979
Nb_2NO_2	0.976	$Nb_3N_2O_2$	0.976	$Nb_4N_3O_2$	0.975
Ta_2NO_2	0.976	$Ta_3N_2O_2$	0.978	$Ta_4N_3O_2$	0.978
Ti_2NO_2	0.974	$Ti_3N_2O_2$	0.974	$Ti_4N_3O_2$	0.974
V_2NO_2	0.976	$V_3N_2O_2$	0.976	$V_4N_3O_2$	0.976
W_2NO_2	0.983	$W_3N_2O_2$	0.986	$W_4N_3O_2$	0.978
Zr_2NO_2	0.975	$Zr_3N_2O_2$	0.975	$Zr_4N_3O_2$	0.975

Table S2. Energies of fcc- and hcp-type oxygen-terminated MXenes. The energies of the most stable configurations are highlighted.

	Energy (eV)			Energy (eV)	
	fcc-type	hcp-type		fcc-type	hcp-type
Sc_2CO_2	-40.43	**-40.82**	Sc_2NO_2	**-42.50**	-41.77
Cr_2CO_2	-42.23	**-42.51**	Cr_2NO_2	-42.08	**-43.03**
Hf_2CO_2	**-50.54**	-48.45	Hf_2NO_2	**-51.05**	-49.97
Mo_2CO_2	-45.35	**-46.22**	Mo_2NO_2	-44.99	**-45.91**
Nb_2CO_2	**-47.94**	-47.23	Nb_2NO_2	**-47.94**	-47.47
Ta_2CO_2	**-51.28**	-50.70	Ta_2NO_2	**-50.90**	-50.82
Ti_2CO_2	**-44.99**	-42.94	Ti_2NO_2	**-45.94**	-44.65
V_2CO_2	**-44.41**	-43.67	V_2NO_2	**-44.79**	-44.35
W_2CO_2	-48.63	**-50.40**	W_2NO_2	-48.36	**-49.26**
Zr_2CO_2	**-47.21**	-45.33	Zr_2NO_2	**-48.06**	-46.77
Sc_3C_2O2	**-56.28**	-56.08	$Sc_3N_2O_2$	**-60.63**	-51.28
$Cr_3C_2O_2$	**-61.16**	-60.95	$Cr_3N_2O_2$	**-60.70**	-59.67
$Hf_3C_2O_2$	**-71.58**	-69.62	$Hf_3N_2O_2$	**-72.97**	-71.85
$Mo_3C_2O_2$	-65.18	**-65.95**	$Mo_3N_2O_2$	-64.57	**-65.78**
$Nb_3C_2O_2$	**-68.33**	-67.56	$Nb_3N_2O_2$	**-68.26**	-68.19
$Ta_3C_2O_2$	**-73.34**	-72.78	$Ta_3N_2O_2$	-72.44	**-72.66**
$Ti_3C_2O_2$	**-63.61**	-62.03	$Ti_3N_2O_2$	**-65.68**	-64.33
$V_3C_2O_2$	**-63.35**	-62.52	$V_3N_2O_2$	**-64.05**	-63.75
$W_3C_2O_2$	-70.34	**-71.57**	$W_3N_2O_2$	-69.36	**-70.38**
$Zr_3C_2O_2$	**-66.68**	-64.90	$Zr_3N_2O_2$	**-68.55**	-67.38
$Sc_4C_3O_2$	-71.88	**-72.35**	$Sc_4N_3O_2$	**-79.65**	-78.38
$Cr_4C_3O_2$	-72.65	**-72.98**	$Cr_4N_3O_2$	-75.02	**-77.84**
$Hf_4C_3O_2$	**-92.61**	-90.64	$Hf_4N_3O_2$	**-94.77**	-93.74
$Mo_4C_3O_2$	-84.93	**-85.51**	$Mo_4N_3O_2$	-84.11	**-85.40**
$Nb_4C_3O_2$	**-88.98**	-88.46	$Nb_4N_3O_2$	**-89.04**	-88.49
$Ta_4C_3O_2$	**-95.60**	-95.22	$Ta_4N_3O_2$	-94.22	**-94.72**
$Ti_4C_3O_2$	**-82.28**	-80.67	$Ti_4N_3O_2$	**-85.28**	-84.10
$V_4C_3O_2$	**-82.52**	-81.91	$V_4N_3O_2$	**-83.38**	-82.81
$W_4C_3O_2$	-92.03	**-93.05**	$W_4N_3O_2$	-90.42	**-91.67**
$Zr_4C_3O_2$	**-86.12**	-84.35	$Zr_4N_3O_2$	**-88.91**	-87.84

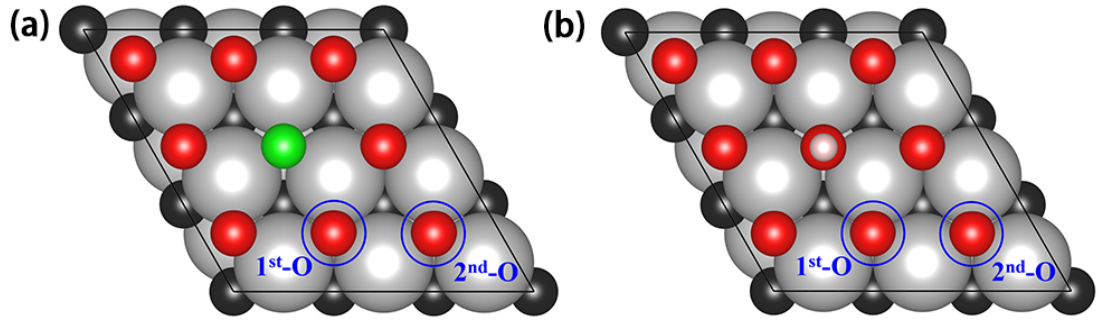

Figure S7. (a) M_2CO_2 with one F substitution for O; (b) M_2CO_2 with one OH substitution for O. 1st-O and 2nd-O indicate the nearest neighbor and next nearest neighbor of the substitution site, respectively, where hydrogen adsorption happens and oxygen vacancy forms. Gray, black, white, green and red spheres represent transition metals, carbon, hydrogen, fluorine and oxygen atoms, respectively.

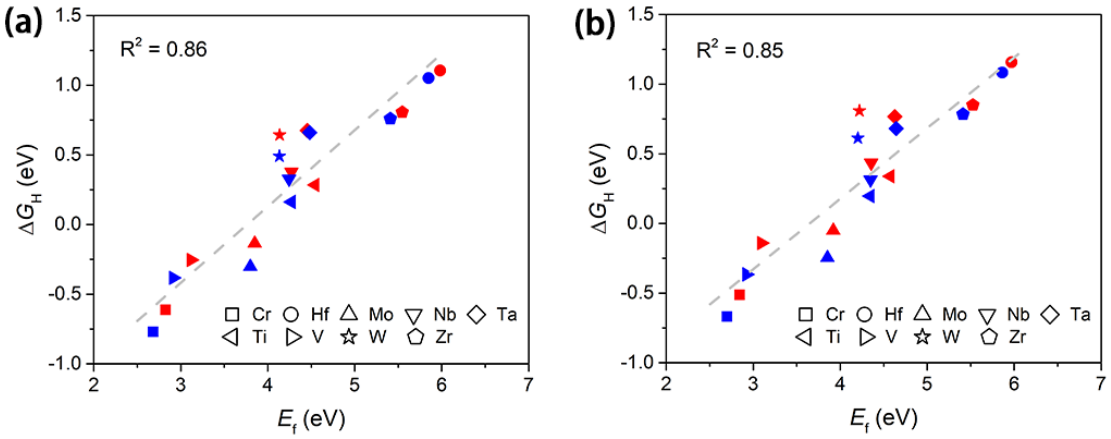

Figure S8. Linear relationship between oxygen vacancy formation energy (E_f) and adsorption free energy of hydrogen (ΔG_H) of oxygen-terminated MXenes with (a) F substitution for O, and (b) OH substitution for O. Red and blue symbols represent H adsorbed on the nearest neighbor of the substitution site and next nearest neighbor of the substitution site, respectively.

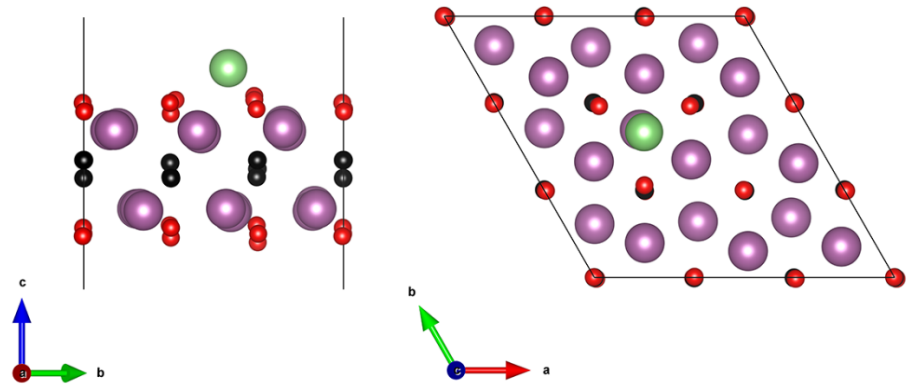

Figure S9. Structure distortion of Sc_2CO_2 after Li adsorption. Black, purple, red and green spheres represent carbon, scandium, oxygen and lithium, respectively.