

Supplementary Information

Surface states of dual-atom catalysts should be considered for analysis of electrocatalytic activity

Weijie Yang¹, Zhenhe Jia¹, Binghui Zhou¹, Li Wei², Zhengyang Gao^{1 *}, Hao Li^{3 *}

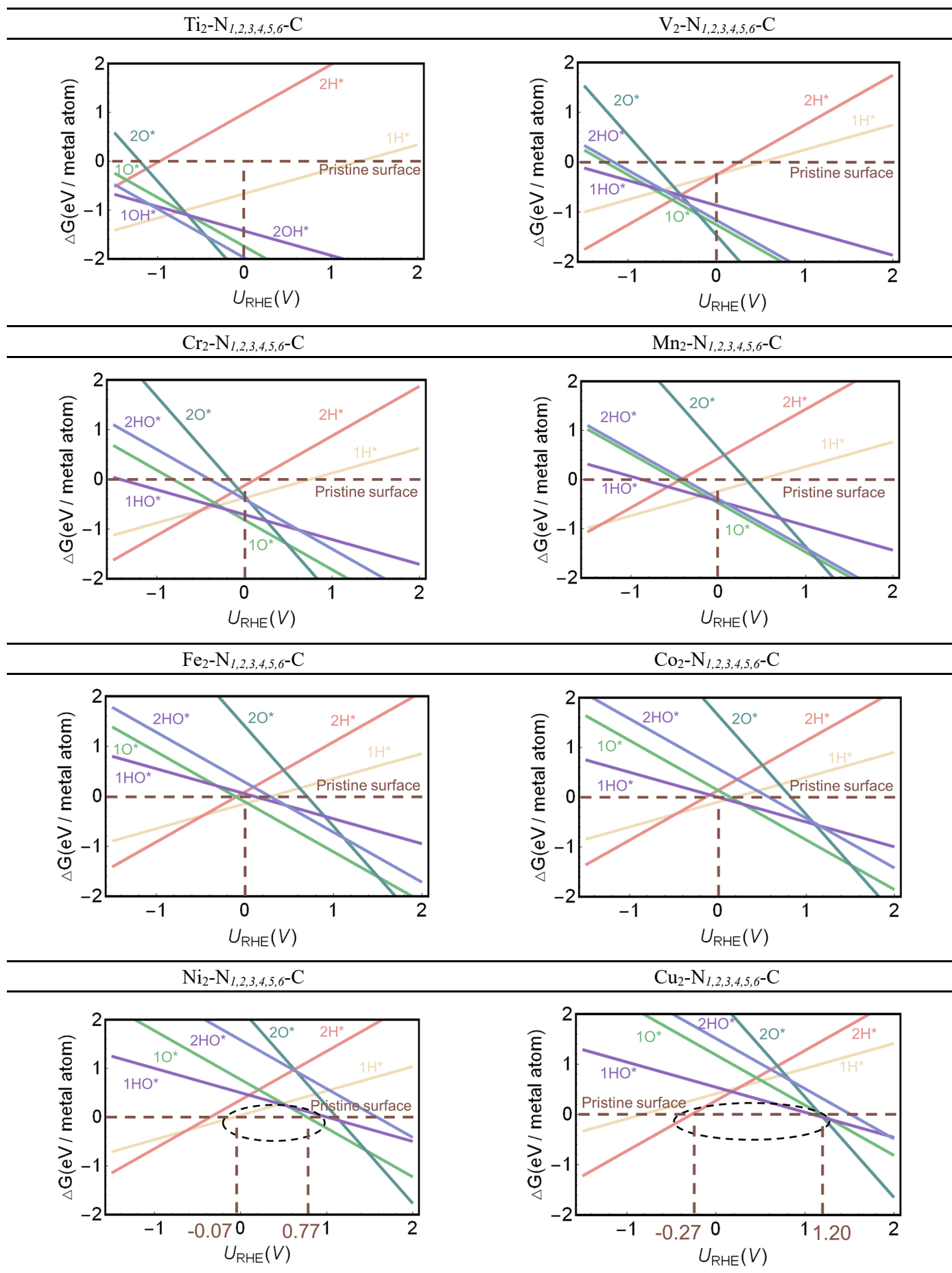
¹ Department of Power Engineering, School of Energy, Power and Mechanical Engineering, North China Electric Power University, Baoding 071003, China

² School of Chemical and Biomolecule Engineering, The University of Sydney, Darlington 2006, NSW, Australia

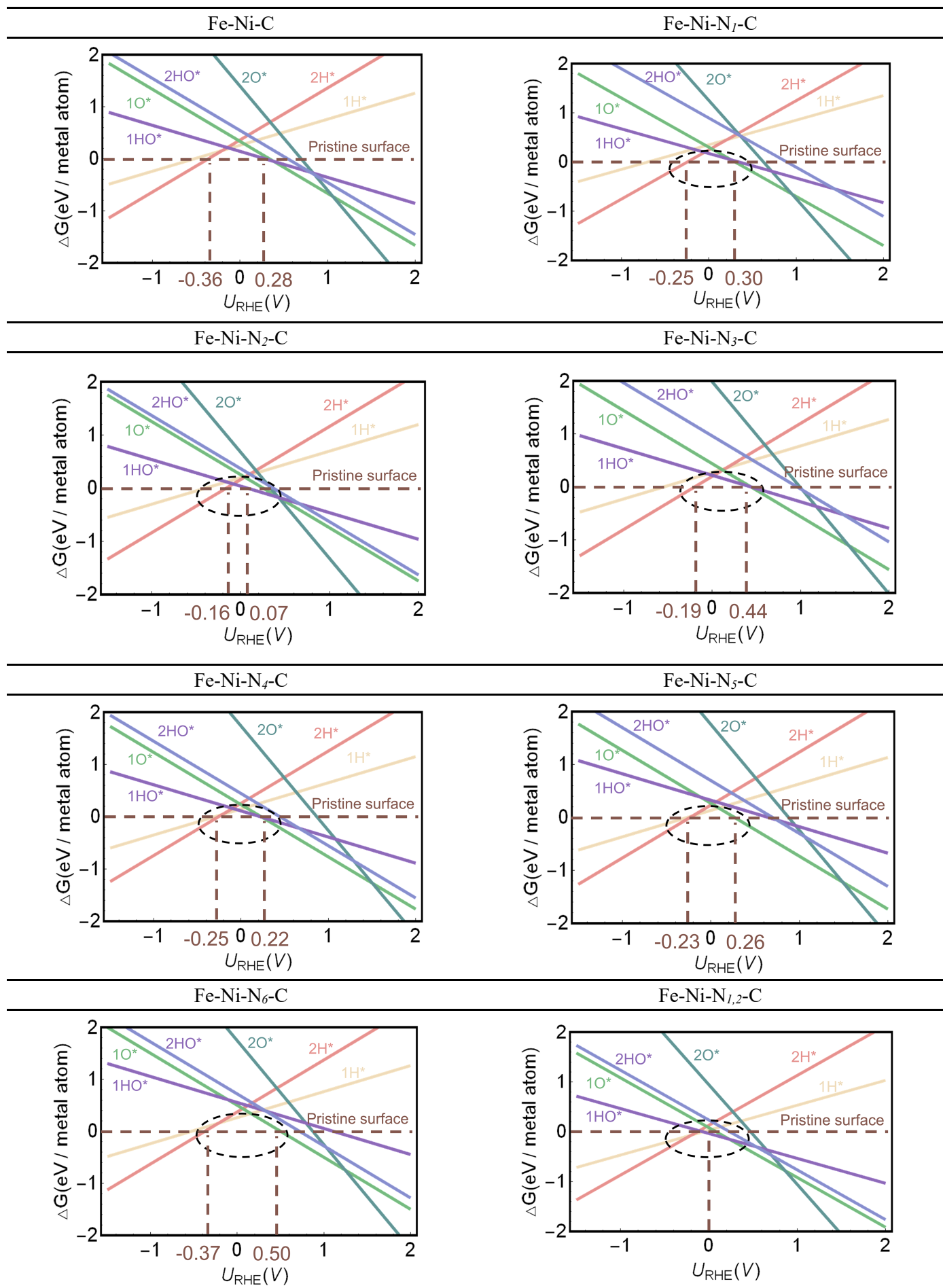
³ Advanced Institute for Materials Research (WPI-AIMR), Tohoku University, Sendai 980-8577, Japan

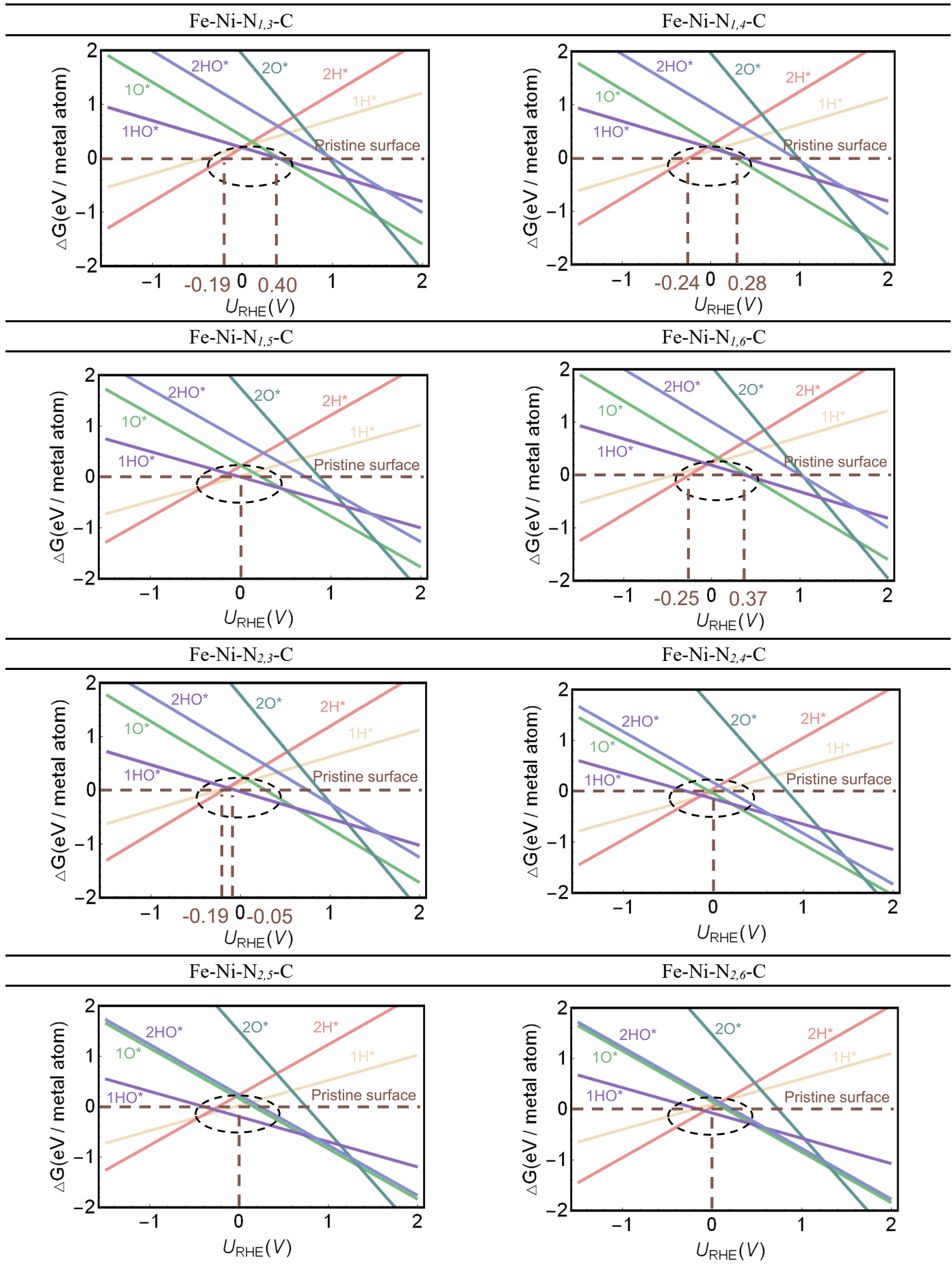
* e-mail: gaozhyan@163.com; li.hao.b8@tohoku.ac.jp

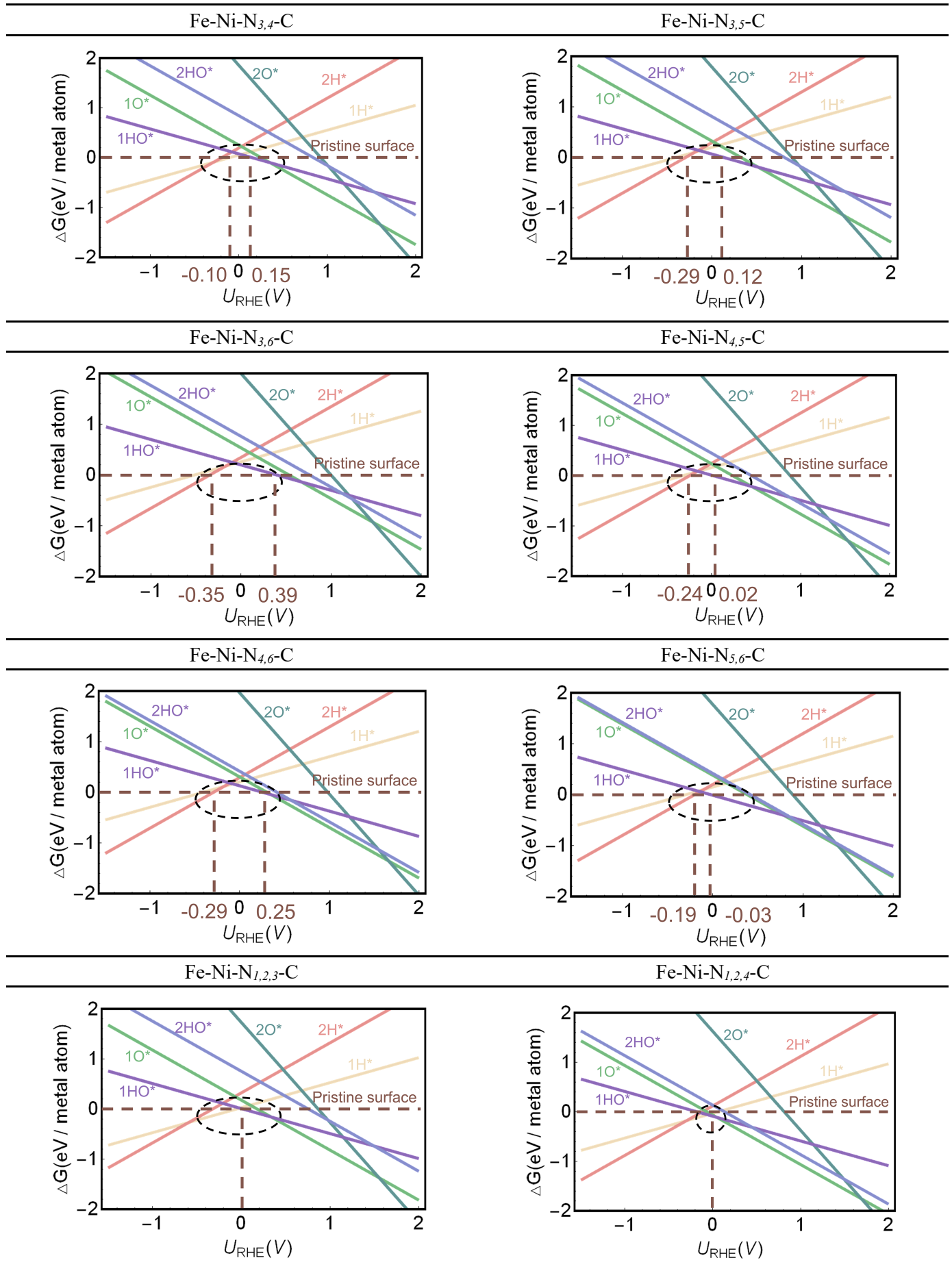
Supplementary Table 1. Surface Pourbaix diagrams of 3d homonuclear DACs at pH = 0.

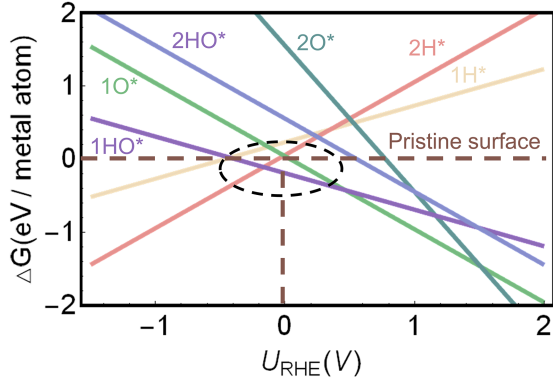
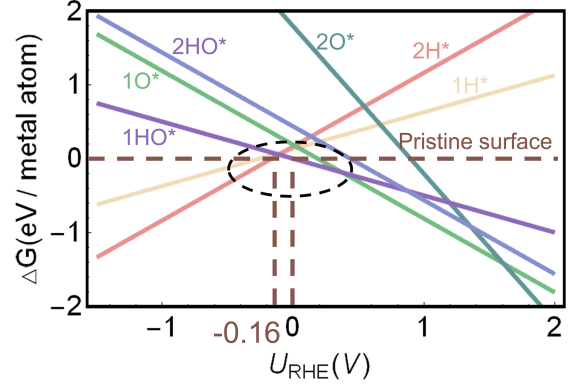
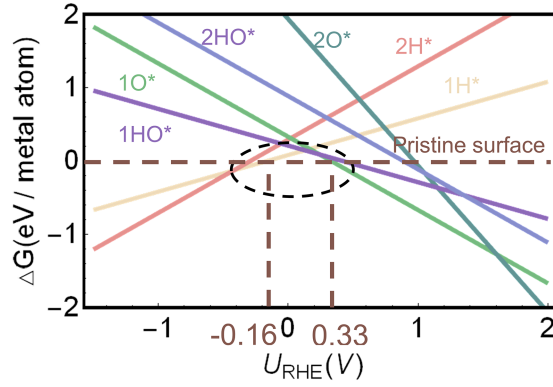
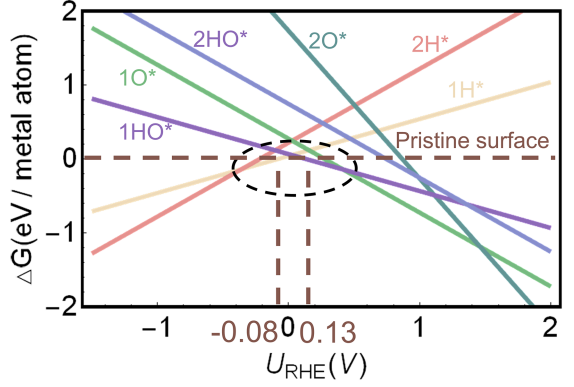
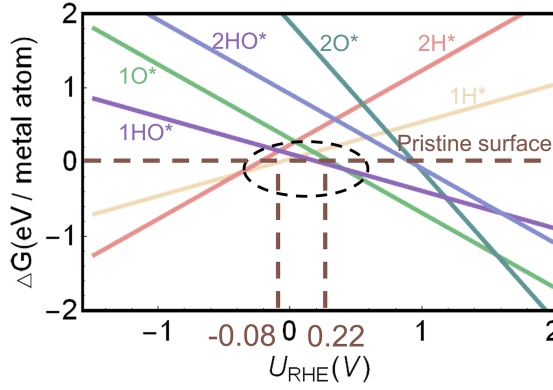
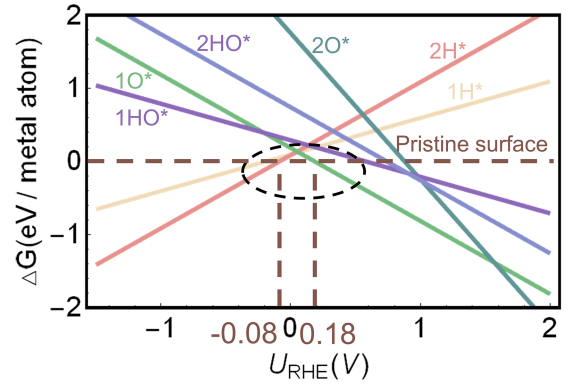
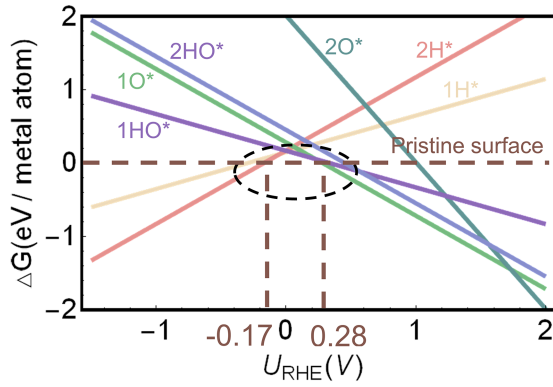
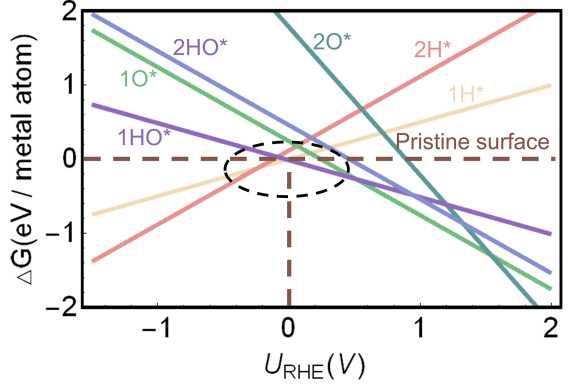


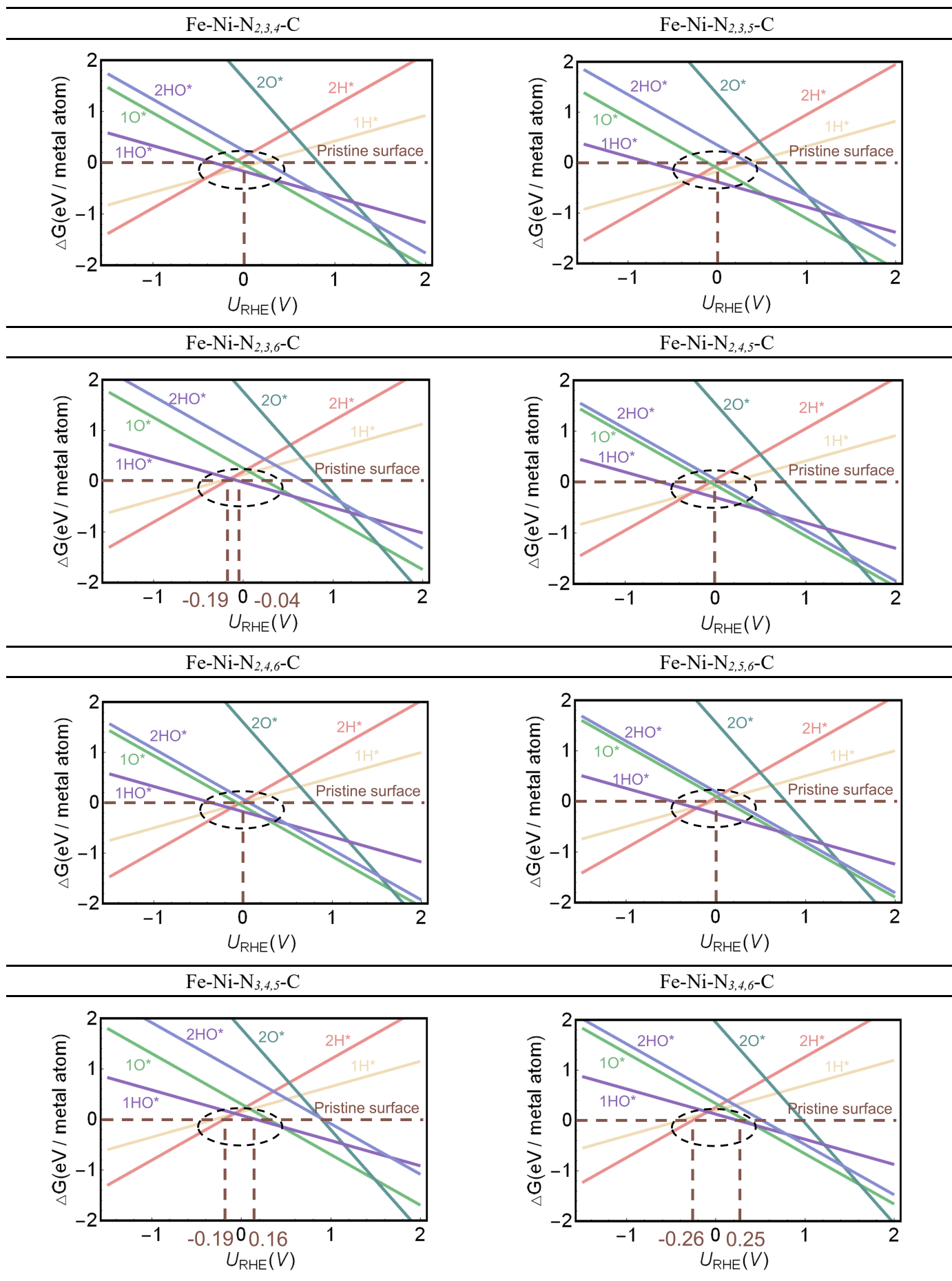
Supplementary Table 2. Surface Pourbaix diagrams of Fe-Ni-N_x-C at pH = 0

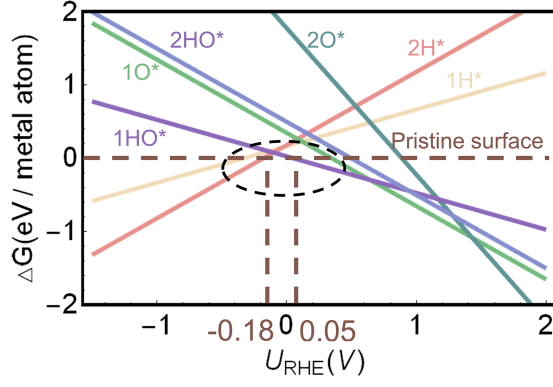
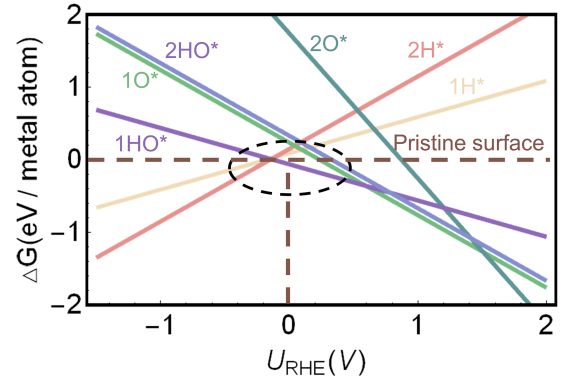
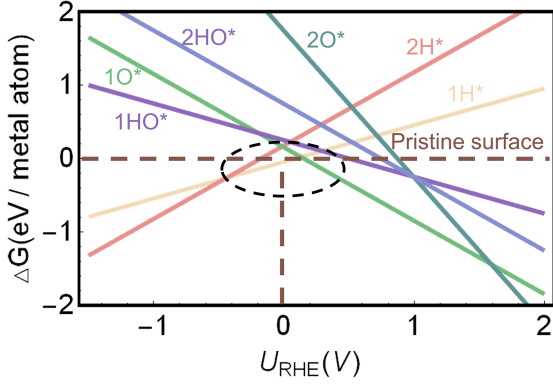
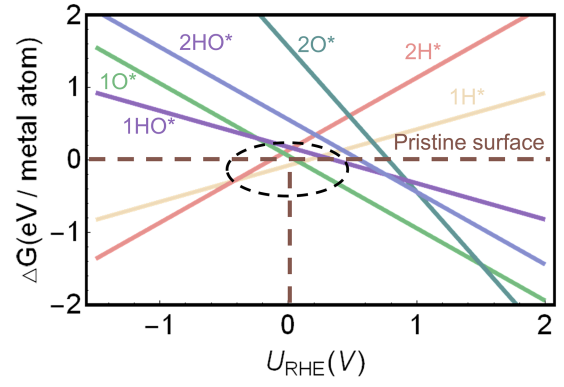
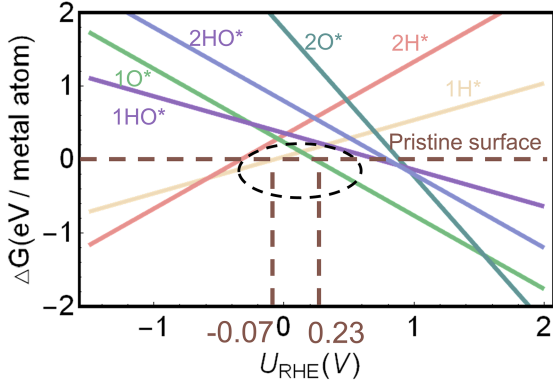
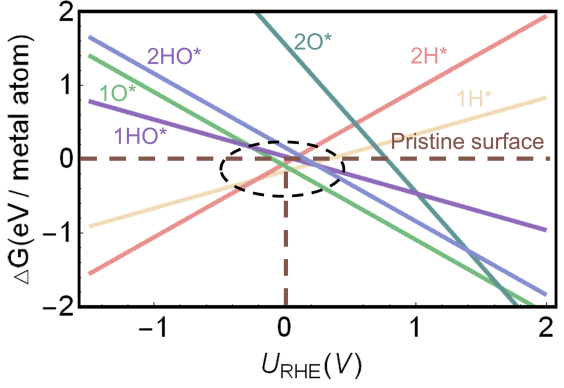
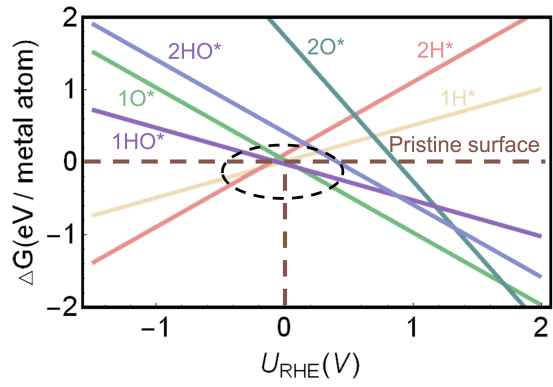
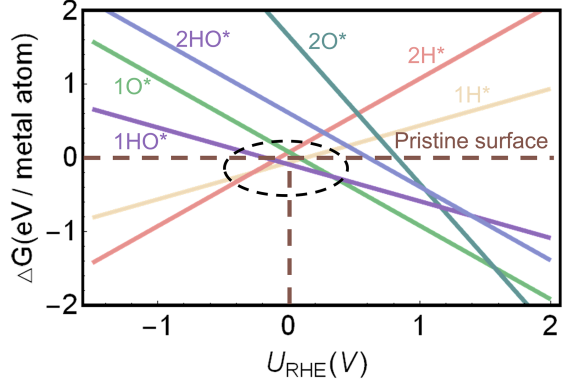


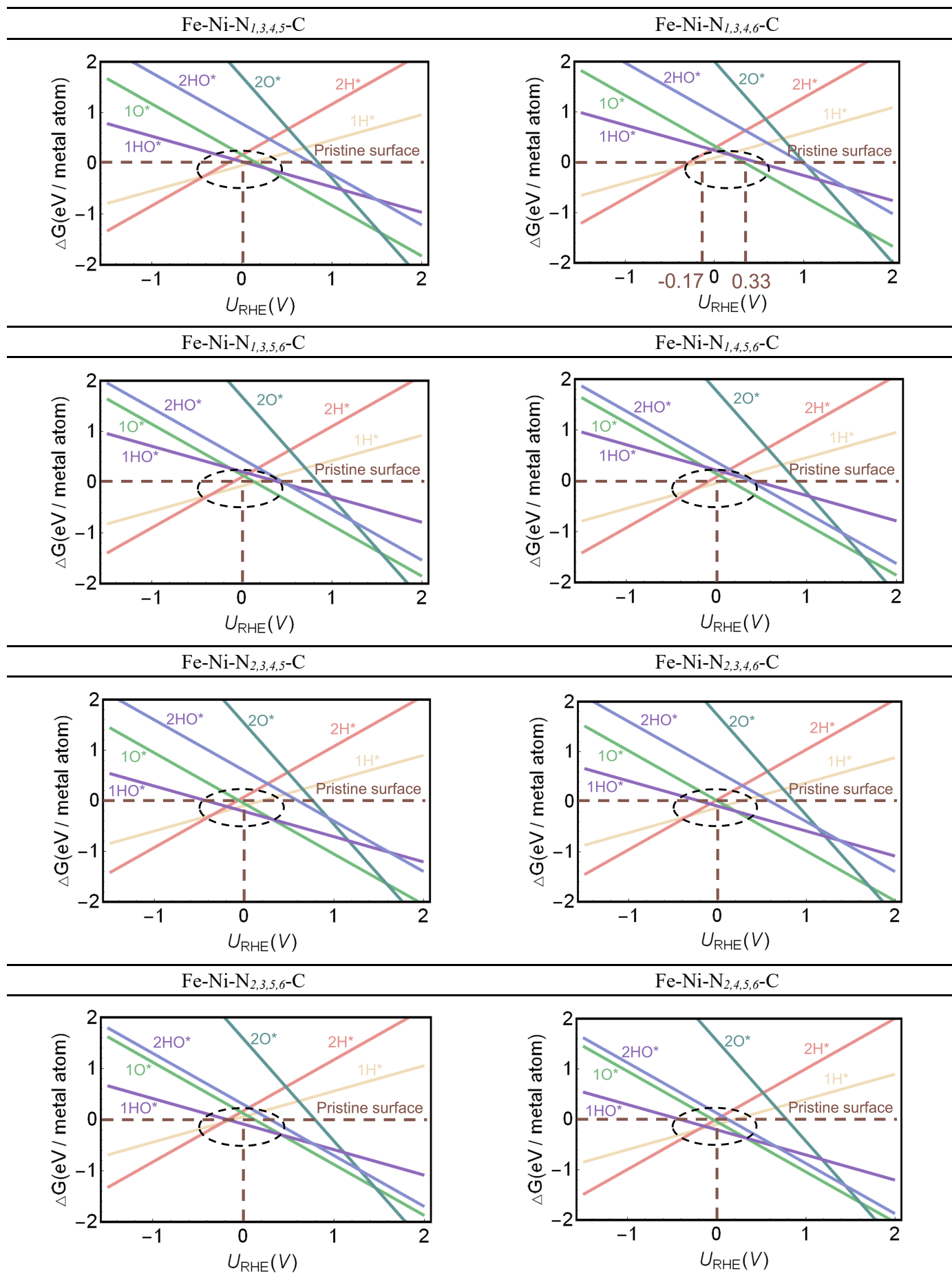


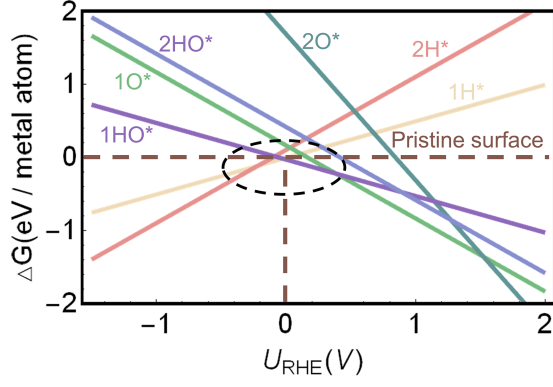
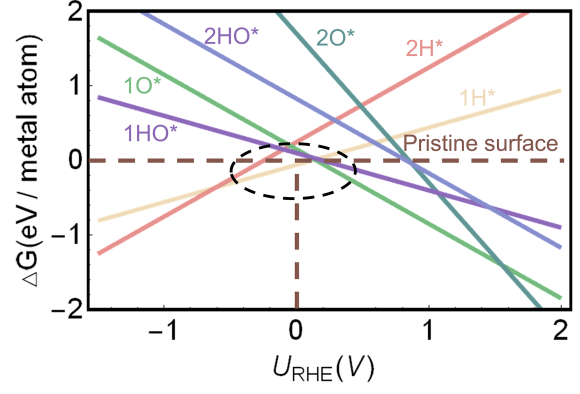
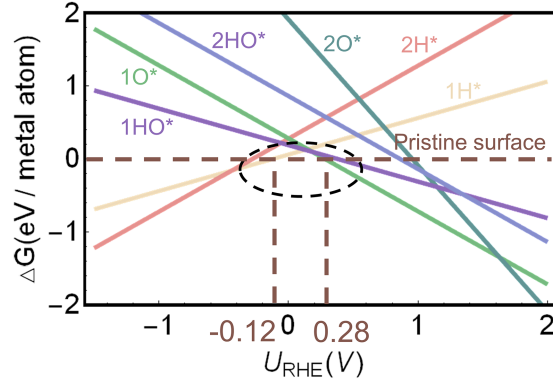
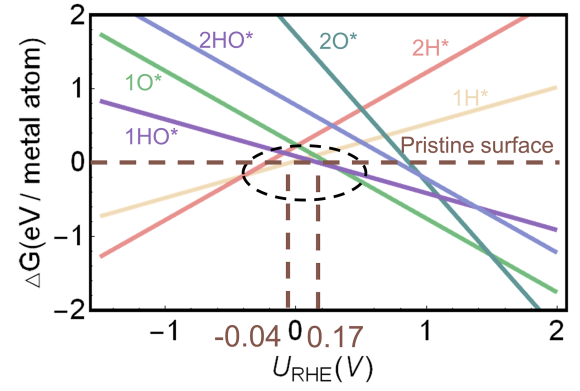
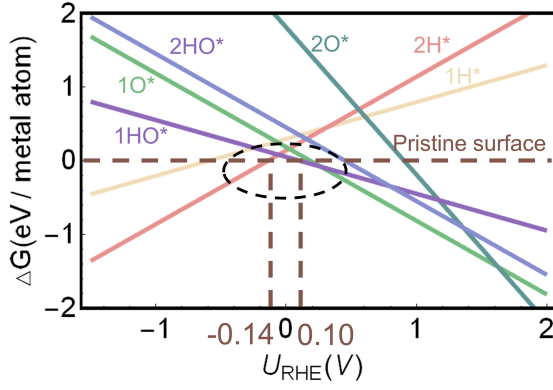
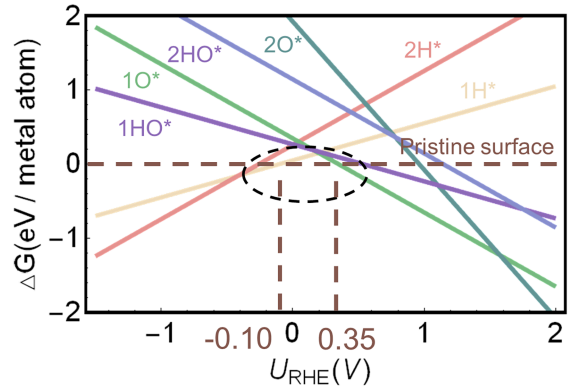
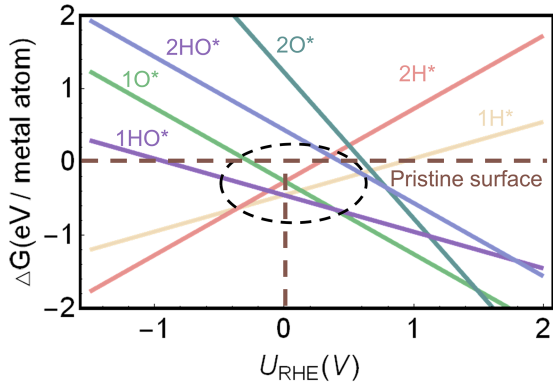
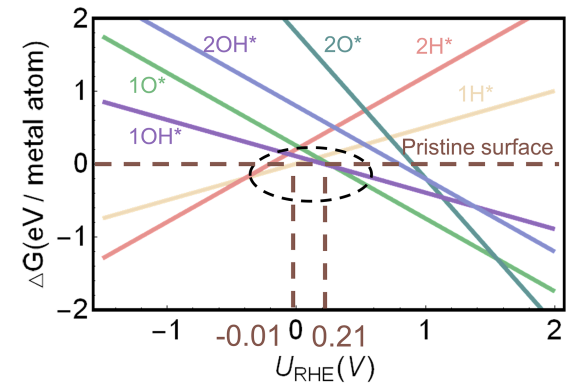


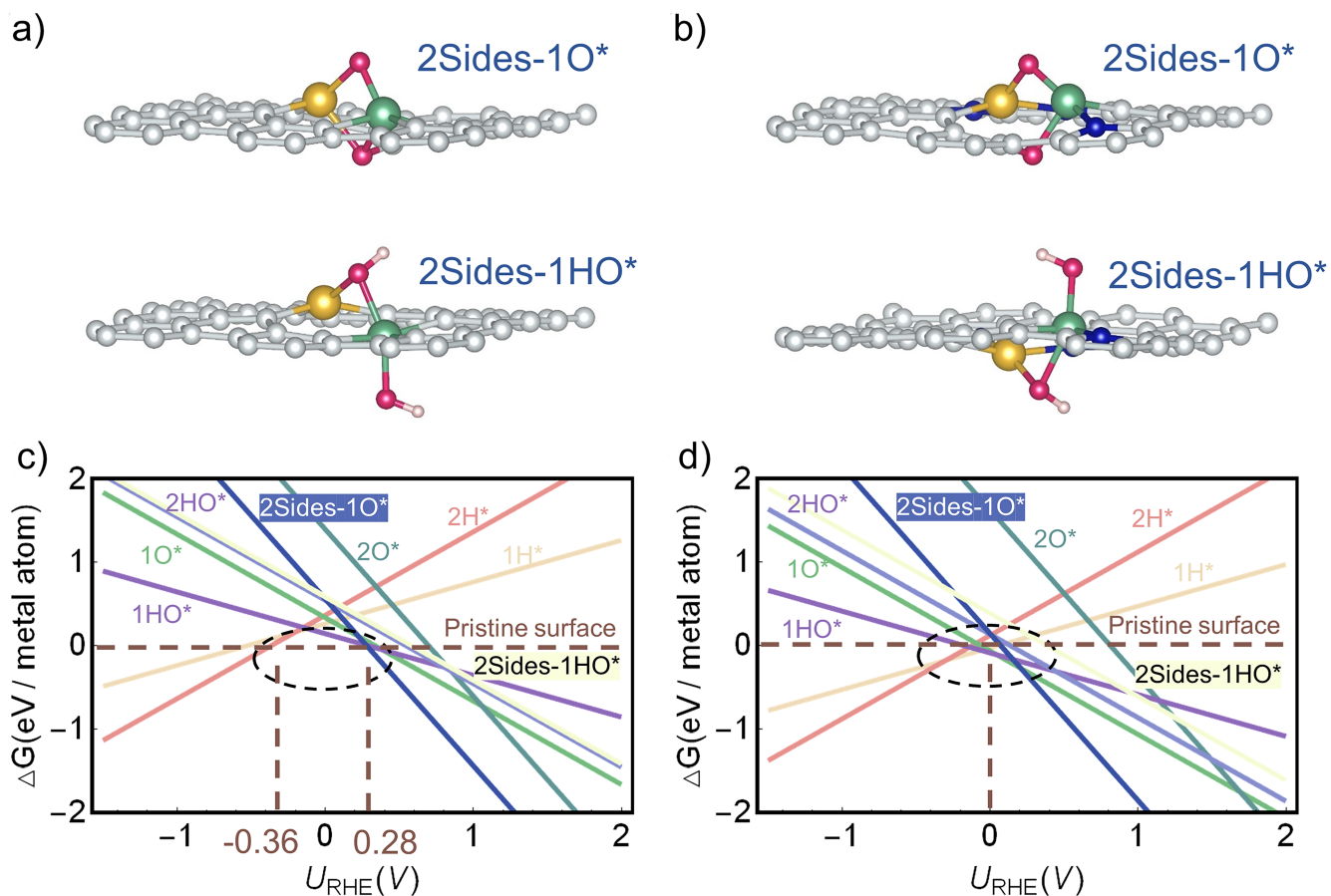
Fe-Ni-N_{I,2,5}-CFe-Ni-N_{I,2,6}-CFe-Ni-N_{I,3,4}-CFe-Ni-N_{I,3,5}-CFe-Ni-N_{I,3,6}-CFe-Ni-N_{I,4,5}-CFe-Ni-N_{I,4,6}-CFe-Ni-N_{I,5,6}-C



Fe-Ni-N_{3,5,6}-CFe-Ni-N_{4,5,6}-CFe-Ni-N_{I,2,3,4}-CFe-Ni-N_{I,2,3,5}-CFe-Ni-N_{I,2,3,6}-CFe-Ni-N_{I,2,4,5}-CFe-Ni-N_{I,2,4,6}-CFe-Ni-N_{I,2,5,6}-C

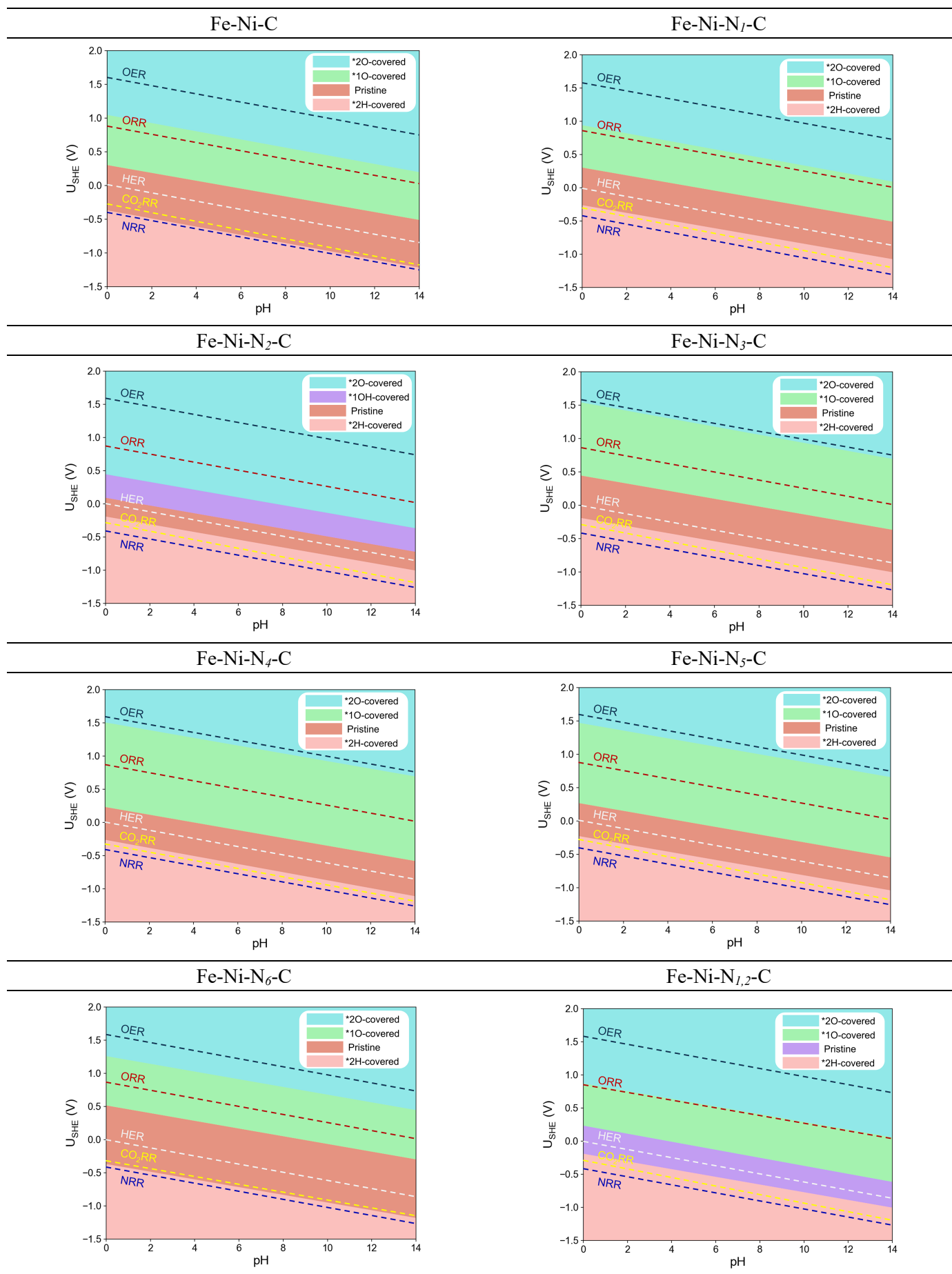


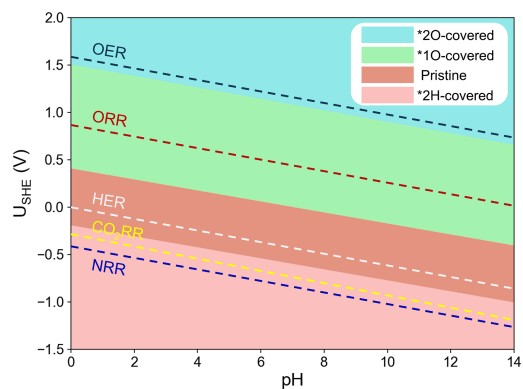
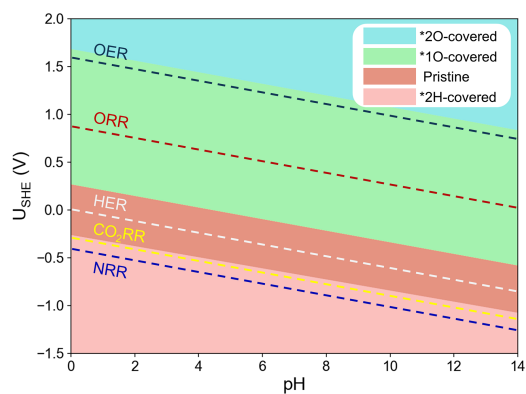
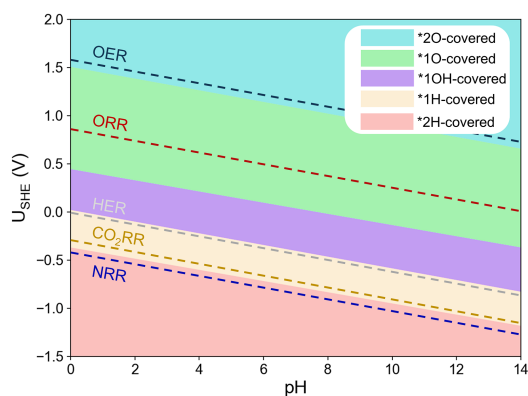
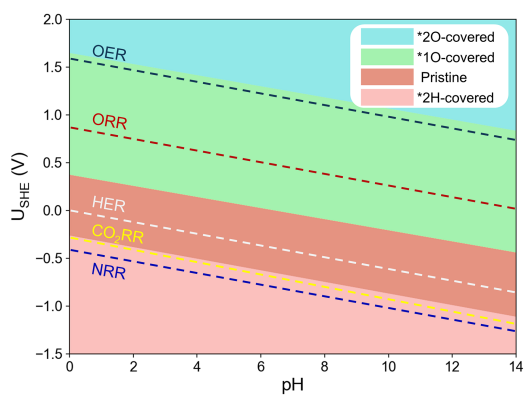
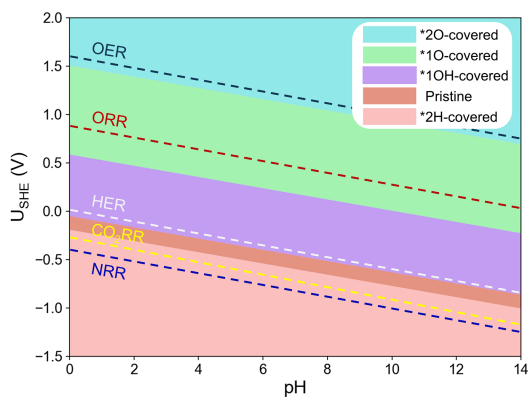
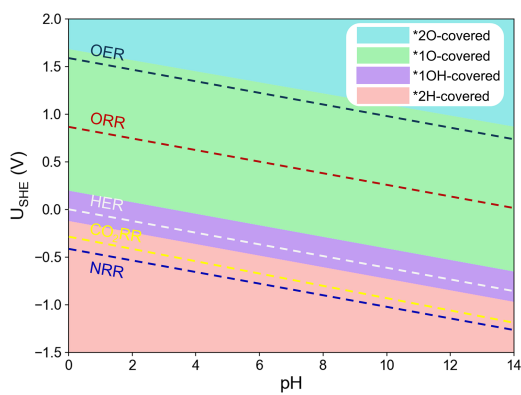
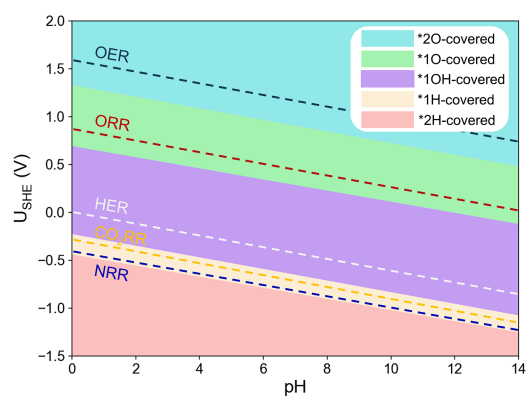
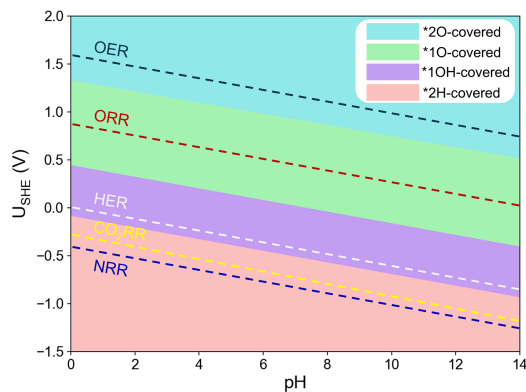
Fe-Ni-N_{3,4,5,6}-CFe-Ni-N_{1,2,3,4,5}-CFe-Ni-N_{1,2,3,4,6}-CFe-Ni-N_{1,2,3,5,6}-CFe-Ni-N_{1,2,4,5,6}-CFe-Ni-N_{1,3,4,5,6}-CFe-Ni-N_{2,3,4,5,6}-CFe-Ni-N_{1,2,3,4,5,6}-C

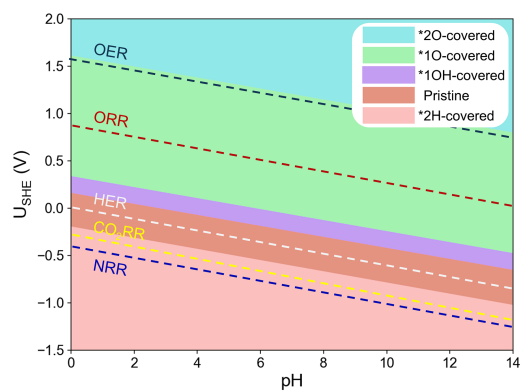
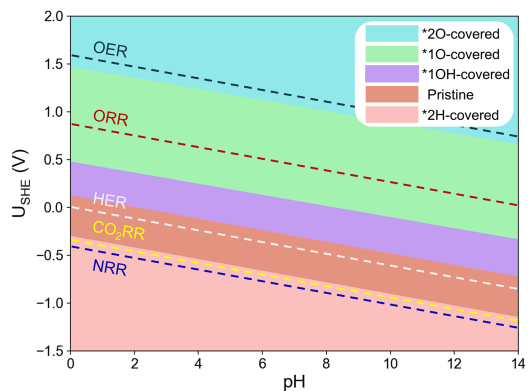
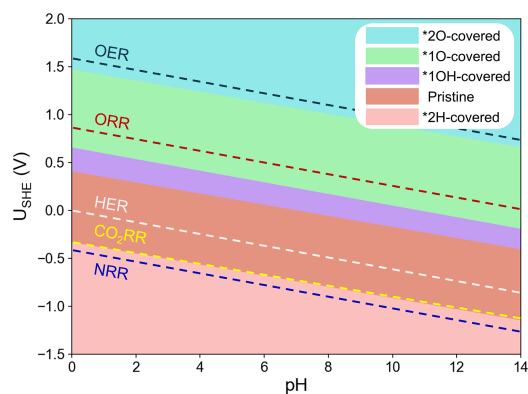
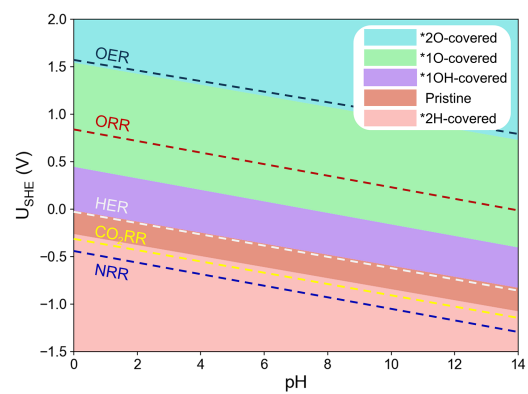
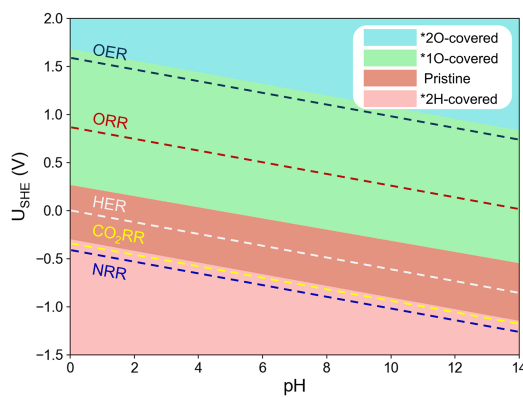
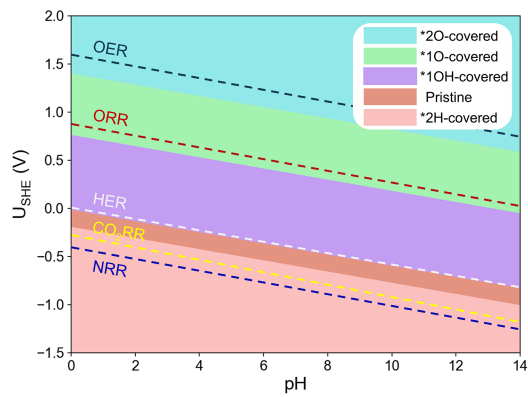
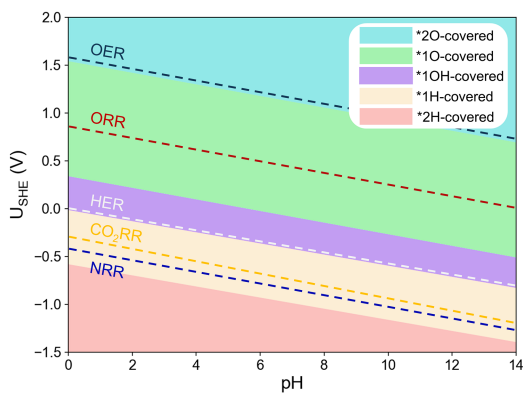
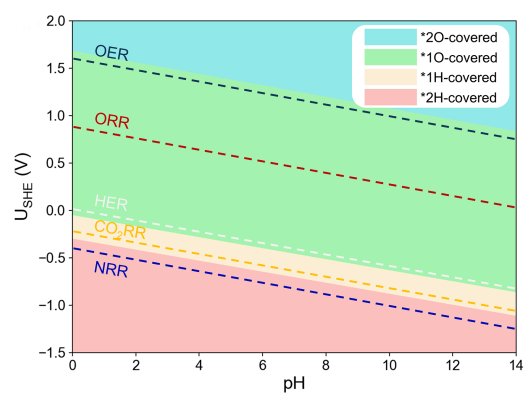


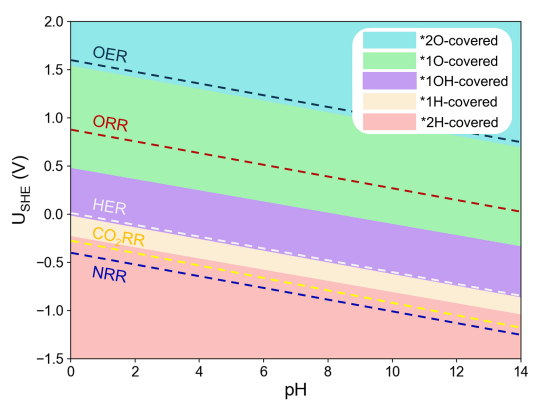
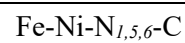
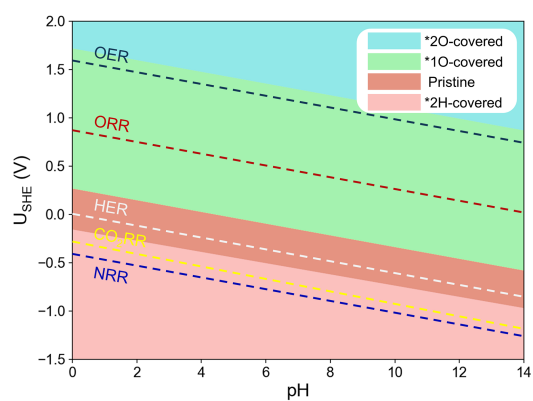
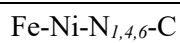
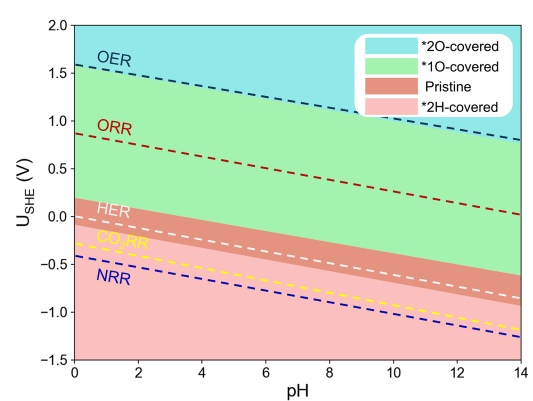
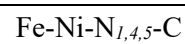
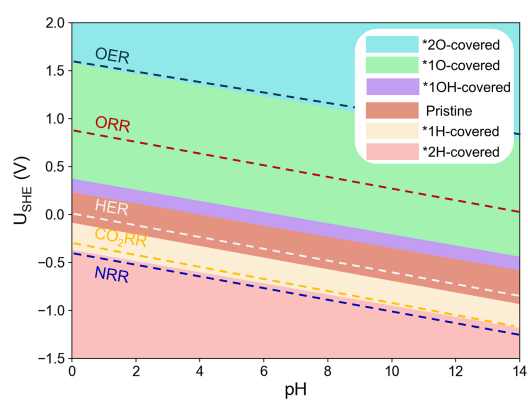
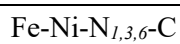
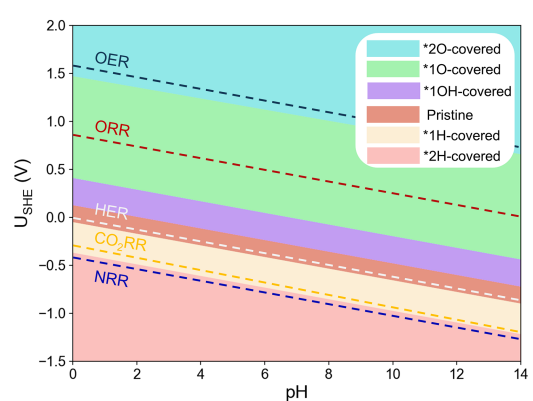
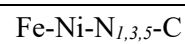
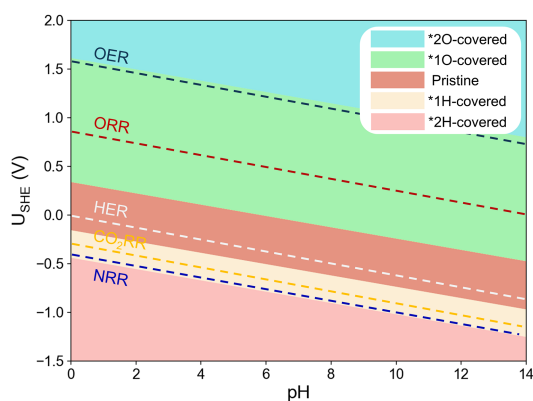
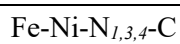
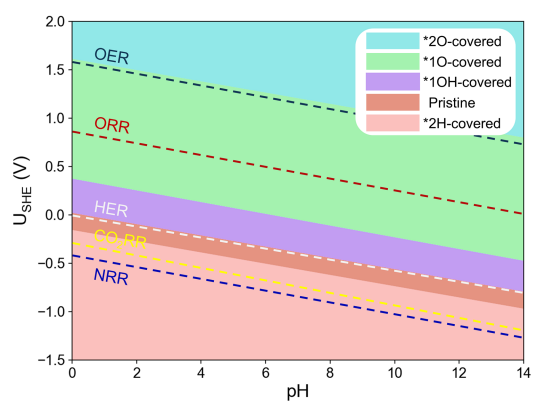
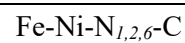
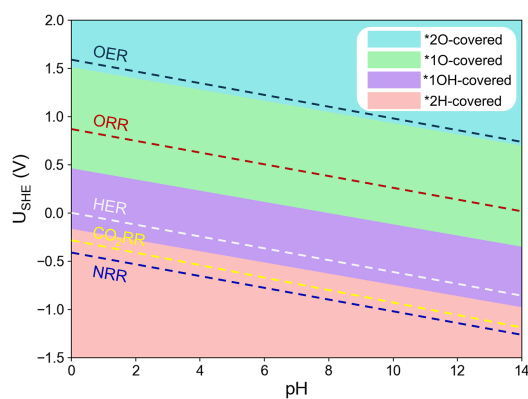
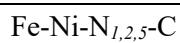
Supplementary Figure 1. Examples of the surface Pourbaix diagrams of DACs at pH=0. **a)** Two-side adsorption configurations of Fe-Ni-C, **b)** and Fe-Ni-N_{1,2,4}-C, where yellow, green, blue, silver, red, and light pink spheres represent Ni, Fe, N, C, O, and H respectively. **c)** The surface Pourbaix diagrams of Fe-Ni-C, **d)** and Fe-Ni-N_{1,2,4}-C.

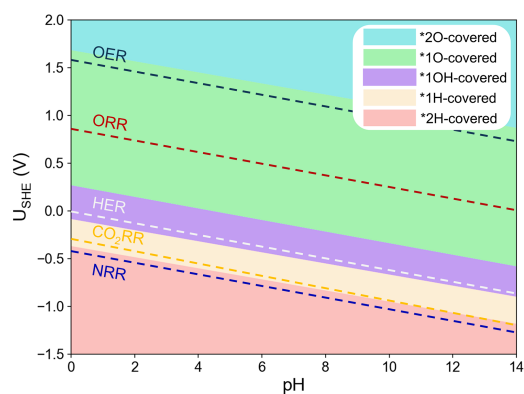
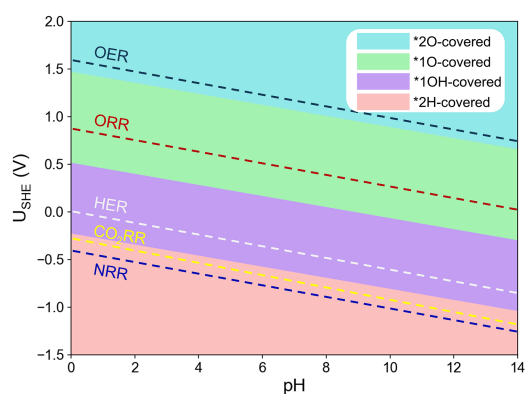
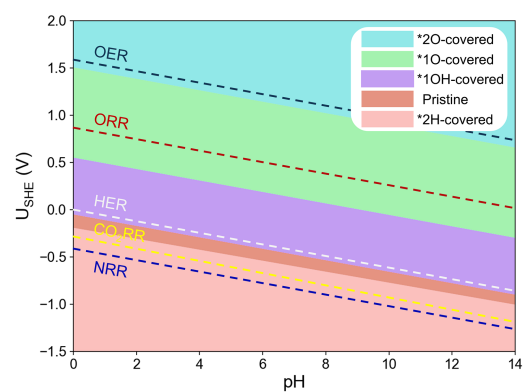
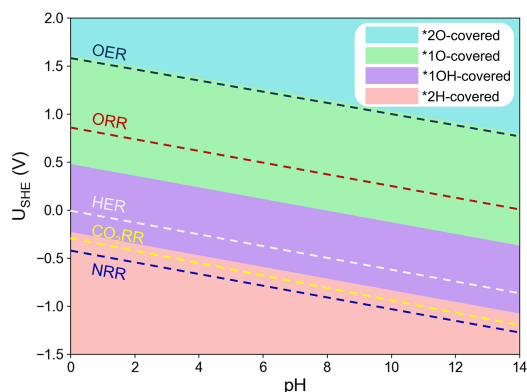
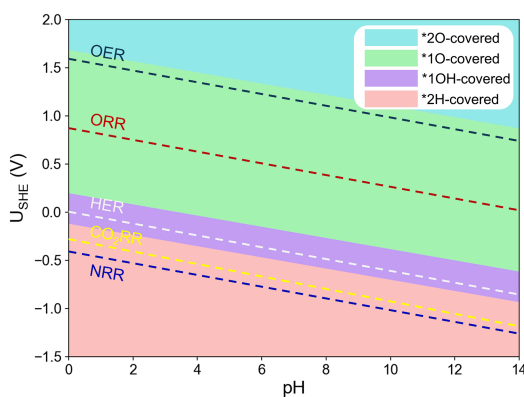
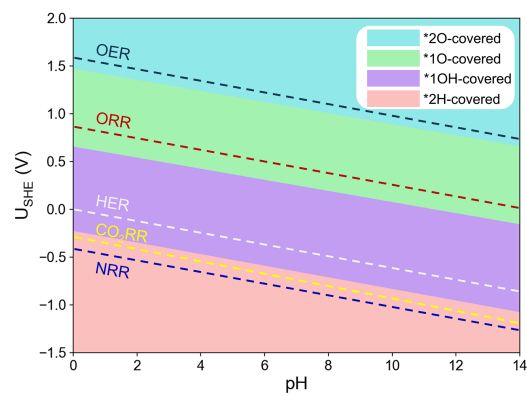
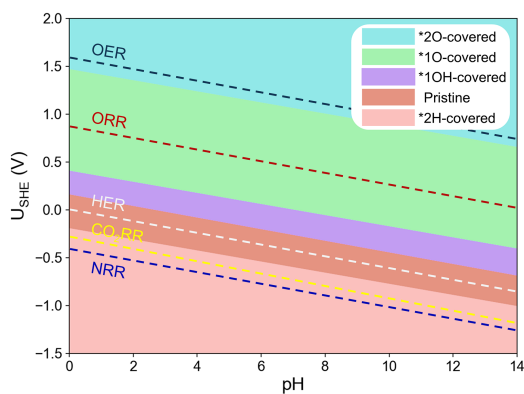
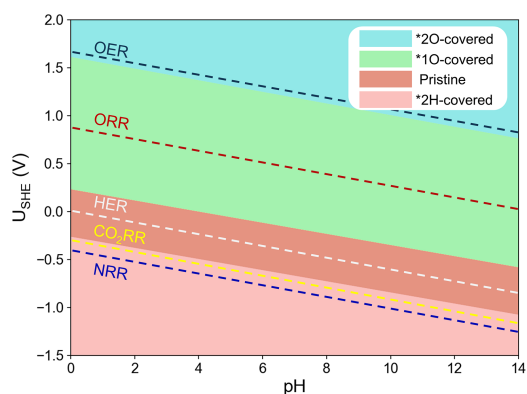
Supplementary Table 3. Surface Pourbaix diagrams of Fe-Ni-N_x-C in both acid and base.

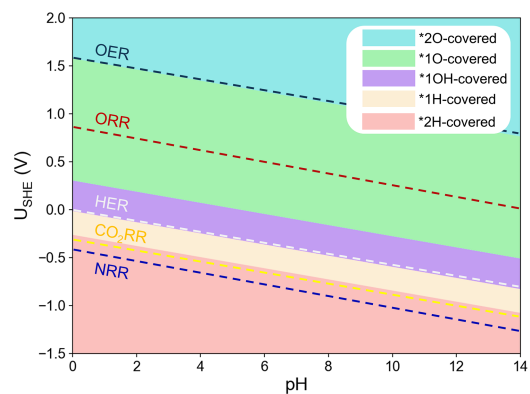
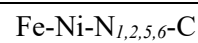
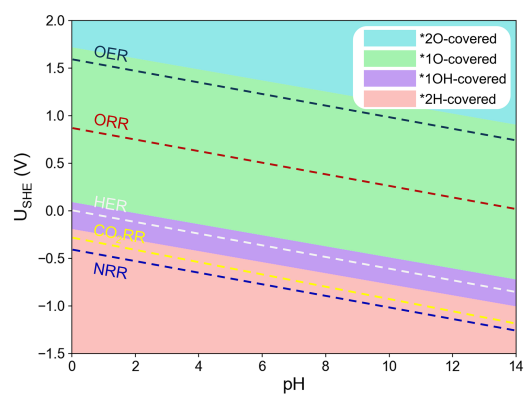
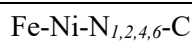
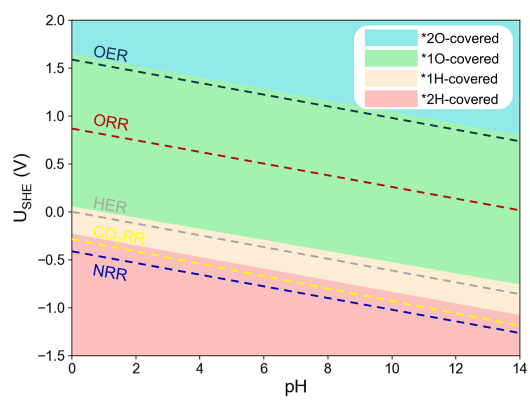
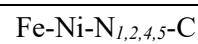
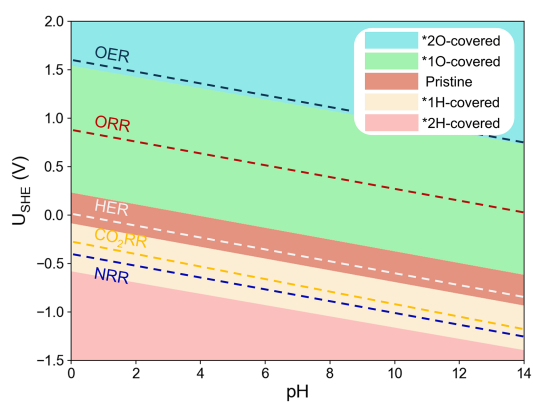
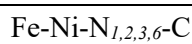
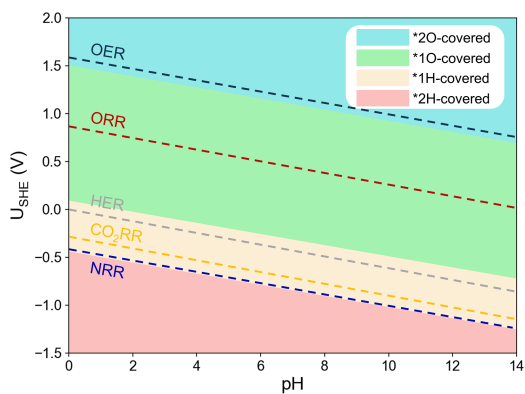
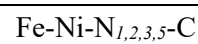
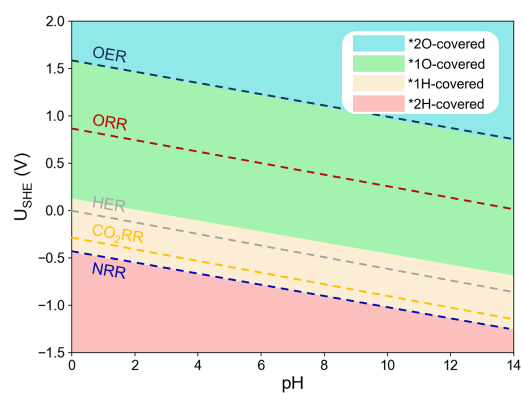
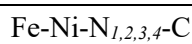
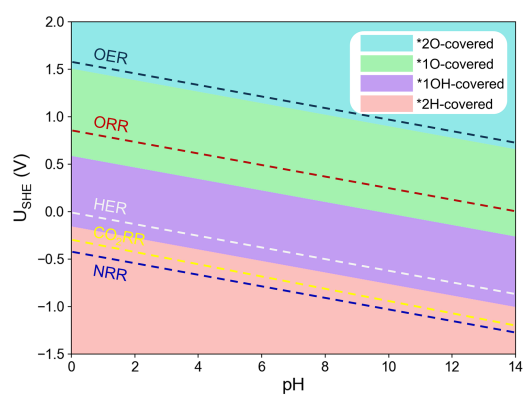
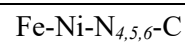
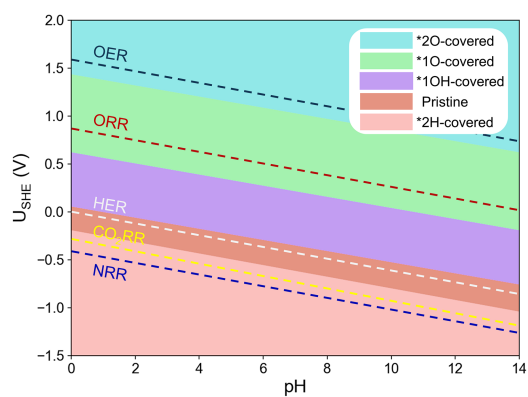
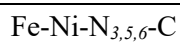


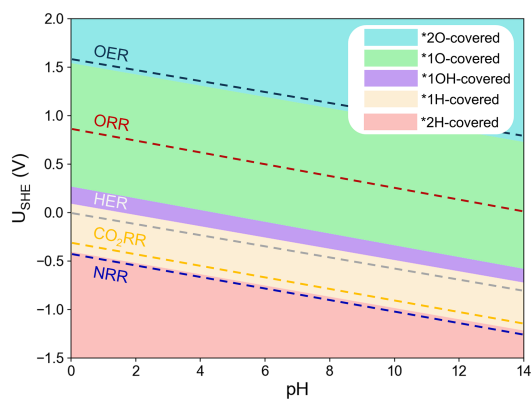
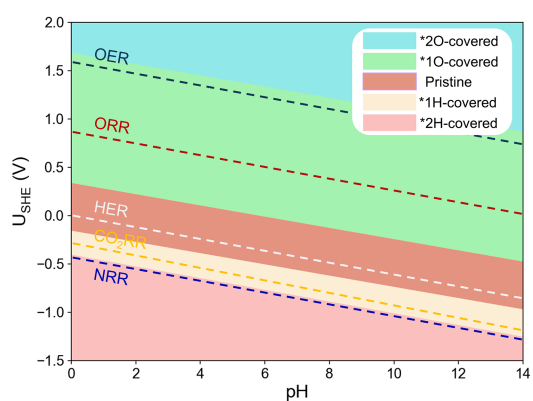
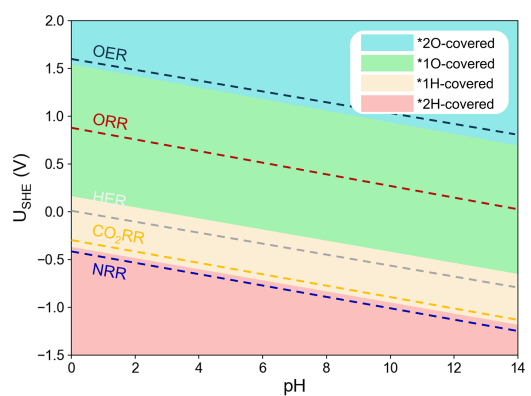
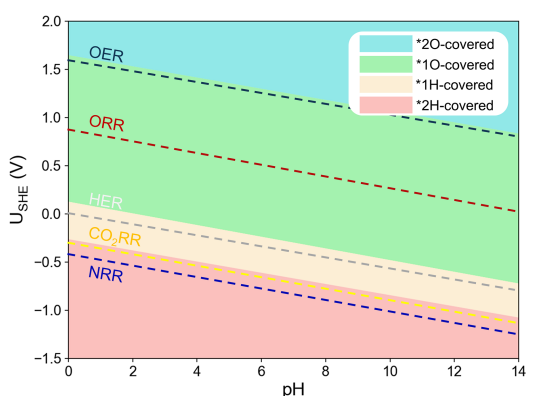
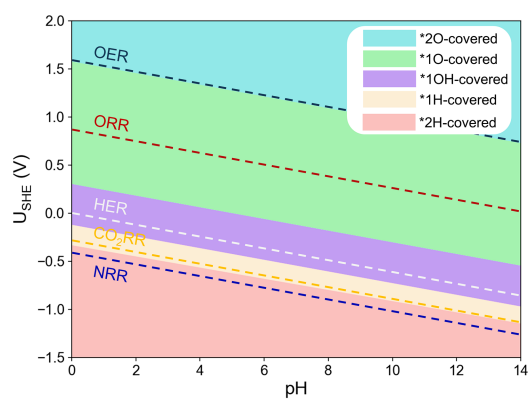
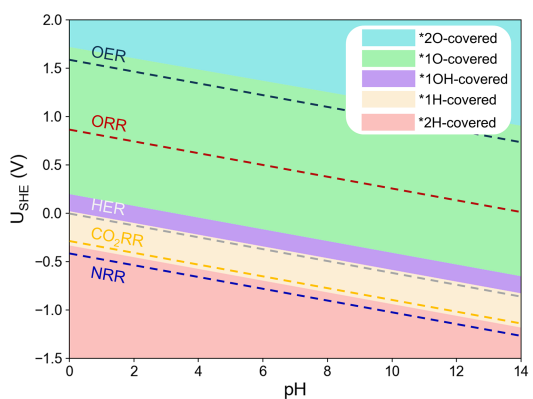
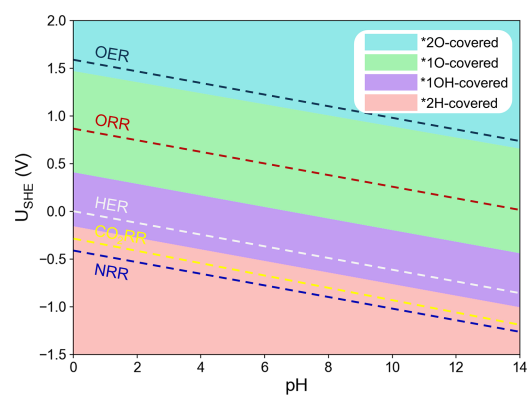
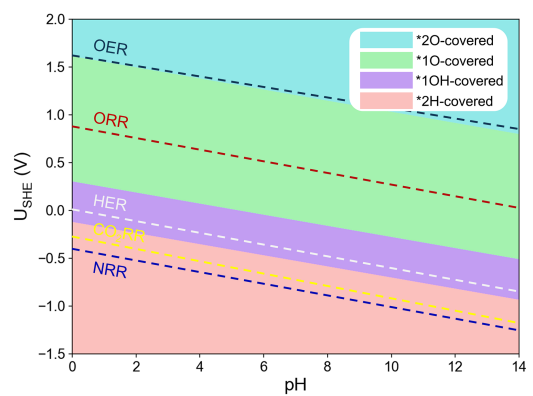
Fe-Ni-N_{1,3}-CFe-Ni-N_{1,4}-CFe-Ni-N_{1,5}-CFe-Ni-N_{1,6}-CFe-Ni-N_{2,3}-CFe-Ni-N_{2,4}-CFe-Ni-N_{2,5}-CFe-Ni-N_{2,6}-C

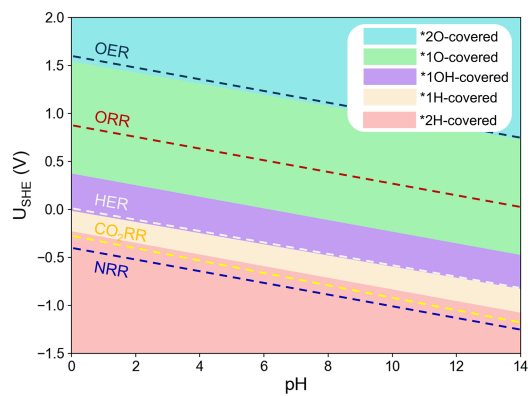
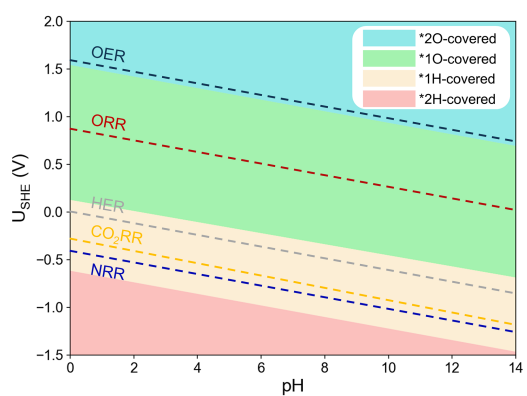
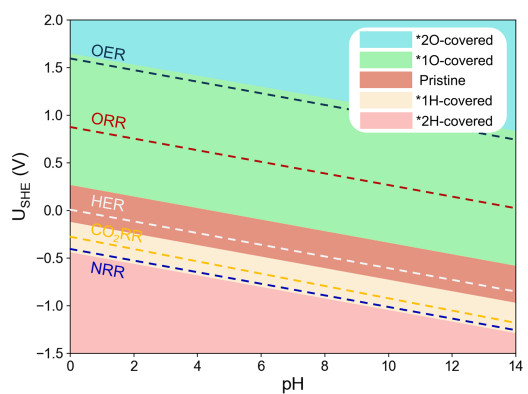
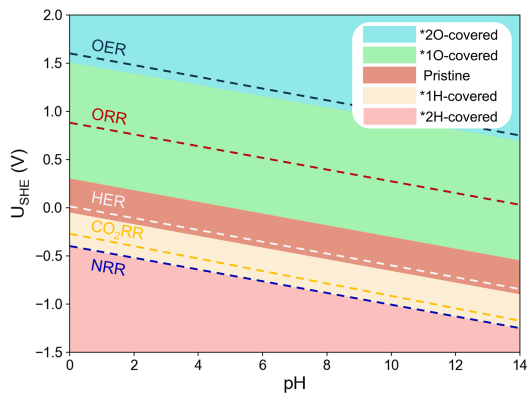
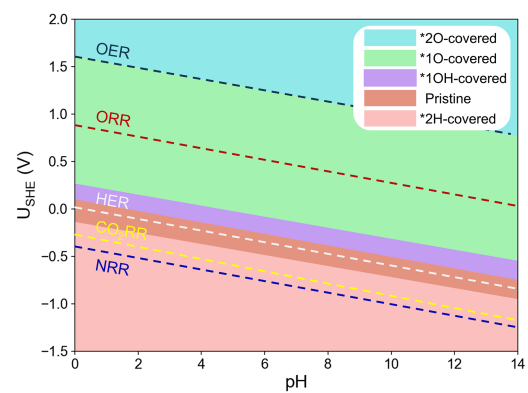
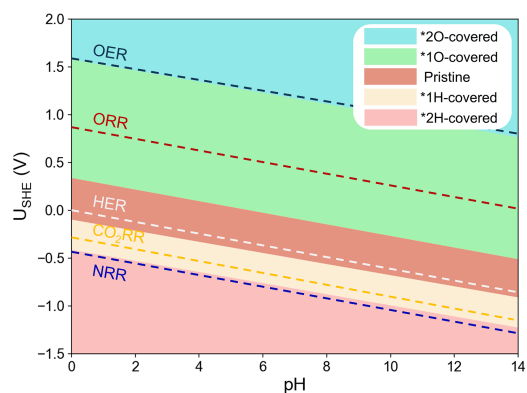
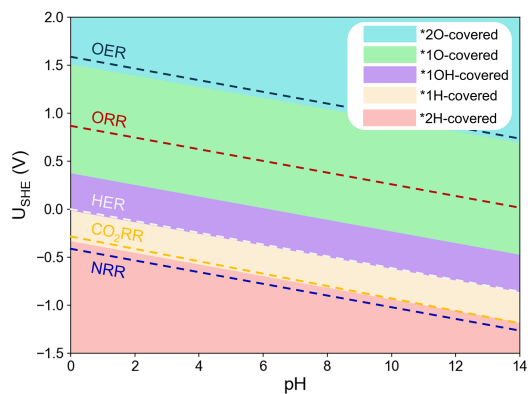
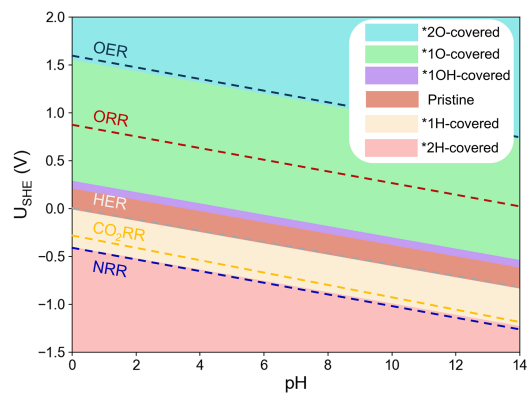
Fe-Ni-N_{3,4}-CFe-Ni-N_{3,5}-CFe-Ni-N_{3,6}-CFe-Ni-N_{4,5}-CFe-Ni-N_{4,6}-CFe-Ni-N_{5,6}-CFe-Ni-N_{I,2,3}-CFe-Ni-N_{I,2,4}-C



Fe-Ni-N_{2,3,4}-CFe-Ni-N_{2,3,5}-CFe-Ni-N_{2,3,6}-CFe-Ni-N_{2,4,5}-CFe-Ni-N_{2,4,6}-CFe-Ni-N_{2,5,6}-CFe-Ni-N_{3,4,5}-CFe-Ni-N_{3,4,6}-C



Fe-Ni-N_{1,3,4,5}-CFe-Ni-N_{1,3,4,6}-CFe-Ni-N_{1,3,5,6}-CFe-Ni-N_{1,4,5,6}-CFe-Ni-N_{2,3,4,5}-CFe-Ni-N_{2,3,4,6}-CFe-Ni-N_{2,3,5,6}-CFe-Ni-N_{2,4,5,6}-C

Fe-Ni-N_{3,4,5,6}-CFe-Ni-N_{1,2,3,4,5}-CFe-Ni-N_{1,2,3,4,6}-CFe-Ni-N_{1,2,3,5,6}-CFe-Ni-N_{1,2,4,5,6}-CFe-Ni-N_{1,3,4,5,6}-CFe-Ni-N_{2,3,4,5,6}-CFe-Ni-N_{1,2,3,4,5,6}-C

Supplementary Table 4. Surface states of Fe-Ni-N_x-C at the common potentials of HER, OER, ORR, CO₂RR, and NRR.

Models	Potential window (V)	HER (0 V)	OER (1.60 V)	ORR (0.78 V)	CO ₂ RR (-0.35 V)	NRR (-0.40V)
Fe-Ni-C	-0.36 ~ 0.28	Pristine	2O*	1O*	Pristine	2H*
Fe-Ni-N ₁ -C	-0.25 ~ 0.30	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N ₂ -C	-0.16 ~ 0.07	Pristine	2O*	2O*	2H*	2H*
Fe-Ni-N ₃ -C	-0.19 ~ 0.44	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N ₄ -C	-0.25 ~ 0.22	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N ₅ -C	-0.23 ~ 0.26	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N ₆ -C	-0.37 ~ 0.50	Pristine	2O*	1O*	Pristine	2H*
Fe-Ni-N _{1,2} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{1,3} -C	-0.19 ~ 0.40	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N _{1,4} -C	-0.24 ~ 0.28	Pristine	1O*	1O*	2H*	2H*
Fe-Ni-N _{1,5} -C	N/A	1HO*	2O*	1O*	1H*	2H*
Fe-Ni-N _{1,6} -C	-0.25 ~ 0.37	Pristine	1O*	1O*	2H*	2H*
Fe-Ni-N _{2,3} -C	-0.19 ~ -0.05	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{2,4} -C	N/A	1HO*	1O*	1O*	2H*	2H*
Fe-Ni-N _{2,5} -C	N/A	1HO*	2O*	1OH*	1H*	1H*
Fe-Ni-N _{2,6} -C	N/A	1HO*	2O*	1OH*	2H*	2H*
Fe-Ni-N _{3,4} -C	-0.10 ~ 0.15	Pristine	1O*	1O*	2H*	2H*
Fe-Ni-N _{3,5} -C	-0.29 ~ 0.12	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N _{3,6} -C	-0.35 ~ 0.39	Pristine	2O*	1O*	Pristine	2H*
Fe-Ni-N _{4,5} -C	-0.24 ~ 0.02	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N _{4,6} -C	-0.29 ~ 0.25	Pristine	1O*	1O*	2H*	2H*
Fe-Ni-N _{5,6} -C	-0.19 ~ -0.03	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{1,2,3} -C	N/A	1HO*	2O*	1O*	1H*	1H*
Fe-Ni-N _{1,2,4} -C	N/A	1O*	1O*	1O*	1H*	2H*
Fe-Ni-N _{1,2,5} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{1,2,6} -C	-0.16 ~ 0	Pristine	1O*	1O*	2H*	2H*
Fe-Ni-N _{1,3,4} -C	-0.16 ~ 0.33	Pristine	1O*	1O*	1H*	1H*

Fe-Ni-N _{1,3,5} -C	-0.08 ~ 0.13	Pristine	2O*	1O*	1H*	2H*
Fe-Ni-N _{1,3,6} -C	-0.08 ~ 0.22	Pristine	2O*	1O*	1H*	2H*
Fe-Ni-N _{1,4,5} -C	-0.08 ~ 0.18	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N _{1,4,6} -C	-0.17 ~ 0.28	Pristine	1O*	1O*	2H*	2H*
Fe-Ni-N _{1,5,6} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{2,3,4} -C	N/A	1HO*	2O*	1O*	1H*	2H*
Fe-Ni-N _{2,3,5} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{2,3,6} -C	-0.19 ~ -0.04	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{2,4,5} -C	N/A	1HO*	1O*	1O*	2H*	2H*
Fe-Ni-N _{2,4,6} -C	N/A	1HO*	1O*	1O*	2H*	2H*
Fe-Ni-N _{2,5,6} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{3,4,5} -C	-0.19 ~ 0.16	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N _{3,4,6} -C	-0.26 ~ 0.25	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N _{3,5,6} -C	-0.18 ~ 0.05	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N _{4,5,6} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{1,2,3,4} -C	N/A	1H*	2O*	1O*	1H*	1H*
Fe-Ni-N _{1,2,3,5} -C	N/A	1H*	2O*	1O*	1H*	1H*
Fe-Ni-N _{1,2,3,6} -C	-0.07 ~ 0.23	Pristine	2O*	1O*	1H*	1H*
Fe-Ni-N _{1,2,4,5} -C	N/A	1H*	1O*	1O*	2H*	2H*
Fe-Ni-N _{1,2,4,6} -C	N/A	1HO*	1O*	1O*	2H*	2H*
Fe-Ni-N _{1,2,5,6} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{1,3,4,5} -C	N/A	1H*	2O*	1O*	1H*	2H*
Fe-Ni-N _{1,3,4,6} -C	-0.17 ~ 0.33	Pristine	1O*	1O*	1H*	2H*
Fe-Ni-N _{1,3,5,6} -C	N/A	1H*	2O*	1O*	1H*	2H*
Fe-Ni-N _{1,4,5,6} -C	N/A	1H*	1O*	1O*	2H*	2H*
Fe-Ni-N _{2,3,4,5} -C	N/A	1HO*	2O*	1O*	1H*	2H*
Fe-Ni-N _{2,3,4,6} -C	N/A	1H*	1O*	1O*	1H*	2H*
Fe-Ni-N _{2,3,5,6} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{2,4,5,6} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{3,4,5,6} -C	N/A	1HO*	2O*	1O*	2H*	2H*
Fe-Ni-N _{1,2,3,4,5} -C	N/A	1H*	2O*	1O*	1H*	1H*

Fe-Ni-N _{1,2,3,4,6} -C	-0.12 ~ 0.28	Pristine	1O*	1O*	1H*	1H*
Fe-Ni-N _{1,2,3,5,6} -C	-0.04 ~ 0.17	Pristine	2O*	1O*	1H*	1H*
Fe-Ni-N _{1,2,4,5,6} -C	-0.14 ~ 0.10	Pristine	2O*	1O*	2H*	2H*
Fe-Ni-N _{1,3,4,5,6} -C	-0.10 ~ 0.35	Pristine	2O*	1O*	1H*	2H*
Fe-Ni-N _{2,3,4,5,6} -C	N/A	1HO*	2O*	1O*	1H*	2H*
Fe-Ni-N _{1,2,3,4,5,6} -C	-0.01 ~ 0.21	Pristine	2O*	1O*	1H*	2H*