**Supplementary Figure 1** Surface Pourbaix diagrams of Fe-Ni-N*x*-C at pH = 0

|  |  |
| --- | --- |
| Fe-Ni-C | Fe-Ni-N*1*-C |
|  |  |
| Fe-Ni-N*2*-C | Fe-Ni-N*3*-C |
|  |  |
| Fe-Ni-N*4*-C | Fe-Ni-N*5*-C |
|  |  |
| Fe-Ni-N*6*-C | Fe-Ni-N*1,2*-C |
|  |  |

|  |  |
| --- | --- |
| Fe-Ni-N*1,3*-C | Fe-Ni-N*1,4*-C |
|  |  |
| Fe-Ni-N*1,5*-C | Fe-Ni-N*1,6*-C |
|  |  |
| Fe-Ni-N*2,3*-C | Fe-Ni-N*2,4*-C |
|  |  |
| Fe-Ni-N*2,5*-C | Fe-Ni-N*2,6*-C |
|  |  |

|  |  |
| --- | --- |
| Fe-Ni-N*3,4*-C | Fe-Ni-N*3,5*-C |
|  |  |
| Fe-Ni-N*3,6*-C | Fe-Ni-N*4,5*-C |
|  |  |
| Fe-Ni-N*4,6*-C | Fe-Ni-N*5,6*-C |
|  |  |
| Fe-Ni-N*1,2,3*-C | Fe-Ni-N*1,2,4*-C |
|  |  |

|  |  |
| --- | --- |
| Fe-Ni-N*1,2,5*-C | Fe-Ni-N*1,2,6*-C |
|  |  |
| Fe-Ni-N*1,3,4*-C | Fe-Ni-N*1,3,5*-C |
|  |  |
| Fe-Ni-N*1,3,6*-C | Fe-Ni-N*1,4,5*-C |
|  |  |
| Fe-Ni-N*1,4,6*-C | Fe-Ni-N*1,5,6*-C |
|  |  |

|  |  |
| --- | --- |
| Fe-Ni-N*2,3,4*-C | Fe-Ni-N*2,3,5*-C |
|  |  |
| Fe-Ni-N*2,3,6*-C | Fe-Ni-N*2,4,5*-C |
|  |  |
| Fe-Ni-N*2,4,6*-C | Fe-Ni-N*2,5,6*-C |
|  |  |
| Fe-Ni-N*3,4,5*-C | Fe-Ni-N*3,4,6*-C |
|  |  |

|  |  |
| --- | --- |
| Fe-Ni-N*3,5,6*-C | Fe-Ni-N*4,5,6*-C |
|  |  |
| Fe-Ni-N*1,2,3,4*-C | Fe-Ni-N*1,2,3,5*-C |
|  |  |
| Fe-Ni-N*1,2,3,6*-C | Fe-Ni-N*1,2,4,5*-C |
|  |  |
| Fe-Ni-N*1,2,4,6*-C | Fe-Ni-N*1,2,5,6*-C |
|  |  |

|  |  |
| --- | --- |
| Fe-Ni-N*1,3,4,5*-C | Fe-Ni-N*1,3,4,6*-C |
|  |  |
| Fe-Ni-N*1,3,5,6*-C | Fe-Ni-N*1,4,5,6*-C |
|  |  |
| Fe-Ni-N*2,3,4,5*-C | Fe-Ni-N*2,3,4,6*-C |
|  |  |
| Fe-Ni-N*2,3,5,6*-C | Fe-Ni-N*2,4,5,6*-C |
|  |  |

|  |  |
| --- | --- |
| Fe-Ni-N*3,4,5,6*-C | Fe-Ni-N*1,2,3,4,5*-C |
|  |  |
| Fe-Ni-N*1,2,3,4,6*-C | Fe-Ni-N*1,2,3,5,6*-C |
|  |  |
| Fe-Ni-N*1,2,4,5,6*-C | Fe-Ni-N*1,3,4,5,6*-C |
|  |  |
| Fe-Ni-N*2,3,4,5,6*-C | Fe-Ni-N*1,2,3,4,5,6*-C |
|  |  |

**Supplementary Figure 2** Surface Pourbaix diagrams of Fe-Ni-N*x*-C in both acid and base.

|  |  |
| --- | --- |
| Fe-Ni-C | Fe-Ni-N*1*-C |
|  |  |
| Fe-Ni-N*2*-C | Fe-Ni-N*3*-C |
|  |  |
| Fe-Ni-N*4*-C | Fe-Ni-N*5*-C |
|  |  |
| Fe-Ni-N*6*-C | Fe-Ni-N*1,2*-C |
|  |  |
| Fe-Ni-N*1,3*-C | Fe-Ni-N*1,4*-C |
|  |  |
| Fe-Ni-N*1,5*-C | Fe-Ni-N*1,6*-C |
|  |  |
| Fe-Ni-N*2,3*-C | Fe-Ni-N*2,4*-C |
|  |  |
| Fe-Ni-N*2,5*-C | Fe-Ni-N*2,6*-C |
|  |  |
| Fe-Ni-N*3,4*-C | Fe-Ni-N*3,5*-C |
|  |  |
| Fe-Ni-N*3,6*-C | Fe-Ni-N*4,5*-C |
|  |  |
| Fe-Ni-N*4,6*-C | Fe-Ni-N*5,6*-C |
|  |  |
| Fe-Ni-N*1,2,3*-C | Fe-Ni-N*1,2,4*-C |
|  |  |
| Fe-Ni-N*1,2,5*-C | Fe-Ni-N*1,2,6*-C |
|  |  |
| Fe-Ni-N*1,3,4*-C | Fe-Ni-N*1,3,5*-C |
|  |  |
| Fe-Ni-N*1,3,6*-C | Fe-Ni-N*1,4,5*-C |
|  |  |
| Fe-Ni-N*1,4,6*-C | Fe-Ni-N*1,5,6*-C |
|  |  |
| Fe-Ni-N*2,3,4*-C | Fe-Ni-N*2,3,5*-C |
|  |  |
| Fe-Ni-N*2,3,6*-C | Fe-Ni-N*2,4,5*-C |
|  |  |
| Fe-Ni-N*2,4,6*-C | Fe-Ni-N*2,5,6*-C |
|  |  |
| Fe-Ni-N*3,4,5*-C | Fe-Ni-N*3,4,6*-C |
|  |  |
| Fe-Ni-N*3,5,6*-C | Fe-Ni-N*4,5,6*-C |
|  |  |
| Fe-Ni-N*1,2,3,4*-C | Fe-Ni-N*1,2,3,5*-C |
|  |  |
| Fe-Ni-N*1,2,3,6*-C | Fe-Ni-N*1,2,4,5*-C |
|  |  |
| Fe-Ni-N*1,2,4,6*-C | Fe-Ni-N*1,2,5,6*-C |
|  |  |
| Fe-Ni-N*1,3,4,5*-C | Fe-Ni-N*1,3,4,6*-C |
|  |  |
| Fe-Ni-N*1,3,5,6*-C | Fe-Ni-N*1,4,5,6*-C |
|  |  |
| Fe-Ni-N*2,3,4,5*-C | Fe-Ni-N*2,3,4,6*-C |
|  |  |
| Fe-Ni-N*2,3,5,6*-C | Fe-Ni-N*2,4,5,6*-C |
|  |  |
| Fe-Ni-N*3,4,5,6*-C | Fe-Ni-N*1,2,3,4,5*-C |
|  |  |
| Fe-Ni-N*1,2,3,4,6*-C | Fe-Ni-N*1,2,3,5,6*-C |
|  |  |
| Fe-Ni-N*1,2,4,5,6*-C | Fe-Ni-N*1,3,4,5,6*-C |
|  |  |
| Fe-Ni-N*2,3,4,5,6*-C | Fe-Ni-N*1,2,3,4,5,6*-C |
|  |  |

**Supplementary Table 1** 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) | | CO2RR  (-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 | 0 | 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 | 0 | 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 | 0 | 1HO\* | | 1O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*2,5*-C | 0 | 1HO\* | | 2O\* | | 1OH\* | 1H\* | 1H\* |
| Fe-Ni-N*2,6*-C | 0 | 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 | 0 | 1HO\* | | 2O\* | | 1O\* | 1H\* | 1H\* |
| Fe-Ni-N*1,2,4*-C | 0 | 1O\* | | 1O\* | | 1O\* | 1H\* | 2H\* |
| Fe-Ni-N*1,2,5*-C | 0 | 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 | 0 | 1HO\* | | 2O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*2,3,4*-C | 0 | 1HO\* | | 2O\* | | 1O\* | 1H\* | 2H\* |
| Fe-Ni-N*2,3,5*-C | 0 | 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 | 0 | 1HO\* | | 1O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*2,4,6*-C | 0 | 1HO\* | | 1O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*2,5,6*-C | 0 | 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 | 0 | 1HO\* | | 2O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*1,2,3,4*-C | 0 | 1H\* | | 2O\* | | 1O\* | 1H\* | 1H\* |
| Fe-Ni-N*1,2,3,5*-C | 0 | 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 | 0 | 1H\* | | 1O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*1,2,4,6*-C | 0 | 1HO\* | | 1O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*1,2,5,6*-C | 0 | 1HO\* | | 2O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*1,3,4,5*-C | 0 | 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 | 0 | 1H\* | | 2O\* | | 1O\* | 1H\* | 2H\* |
| Fe-Ni-N*1,4,5,6*-C | 0 | 1H\* | | 1O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*2,3,4,5*-C | 0 | 1HO\* | | 2O\* | | 1O\* | 1H\* | 2H\* |
| Fe-Ni-N*2,3,4,6*-C | 0 | 1H\* | | 1O\* | | 1O\* | 1H\* | 2H\* |
| Fe-Ni-N*2,3,5,6*-C | 0 | 1HO\* | | 2O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*2,4,5,6*-C | 0 | 1HO\* | | 2O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*3,4,5,6*-C | 0 | 1HO\* | | 2O\* | | 1O\* | 2H\* | 2H\* |
| Fe-Ni-N*1,2,3,4,5*-C | 0 | 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 | 0 | 1HO\* | | 2O\* | | 1O\* | 1H\* | 2H\* |
| Fe-Ni-N*1,2,3,4,5,6*-C | 0 | 1H\* | | 2O\* | | 1O\* | 1H\* | 2H\* |