Supporting Information

Boosting screening of non-equiatomic high-entropy electrocatalysts by inverse design via active graph learning

Jun Zhang[a,b,c], Weifeng Su[d], Yingying Li[a,b], Shuaishuai Man[e], Lifeng Liu[d], Shijun Zhao[c,*], Guang-Jie Xia[a,*].

[a]Center for Intelligent Computing and School of Physical Sciences, Great Bay University, Dongguan 523000, China

[b]Department of Chemical Physics, School of Chemistry and Materials Science, University of Science and Technology of China, Hefei 230026, China

[c]Department of Mechanical Engineering, City University of Hong Kong, Hong Kong, China

[d]Songshan Lake Materials Laboratory, Dongguan 523808, China

[e]School of Environment and Ecology, Jiangnan University, Wuxi 214122, China

[*]Corresponding Authors: shijzhao@cityu.edu.hk (S.Z.); xiagj@gbu.edu.cn (G.-J.X.)

The supporting information includes the supporting notes and figures.

Supporting Note 1: AGAT model code structure.

```
==========================================================
PotentialModel(
  (gat_layers): ModuleList(
    (0): Layer(
      (layer): Linear(in_features=6, out_features=300, bias=True)
      (leaky_relu1): LeakyReLU(negative_slope=0.2) (leaky_relu2):
      LeakyReLU(negative_slope=0.2)
    )
    (1-4): 4 x Layer(
      (layer):     Linear(in_features=300,      out_features=300,
bias=True)
      (leaky_relu1): LeakyReLU(negative_slope=0.2)
      (leaky_relu2): LeakyReLU(negative_slope=0.2)
    )
  )
  (energy_readout_layers): ModuleList(
    (0): Linear(in_features=300, out_features=300, bias=True)
    (1): LeakyReLU(negative_slope=0.2)
    (2): Linear(in_features=300, out_features=100, bias=True)
    (3): LeakyReLU(negative_slope=0.2)
    (4): Linear(in_features=100, out_features=50, bias=True)
    (5): LeakyReLU(negative_slope=0.2)
    (6): Linear(in_features=50, out_features=30, bias=True)
    (7): LeakyReLU(negative_slope=0.2)
    (8): Linear(in_features=30, out_features=10, bias=True)
    (9): Linear(in_features=10, out_features=3, bias=True)
    (10): Linear(in_features=3, out_features=1, bias=True)
  )
  (force_readout_layers): ModuleList(
    (0): Linear(in_features=300, out_features=300, bias=True)
    (1): LeakyReLU(negative_slope=0.2)
    (2): Linear(in_features=300, out_features=100, bias=True)
    (3): LeakyReLU(negative_slope=0.2)
    (4): Linear(in_features=100, out_features=50, bias=True)
    (5): LeakyReLU(negative_slope=0.2)
    (6): Linear(in_features=50, out_features=30, bias=True)
    (7): Linear(in_features=30, out_features=10, bias=True)
    (8): Linear(in_features=10, out_features=3, bias=True)
  )
  (stress_readout_layers): ModuleList(
    (0): Linear(in_features=300, out_features=300, bias=True)
    (1): LeakyReLU(negative_slope=0.2)
    (2): Linear(in_features=300, out_features=6, bias=True)
```

```
  )
  (u2e): Linear(in_features=6, out_features=300, bias=False)
)
============================================================
```

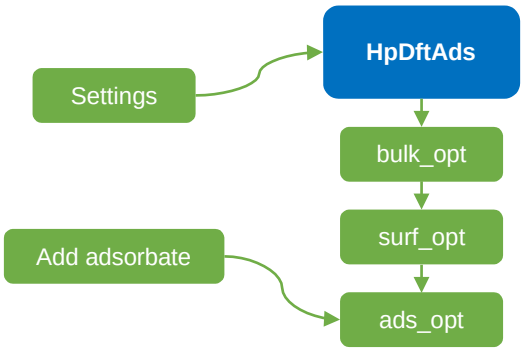

Figure S1 Workflow of high-throughput DFT calculations. The HpDftAds, surf, ads, and opt denote high-throughput DFT calculations, surface, adsorption, and optimization, respectively.

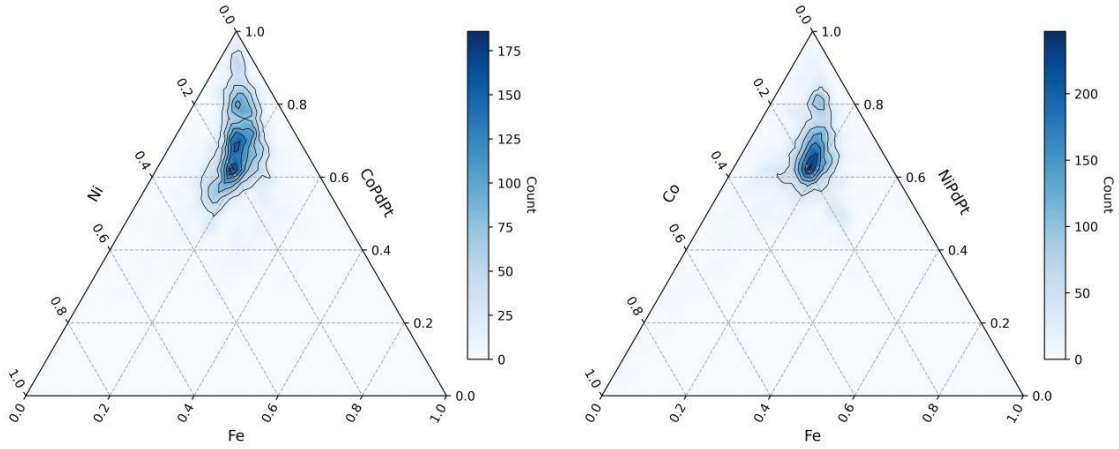

Figure S2 Distribution of all compositions calculated by DFT in all active loops.

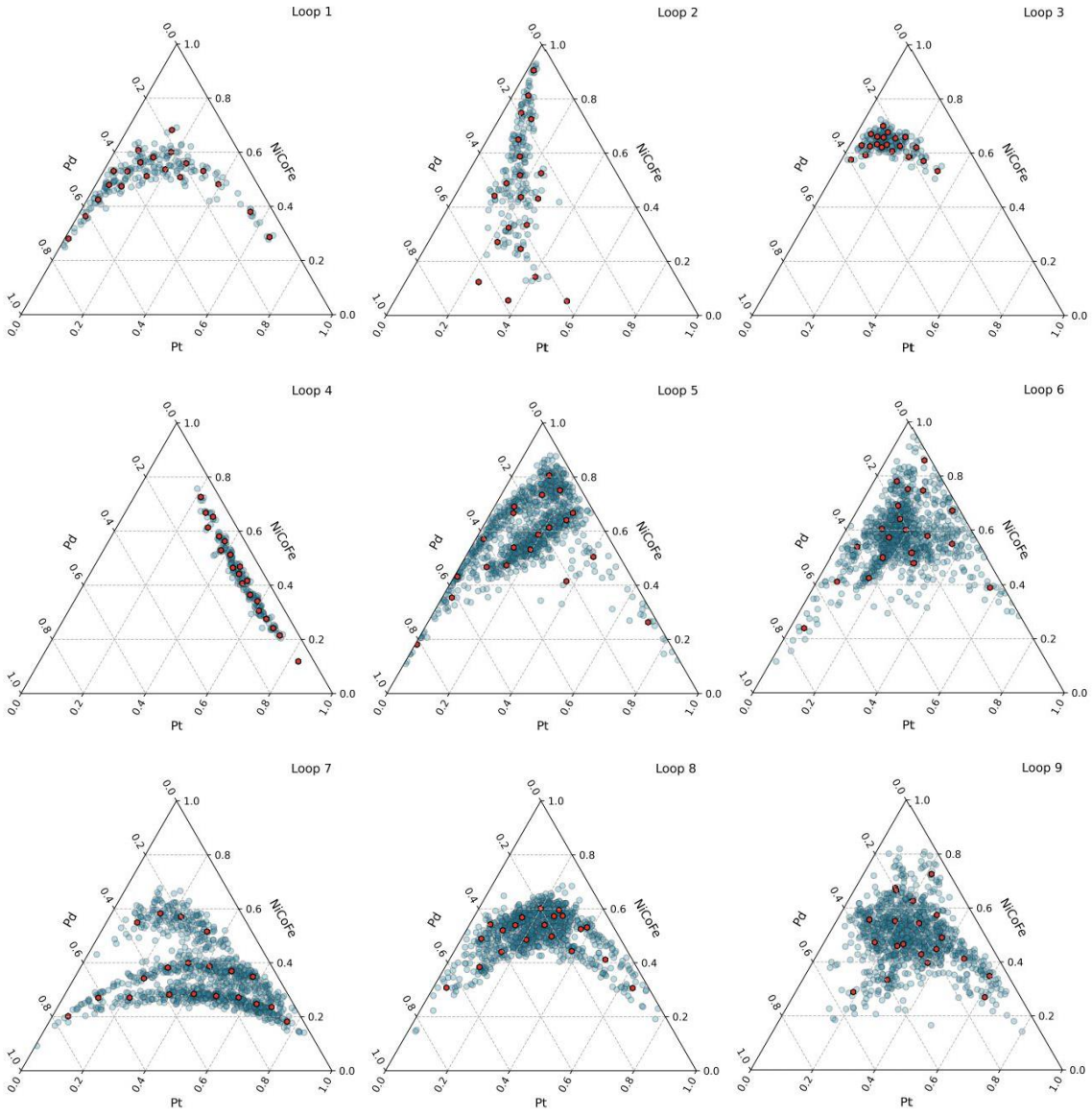

Figure S3 Composition recommendations of cycle 1-9. Blue cycles and red hexagons denote CGAN recommendations and 20 centers of KNN classifications.

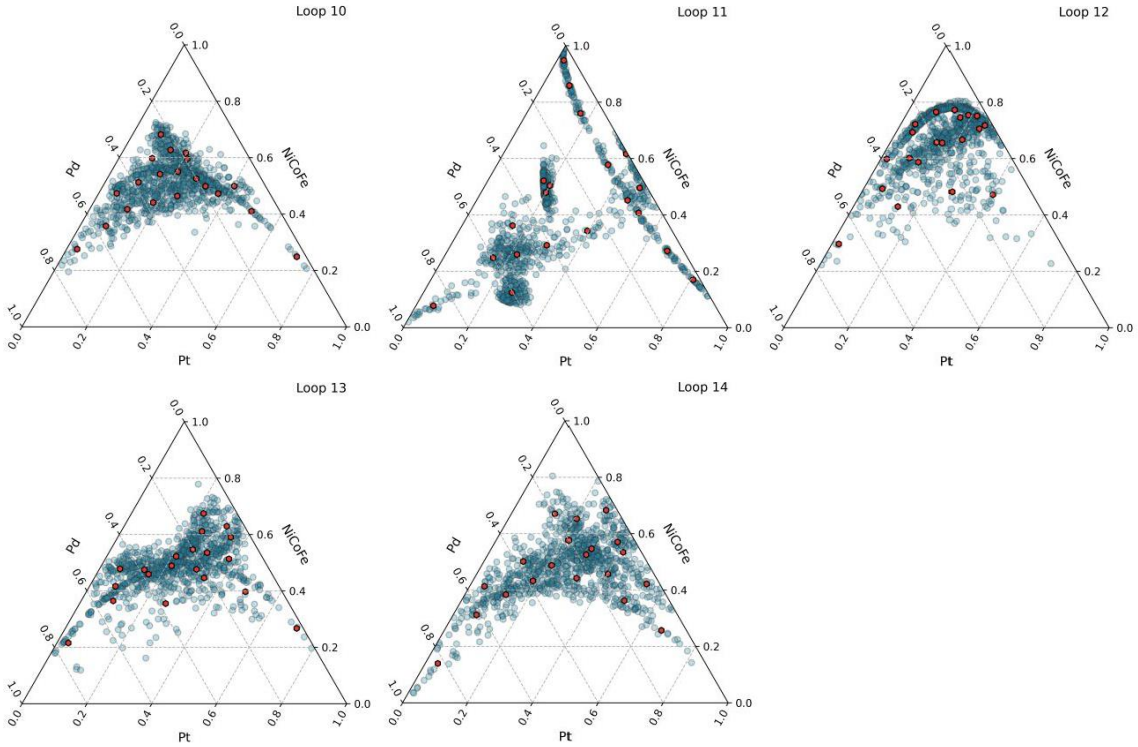

Figure S4 Composition recommendations of cycle 10-14. Blue cycles and red hexagons denote CGAN recommendations and 20 centers of KNN classifications. Note that cycle 11 experienced ill-convergence of geometrical relaxation, which was fixed in following cycles.

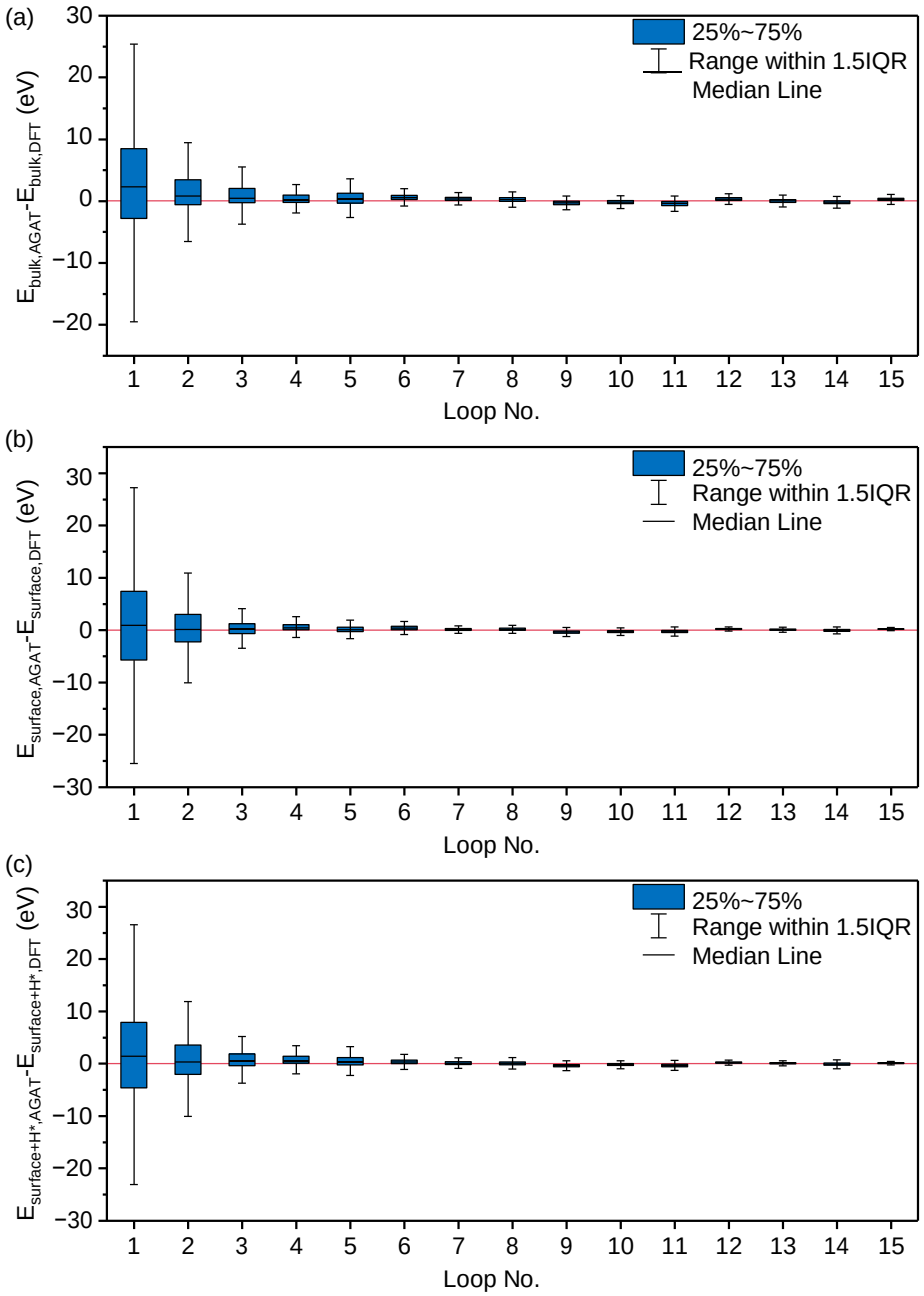

Figure S5 Energy difference between DFT and AGAT predictions of (a) bulk, (b) clean surface, and (c) adsorption structures.

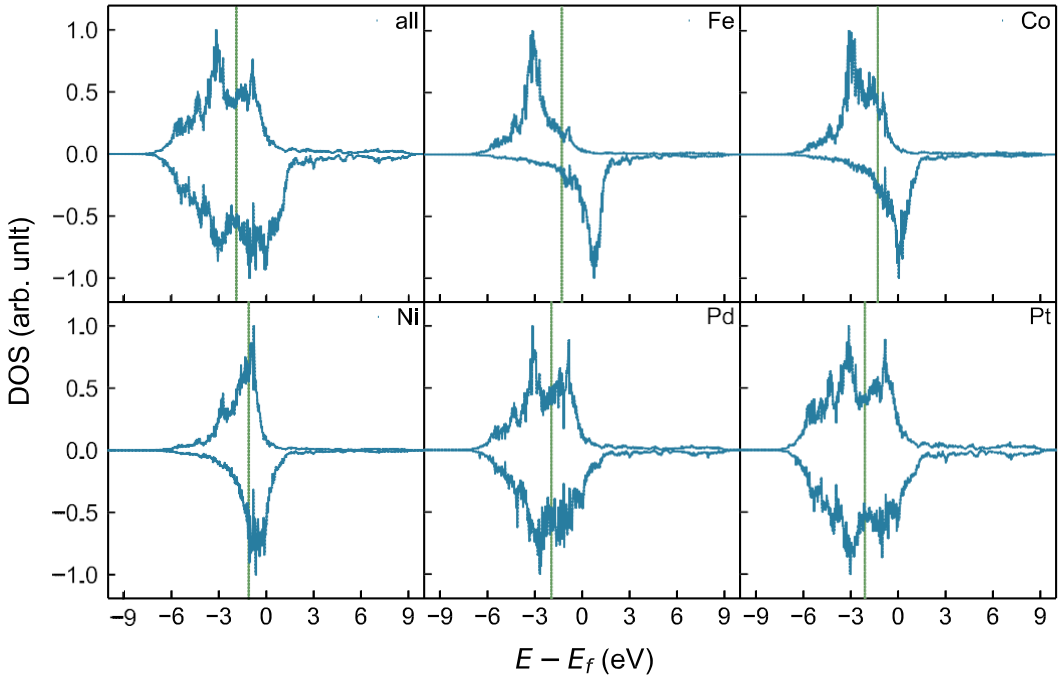

Figure S6 Projected density of city of surface atoms of $Ni_6Co_8Fe_{12}Pd_6Pt_{64}$. Green line denotes the energy level of *d*-band center.

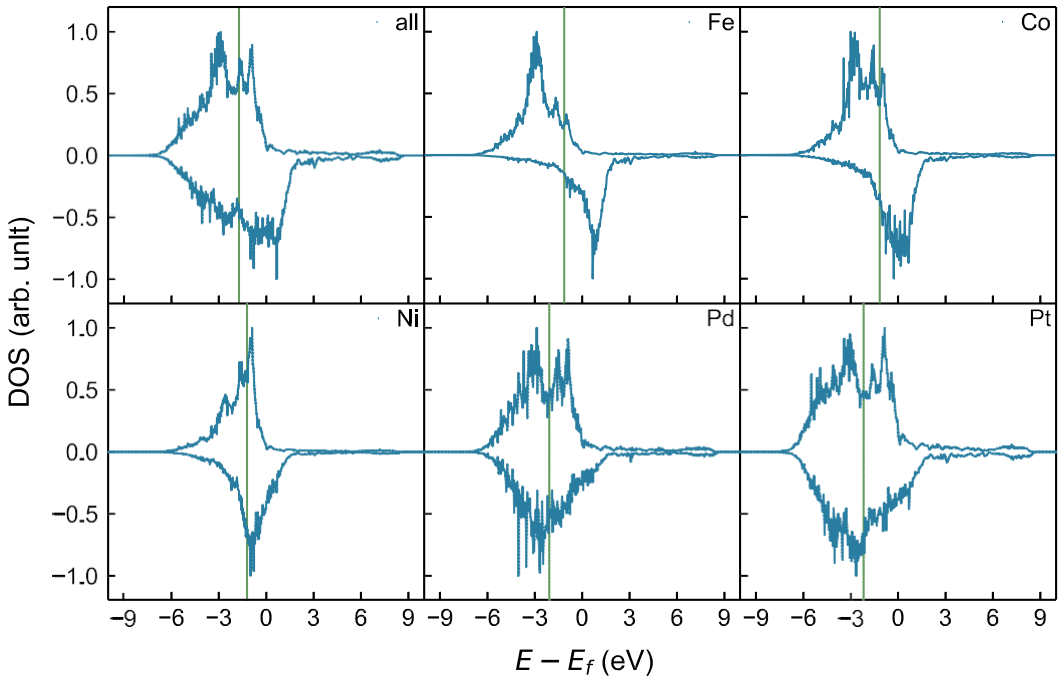

Figure S7 Projected density of states of surface atoms of $Ni_{12}Co_{12}Fe_{21}Pd_8Pt_{43}$. Green line denotes the energy level of *d*-band center.

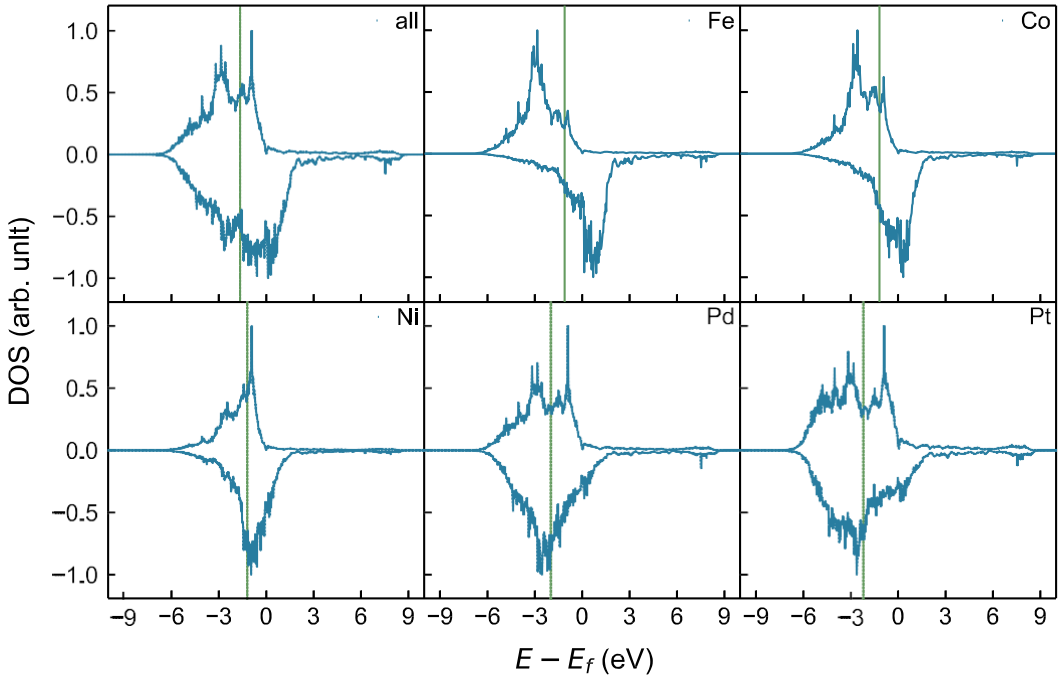

Figure S8 Projected density of states of surface atoms of $Ni_{15}Co_{15}Fe_{19}Pd_{17}Pt_{30}$. Green line denotes the energy level of *d*-band center.

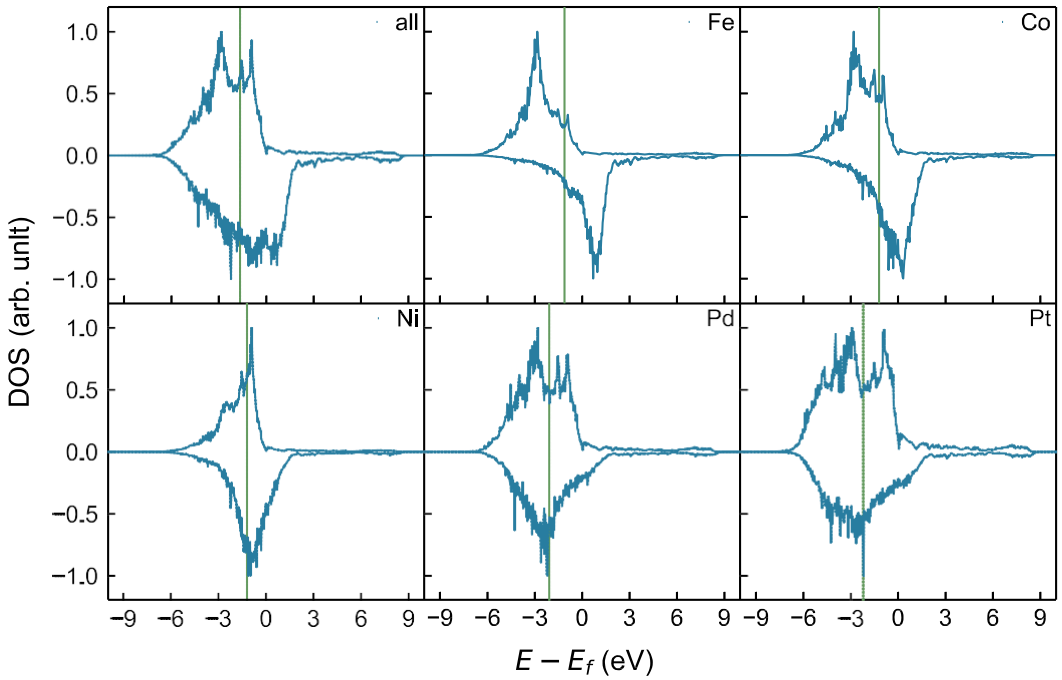

Figure S9 Projected density of states of surface atoms of $Ni_{18}Co_{10}Fe_{23}Pd_{16}Pt_{29}$. Green line denotes the energy level of *d*-band center.

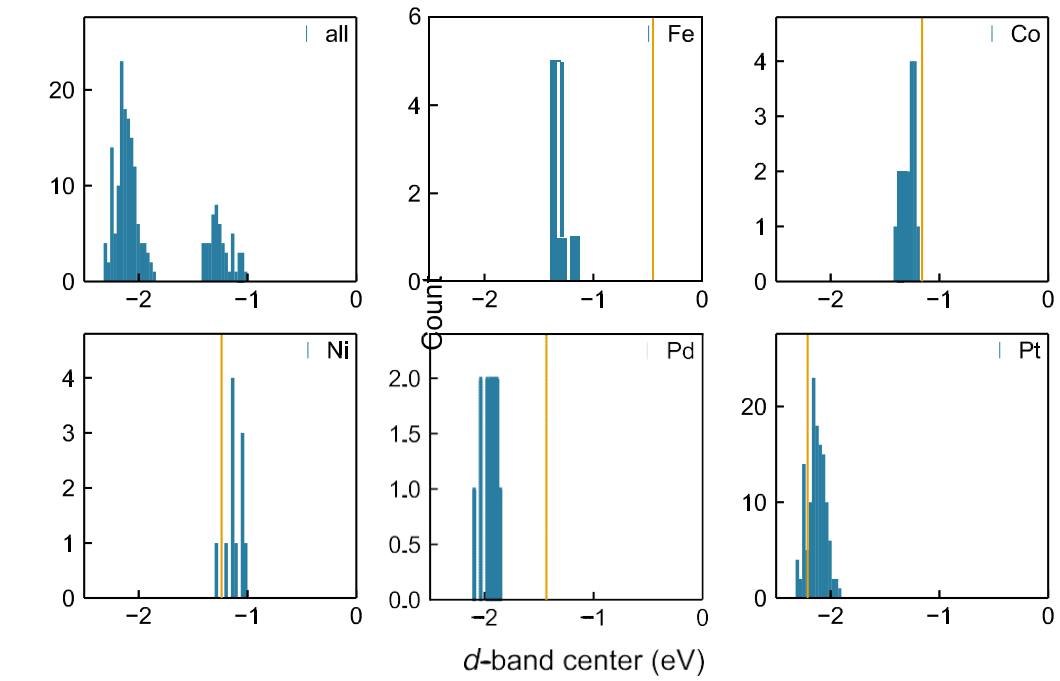

Figure S10 *d*-band center distribution of surface atoms of $Ni_6Co_8Fe_{12}Pd_6Pt_{64}$. Yellow line denotes the energy level of *d*-band center before mixing.

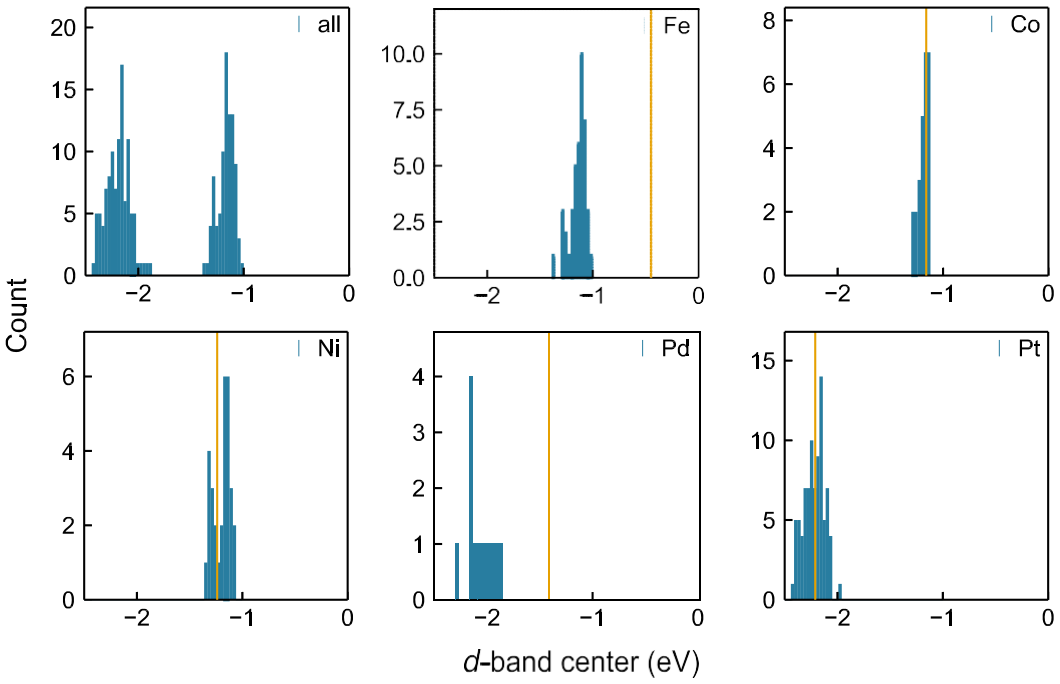

Figure S11 *d*-band center distribution of surface atoms of $Ni_{12}Co_{12}Fe_{21}Pd_8Pt_{43}$. Yellow line denotes the energy level of *d*-band center before mixing.

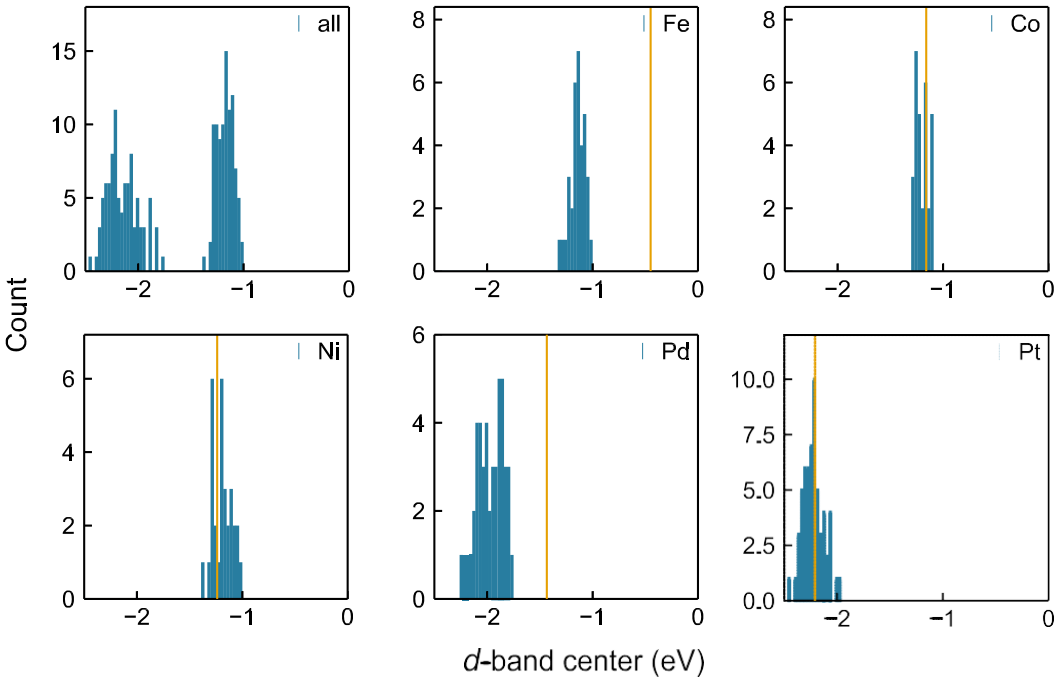

Figure S12 *d*-band center distribution of surface atoms of $Ni_{15}Co_{15}Fe_{19}Pd_{17}Pt_{30}$. Yellow line denotes the energy level of *d*-band center before mixing.

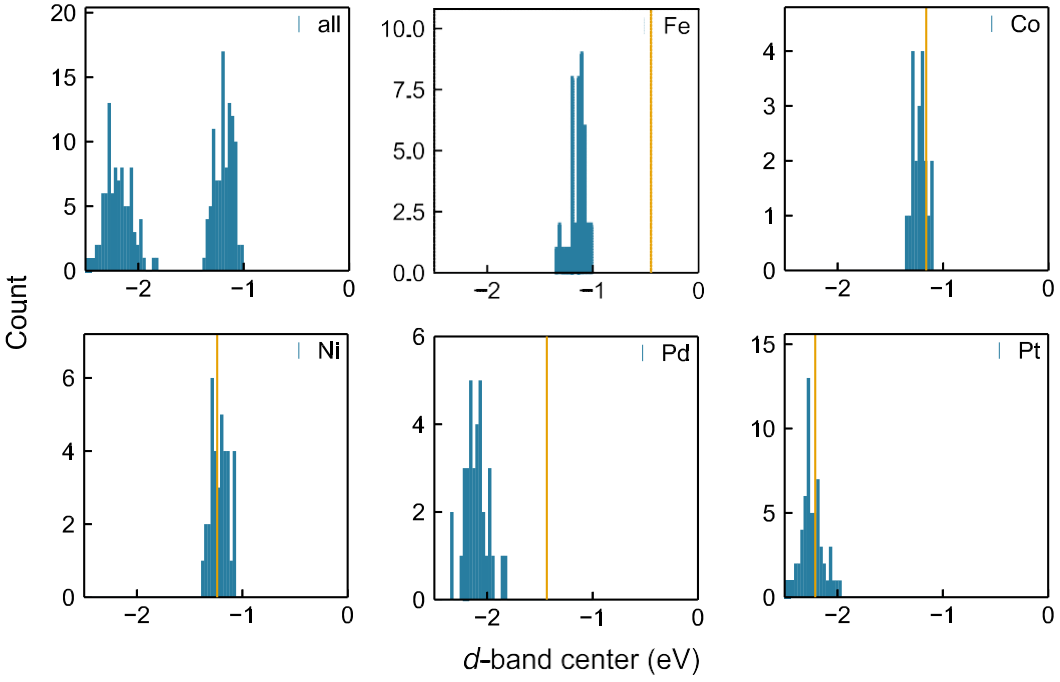

Figure S13 *d*-band center distribution of surface atoms of $Ni_{18}Co_{10}Fe_{23}Pd_{16}Pt_{29}$. Yellow line denotes the energy level of *d*-band center before mixing.