Supporting Information

DFT and Machine Learning for Predicting Hydrogen Adsorption Energies on Rocksalt Complex Oxides

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Table 1: List of Descriptors

Descriptor 1	Bader net charge
Descriptor 2	Average Oxygen 2p (up)
Descriptor 3	Average Oxygen 2p (down)
Descriptor 4	Bond Length-Valence Electron ratio (neighbor metal 1)
Descriptor 5	Bond Length-Valence Electron ratio (neighbor metal 2)
Descriptor 6	Bond Length-Valence Electron ratio (neighbor metal 3)
Descriptor 7	Bond Length-Valence Electron ratio (neighbor metal 4)
Descriptor 8	Bond Length-Valence Electron ratio (neighbor metal 5)
Descriptor 9	Frequency of Ni around Oxygen adsorption site
Descriptor 10	Frequency of Cu around Oxygen adsorption site
Descriptor 11	Frequency of Mg around Oxygen adsorption site
Descriptor 12	Frequency of Zn around Oxygen adsorption site
Descriptor 13	Average difference in electronegativity between Oxygen and neighboring metals
Descriptor 14	Average difference in ionization energy between Oxygen and neighboring metals

Example of how those descriptors are calculated:

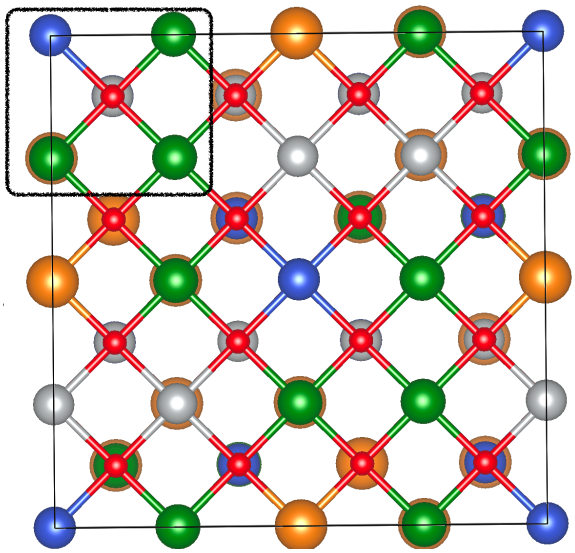

Figure 1: Slab top view example. Color legend: Ni, Mg, Cu, Zn, O are represented by green, orange, blue, gray and red respectively.

Figure 1 is an example of one of the slabs of the $\mathbf{Ni_{16}Mg_{16}Cu_{16}Zn_{16}O_{64}}$ model used to compute the dataset. For simplicity, one entry of the dataset (the set of descriptors for an Oxygen adsorption site) is explained. The same is extended for all the others entries. Remember that our dataset is composed of 336 entries from 21 slabs, each slab has 16 Oxygen adsorption sites. In Figure 1, one Oxygen adsorption site is denoted by the black box at the left top and will be our case explained. Each Oxygen adsorption site is surrounded by 5 metals, in the specific case the 5 neighbors are Ni, Ni, Ni, Cu (in the first layer) and Zn in the second layer.

From a single DFT calculation (optimization of the slab) the geometry is relaxed. Then, using this relaxed structure and after a postprocessing the electronic descriptors are obtained:

Descriptor 1: Bader net charge is obtained as described in the following link:
https://theory.cm.utexas.edu/henkelman/code/bader/

Descriptor 2: Average Oxygen 2p (up)
Descriptor 3: Average Oxygen 2p (down)
Both are calculated as in reference: Dickens, C. F. , Montoya, J. H. , Kulkarni, A. R., Bajdich, M., Nørskov, J. K. An electronic structure descriptor for oxygen reactivity at metal and metal-oxide surfaces. Surf. Sci. 2019, 681, 122–129

$$\bar{\epsilon}_{2p} = \frac{\int_{\epsilon_{min}}^{\epsilon_{max}} \rho_{2p} \epsilon d\epsilon}{\int_{\epsilon_{min}}^{\epsilon_{max}} \rho_{2p} d\epsilon}$$

The VASPKIT package was used to post-processing.(reference: Wang, V., Xu, N., Liu, J.C., Tang, G., Geng, W.T. VASPKIT: A user-friendly interface facilitat- ing high-throughput computing and analysis using VASP code. Computer Physics Communica- tions 2021, 267, 108033)

The valence electrons determined by the periodic table group : Mg equal 2, Ni equal 10, Cu equal 11, Zn equal 12.
Descriptor 4 : Bond Length-Valence Electron ratio (Ni) = distance Oxygen-Ni /10
Descriptor 5 : Bond Length-Valence Electron ratio (Ni) = distance Oxygen-Ni / 10
Descriptor 6 : Bond Length-Valence Electron ratio (Ni) = distance Oxygen-Ni / 10
Descriptor 7 : Bond Length-Valence Electron ratio (Cu) = distance Oxygen-Ni / 11
Descriptor 8 : Bond Length-Valence Electron ratio (Zn) = distance Oxygen-Ni / 12

By counting how many times is repeated a metal around the Oxygen adsorption site is possible to determine the following descriptors:
Descriptor 9 : Frequency of Ni around Oxygen adsorption site = 3
Descriptor 10: Frequency of Cu around Oxygen adsorption site = 1
Descriptor 11: Frequency of Mg around Oxygen adsorption site = 0
Descriptor 12: Frequency of Zn around Oxygen adsorption site = 1

Descriptor 13: Average difference in electronegativity between Oxygen and neighboring metals.
The Electronegativities values (Pauling scale) are : Ni equal 1.91, Mg equal 1.31, Zn equal 1.65, Cu equal 1.90, O equal 3.44. To calculate Descriptor 13, which represents the average difference in electronegativity between oxygen (O) and its neighboring metals (Ni, Mg, Zn, Cu), we first find the electronegativity differences for each metal. The electronegativity of oxygen is 3.44, and for each metal, we subtract its electronegativity from oxygen's. In the case of consideration, for Ni: 3.44 - 1.91 = 1.53. (Take in consideration that surrounding the Oxygen adsorption site there are 3 Ni atoms).For the other two neighbour metals: Cu: 3.44 -1.90=1.54 and Zn: 3.44 - 1.65 = 1.79. The average is then (1.53 + 1.53 + 1.53 + 1.54 + 1.79) / 5 = 1.584. Therefore, the Descriptor 13 value is 1.584.

Descriptor 14: Average difference in ionization energy between Oxygen and neighboring metals.
The Ionization energy values (in eV) are : Ni equal 7.64, Mg equal 7.646, Zn equal 9.394, Cu equal 7.726, O equal 13.618. To calculate the average difference in ionization energy between oxygen (O) and its neighboring metals (Ni, Cu, Zn), we first find the ionization energy difference for each metal. The ionization energy of oxygen is 13.618 eV, and for each metal, we subtract its ionization energy from oxygen's. For example, for Ni: 13.618 - 7.64 = 5.978. For the other two neighbour metals: Cu: 13.618 - 7.726 = 5.892 and Zn: 13.618 - 9.394 = 4.224. The average is then (5.978 + 5.978 + 5.978 + 5.892 + 4.224) / 5 = 5.61. Therefore, the Descriptor 14 value is 5.61.

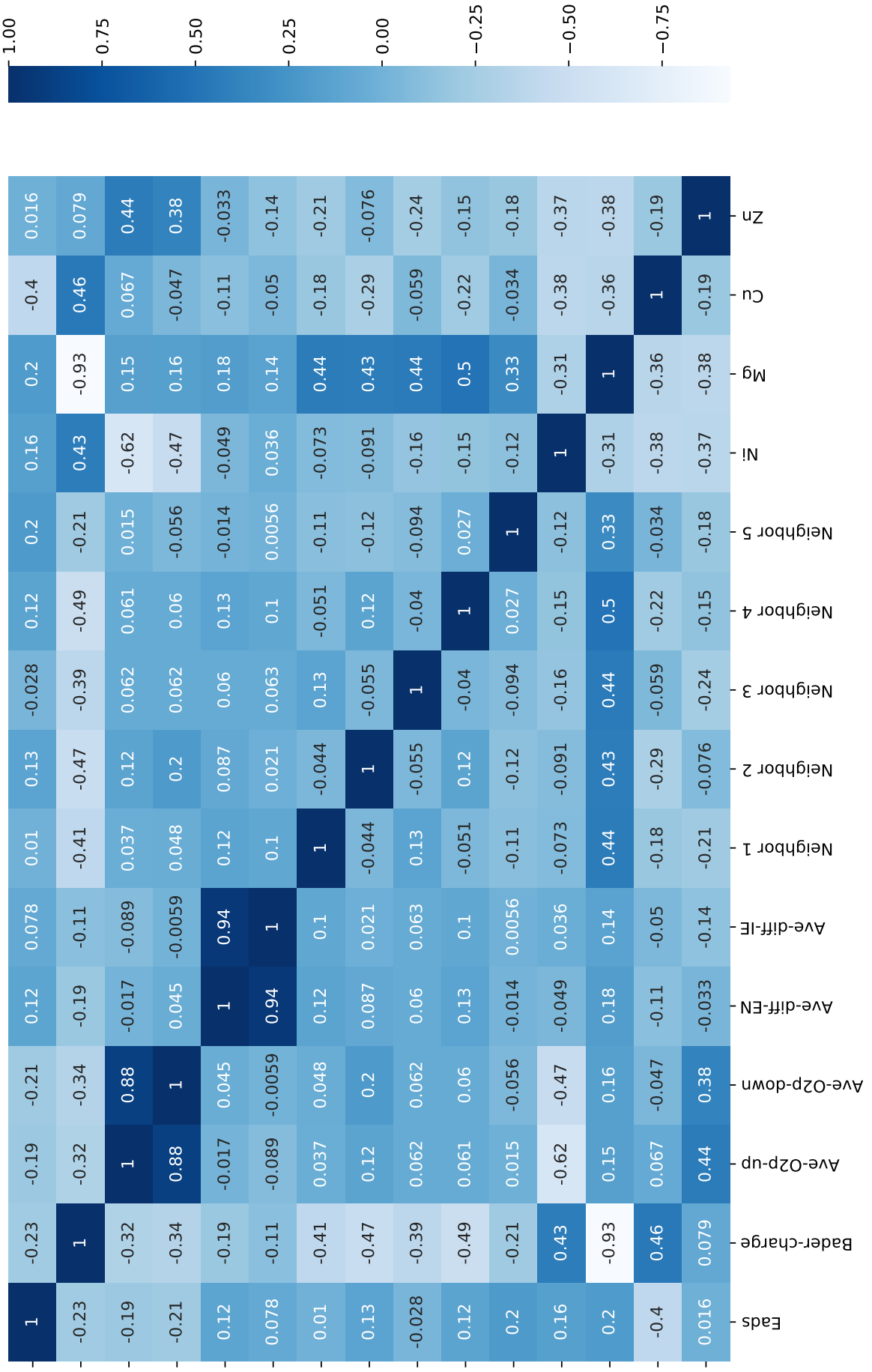

Figure 2: Correlation matrix