Stable perovskite discovery with machine learning

June 2, 2022

[1]:
```python
import pandas as pd
import numpy as np
import re
import warnings
warnings.filterwarnings("ignore")

# Importing plotting packages
import matplotlib.pyplot as plt
%matplotlib inline
import seaborn as sns
color = sns.color_palette("pastel")
sns.set_style("white")
sns.set(rc={'figure.figsize':(8,8)})
import plotly.express as px
import plotly.graph_objects as go
from plotly.subplots import make_subplots

# Importing Pandas and NumPy
import pandas as pd
import numpy as np
np.random.seed(0)

# Importing plotting packages
import matplotlib.pyplot as plt
%matplotlib inline
import seaborn as sns
color = sns.color_palette("pastel")
sns.set_style("white")
sns.set(rc={'figure.figsize':(8,8)})
import plotly.express as px
import plotly.graph_objects as go
from plotly.subplots import make_subplots

from tabulate import tabulate

import joblib
```

```python
# Import machine learning packages
from sklearn.model_selection import train_test_split, GridSearchCV,␣
 ↪cross_val_predict
from sklearn.preprocessing import StandardScaler
from sklearn.pipeline import Pipeline
from sklearn.base import BaseEstimator, TransformerMixin
from sklearn.model_selection import cross_validate
from sklearn.metrics import mean_squared_error
from sklearn.linear_model import LinearRegression
from sklearn.svm import SVR
from sklearn.ensemble import RandomForestRegressor
from sklearn.neural_network import MLPRegressor

import sklearn.neighbors._base
from xgboost import XGBRegressor

from scipy.stats import zscore

import sys
sys.modules['sklearn.neighbors.base'] = sklearn.neighbors._base

import warnings
warnings.filterwarnings("ignore")
```

[2]:
```python
# DFT Calculations
dft_calc = pd.read_csv("data/dft_calc.csv").replace(" ", 0)

# Elemental Properties
elemental_prop = pd.read_csv("data/elemental_properties.csv")
elemental_prop = elemental_prop.replace(" ", 0)

# Shannon Radii
shan_rad = pd.read_csv("data/shannon_radius.csv")

# Copy of dft_calc
df = dft_calc.copy()
```

[3]:
```python
a_shan_rad = shan_rad[["A_SITE","A_RADII"]]
b_shan_rad = shan_rad[["B_SITE","B_RADII"]]
```

[4]:
```python
a_shan_rad_sites = list(a_shan_rad["A_SITE"])
b_shan_rad_sites = list(b_shan_rad["B_SITE"])

a_shan_rad = list(a_shan_rad["A_RADII"])
b_shan_rad = list(b_shan_rad["B_RADII"])
```

0.1 Data Setup

The dataset is derived from the work of Jacobs et al. who used density functional theory (DFT) methods to simulate 1,926 perovskite oxides to calculate their thermodynamic stability [1]. As shown below, their dataset contains 11 columns which includes the material name, the ions in the A-site, B-site and X-site, their energy above hull E_{hull} in meV/atom, and their formation energy in meV/atom.

[5]:
```
dft_calc.head()
```

[5]:
```
        COMPOSITION A_SITE_1 A_SITE_2 A_SITE_3 B_SITE_1 B_SITE_2 B_SITE_3  \
0       Ba1Sr7V8O24       Ba       Sr      NaN        V      NaN      NaN
1   Ba2Bi2Pr4Co8O24       Ba       Bi       Pr       Co      NaN      NaN
2     Ba2Ca6Fe8O24       Ba       Ca      NaN       Fe      NaN      NaN
3   Ba2Cd2Pr4Ni8O24       Ba       Cd       Pr       Ni      NaN      NaN
4     Ba2Dy6Fe8O24       Ba       Dy      NaN       Fe      NaN      NaN

  X_SITE  NUM_ELEMS  ENERGY_ABOVE_HULL  FORMATION_ENERGY
0      O          4          29.747707         -2.113335
1      O          5         106.702335         -1.311863
2      O          4         171.608093         -1.435607
3      O          5         284.898190         -0.868639
4      O          4         270.007913         -1.746806
```

[6]:
```
dft_calc.info()
```

```
<class 'pandas.core.frame.DataFrame'>
RangeIndex: 1926 entries, 0 to 1925
Data columns (total 11 columns):
 #   Column             Non-Null Count  Dtype
---  ------             --------------  -----
 0   COMPOSITION        1926 non-null   object
 1   A_SITE_1           1926 non-null   object
 2   A_SITE_2           1159 non-null   object
 3   A_SITE_3           34 non-null     object
 4   B_SITE_1           1926 non-null   object
 5   B_SITE_2           1247 non-null   object
 6   B_SITE_3           33 non-null     object
 7   X_SITE             1926 non-null   object
 8   NUM_ELEMS          1926 non-null   int64
 9   ENERGY_ABOVE_HULL  1926 non-null   float64
 10  FORMATION_ENERGY   1926 non-null   float64
dtypes: float64(2), int64(1), object(8)
memory usage: 165.6+ KB
```

Another dataset from the work of Logan et al. is used to supplement the `dft_calc` dataset [2]. As seen below, the dataset contains information regarding the chemical and properties for each element in the periodic table.

[7]:
```
elemental_prop.head()
```

[7]:
```
  SYMBOL IONIC_RADIUS  MOD_OF_ELASTICITY     BP     MP DENSITY    AT_WT  \
0      H         1.54                NaN  20.28  13.81  0.0899  1.00797
1     He            0                NaN  4.216   0.95  0.1785   4.0026
2     Li         0.76               10.0   1615  453.7    0.53    6.941
3     Be         0.45              301.0   3243   1560    1.85  9.01218
4      B         0.23              441.0   4275   2365    2.34   10.811

   BCC_EFF_LAT_CNT  BCC_ENERGY  BCC_ENERGY_DIFF  …  IS_NONMETAL  \
0         3.589268   -2.135811         1.195480  …            1
1         5.373995   -2.000673        -2.001808  …            1
2         6.416364   -1.865535         0.004352  …            0
3         4.997332   -3.655272         0.099767  …            0
4         4.606670   -4.966431         1.711267  …            0

   ND_UNFILLED  ND_VALENCE  NF_UNFILLED  NF_VALENCE  NP_UNFILLED  NP_VALENCE  \
0            0           0            0           0            0           0
1            0           0            0           0            0           0
2            0           0            0           0            0           0
3            0           0            0           0            0           0
4            0           0            0           0            5           1

   NS_UNFILLED  NS_VALENCE  N_UNFILLED
0            1           1           1
1            0           2           0
2            1           1           1
3            0           2           0
4            0           2           5

[5 rows x 82 columns]
```

[8]:
```
elemental_prop.info()
```

```
<class 'pandas.core.frame.DataFrame'>
RangeIndex: 110 entries, 0 to 109
Data columns (total 82 columns):
 #   Column                  Non-Null Count  Dtype
---  ------                  --------------  -----
 0   SYMBOL                  110 non-null    object
 1   IONIC_RADIUS            110 non-null    object
 2   MOD_OF_ELASTICITY       81 non-null     float64
 3   BP                      110 non-null    object
 4   MP                      110 non-null    object
 5   DENSITY                 110 non-null    object
 6   AT_WT                   110 non-null    object
 7   BCC_EFF_LAT_CNT         110 non-null    float64
```

8	BCC_ENERGY	110 non-null	float64
9	BCC_ENERGY_DIFF	110 non-null	float64
10	BCC_FERMI	110 non-null	float64
11	BCC_MAG_MOM	110 non-null	float64
12	BCC_VOLUME_PA	110 non-null	float64
13	BCC_VOLUME_DIFF	110 non-null	float64
14	GS_BANDGAP	110 non-null	float64
15	GS_EFF_LAT_CNT	110 non-null	float64
16	GS_ENERGY	110 non-null	float64
17	GS_MAG_MOM	110 non-null	float64
18	GS_VOLUME_PA	110 non-null	float64
19	HH_IP	77 non-null	float64
20	HH_IR	77 non-null	float64
21	ICSD_VOLUME	110 non-null	float64
22	COV_RAD	110 non-null	object
23	ION_ERGY	110 non-null	object
24	ATOM_RAD	110 non-null	object
25	ELECT_AFF	110 non-null	object
26	AT_RAD	110 non-null	object
27	AT_VOL	110 non-null	object
28	MEN_NUM	103 non-null	float64
29	N_WS_THIRD	73 non-null	float64
30	1_ION_POT	110 non-null	object
31	2_ION_POT	110 non-null	object
32	3_ION_POT	110 non-null	object
33	CTE	110 non-null	object
34	SP_HEAT_CAP	110 non-null	object
35	THERMAL_COND	110 non-null	object
36	CONDUCTIVITY	110 non-null	object
37	HEAT_OF_FUSION	110 non-null	object
38	HEAT_OF_VAP	110 non-null	object
39	ELECTRONEGATIVITY	110 non-null	object
40	AT_NUM	110 non-null	int64
41	PERIOD	110 non-null	int64
42	GRP	110 non-null	int64
43	VALENCE	110 non-null	object
44	IS_HEXAGONAL	110 non-null	int64
45	IS_BCC	110 non-null	int64
46	IS_CUBIC	110 non-null	int64
47	IS_FCC	110 non-null	int64
48	IS_ORTHO	110 non-null	int64
49	IS_RHOMBO	110 non-null	int64
50	IS_MONO	110 non-null	int64
51	IS_TETRA	110 non-null	int64
52	IS_ALKALI	110 non-null	int64
53	IS_ALKALI_EARTH	110 non-null	int64
54	IS_BORON	110 non-null	int64
55	IS_CARBON	110 non-null	int64

```
 56  IS_CHALCOGEN            110 non-null    int64
 57  IS_HALOGEN              110 non-null    int64
 58  IS_HYDROGEN             110 non-null    int64
 59  IS_NOBLE_GAS            110 non-null    int64
 60  IS_PINICTIDE            110 non-null    int64
 61  IS_RARE_EARCH           110 non-null    int64
 62  IS_TRANS_METAL          110 non-null    int64
 63  S_ORBITAL               110 non-null    int64
 64  P_ORBITAL               110 non-null    int64
 65  D_ORBITAL               110 non-null    int64
 66  F-ORBITAL               110 non-null    int64
 67  STABLE_OXIDATION_STATE  110 non-null    object
 68  IS_D_BLOCK              110 non-null    int64
 69  IS_F_BLOCK              110 non-null    int64
 70  IS_METAL                110 non-null    int64
 71  IS_METALLOID            110 non-null    int64
 72  IS_NONMETAL             110 non-null    int64
 73  ND_UNFILLED             110 non-null    int64
 74  ND_VALENCE              110 non-null    int64
 75  NF_UNFILLED             110 non-null    int64
 76  NF_VALENCE              110 non-null    int64
 77  NP_UNFILLED             110 non-null    int64
 78  NP_VALENCE              110 non-null    int64
 79  NS_UNFILLED             110 non-null    int64
 80  NS_VALENCE              110 non-null    int64
 81  N_UNFILLED              110 non-null    int64
dtypes: float64(18), int64(40), object(24)
memory usage: 70.6+ KB
```

0.2 Data pre-processing

Most numerical features in the `elemental_prop` dataset are encoded as strings and should be converted to a data type that machine learning models can work on. Additionally, the column for ionization energy is encoded as strings with commas. The commas should be removed before converting str to float. The first column in `dft_calc`, for example, is shown below:

[9]:
```
# Commas in the ionization energy column
elemental_prop["ION_ERGY"]
```

[9]:
```
0          1312
1      2,372.30
2           520
3        899.40
4        800.60
         …
105           0
106           0
107           0
```

```
108          0
109          0
Name: ION_ERGY, Length: 110, dtype: object
```

[10]:
```
# Removing commas on column `ION_ERGY`
elemental_prop["ION_ERGY"] = elemental_prop["ION_ERGY"].str.replace(",","")

# Creating list of numerical columns in elemental_prop that should be converted␣
↪to float
col_names_no_symbol = [i for i in elemental_prop.columns if i != "SYMBOL"]

# Conversion to float
elemental_prop[col_names_no_symbol] = elemental_prop[col_names_no_symbol].
↪astype("float")
```

0.3 Dataset building

To build the actual dataset to be used for modeling, `dft_calc` and `elemental_prop` are joined using the elements in the different sites as key.

[11]:
```
dft_calc.iloc[0,:]
```

[11]:
```
COMPOSITION          Ba1Sr7V8O24
A_SITE_1                      Ba
A_SITE_2                      Sr
A_SITE_3                     NaN
B_SITE_1                       V
B_SITE_2                     NaN
B_SITE_3                     NaN
X_SITE                         O
NUM_ELEMS                      4
ENERGY_ABOVE_HULL      29.747707
FORMATION_ENERGY       -2.113335
Name: 0, dtype: object
```

It has Ba and Sr in its A-sites, and V in its B-sites. The O in its X-site can be disregarded since all materials in the dataset are perovskite oxides and have O in their X-sites. The elemental properties of Ba, Sr, and V are joined into `dft_calc` with column names appended by the site the element is in.

[12]:
```
# List of suffixes to append to elemental properties
# ['A1', 'A2', 'A3', 'B1', 'B2', 'B3']
suffixes = "A1 A2 A3 B1 B2 B3".split()

# List of columns under dft_calc showing sites of elements
# To be used as key in merging
# ['A_SITE_1', 'A_SITE_2', 'A_SITE_3', 'B_SITE_1', 'B_SITE_2', 'B_SITE_3']
site_names = list(dft_calc.columns[1:7])
```

```
# List of column names for symbols under placeholder dataframe
# ['A1_SYMBOL', 'A2_SYMBOL', 'A3_SYMBOL', 'B1_SYMBOL', 'B2_SYMBOL', 'B3_SYMBOL']
symbol_names = [i+"_SYMBOL" for i in suffixes]

# Initializing empty list containing column names for
# A-sites and B-sites
elemental_prop_col_names = []

# Populating elemental_prop_col_names
# Loops through all sites and creates site-specific column names for each
# elemental property
for i in range(6):
    placeholder_df = elemental_prop.copy()
    placeholder_df.columns = suffixes[i] + "_" + placeholder_df.columns.values
    dft_calc = pd.merge(dft_calc, placeholder_df, how="left",␣
 ↪left_on=site_names[i], right_on=symbol_names[i])

    placeholder_df = placeholder_df.drop(columns=[suffixes[i] + "_SYMBOL"])
    elemental_prop_col_names.append(placeholder_df.columns)
```

Shown below is a list containing new column names for Site A1. In the case of Ba1Sr7V8O24, the columns below correspond to the elemental properties of Ba since it is the first element in its A-site.

[13]:
```
# elemental_prop_col_name is a list of lists
elemental_prop_col_names[0]
```

[13]:
```
Index(['A1_IONIC_RADIUS', 'A1_MOD_OF_ELASTICITY', 'A1_BP', 'A1_MP',
       'A1_DENSITY', 'A1_AT_WT', 'A1_BCC_EFF_LAT_CNT', 'A1_BCC_ENERGY',
       'A1_BCC_ENERGY_DIFF', 'A1_BCC_FERMI', 'A1_BCC_MAG_MOM',
       'A1_BCC_VOLUME_PA', 'A1_BCC_VOLUME_DIFF', 'A1_GS_BANDGAP',
       'A1_GS_EFF_LAT_CNT', 'A1_GS_ENERGY', 'A1_GS_MAG_MOM', 'A1_GS_VOLUME_PA',
       'A1_HH_IP', 'A1_HH_IR', 'A1_ICSD_VOLUME', 'A1_COV_RAD', 'A1_ION_ERGY',
       'A1_ATOM_RAD', 'A1_ELECT_AFF', 'A1_AT_RAD', 'A1_AT_VOL', 'A1_MEN_NUM',
       'A1_N_WS_THIRD', 'A1_1_ION_POT', 'A1_2_ION_POT', 'A1_3_ION_POT',
       'A1_CTE', 'A1_SP_HEAT_CAP', 'A1_THERMAL_COND', 'A1_CONDUCTIVITY',
       'A1_HEAT_OF_FUSION', 'A1_HEAT_OF_VAP', 'A1_ELECTRONEGATIVITY',
       'A1_AT_NUM', 'A1_PERIOD', 'A1_GRP', 'A1_VALENCE', 'A1_IS_HEXAGONAL',
       'A1_IS_BCC', 'A1_IS_CUBIC', 'A1_IS_FCC', 'A1_IS_ORTHO', 'A1_IS_RHOMBO',
       'A1_IS_MONO', 'A1_IS_TETRA', 'A1_IS_ALKALI', 'A1_IS_ALKALI_EARTH',
       'A1_IS_BORON', 'A1_IS_CARBON', 'A1_IS_CHALCOGEN', 'A1_IS_HALOGEN',
       'A1_IS_HYDROGEN', 'A1_IS_NOBLE_GAS', 'A1_IS_PINICTIDE',
       'A1_IS_RARE_EARCH', 'A1_IS_TRANS_METAL', 'A1_S_ORBITAL', 'A1_P_ORBITAL',
       'A1_D_ORBITAL', 'A1_F-ORBITAL', 'A1_STABLE_OXIDATION_STATE',
       'A1_IS_D_BLOCK', 'A1_IS_F_BLOCK', 'A1_IS_METAL', 'A1_IS_METALLOID',
       'A1_IS_NONMETAL', 'A1_ND_UNFILLED', 'A1_ND_VALENCE', 'A1_NF_UNFILLED',
       'A1_NF_VALENCE', 'A1_NP_UNFILLED', 'A1_NP_VALENCE', 'A1_NS_UNFILLED',
```

```
 'A1_NS_VALENCE', 'A1_N_UNFILLED'],
dtype='object')
```

Below is a function that uses RegEx to determine the number of each atom in a mole of the material. For instance, for Ba1Sr7V8O24, should yield the following: - NUM_A1: 1 (Ba) - NUM_A2: 7 (Sr) - NUM_A3: NaN - NUM_B1: 8 (V) - NUM_B2: NaN - NUM_B3: NaN

[14]:
```
def num_of_sites(site):
    nums =[]

    # Loops through all rows of dft_calc
    for i in range(dft_calc.shape[0]):

        # Applies RegEx filter to COMPOSITION yielding a list of tuples
        # [('Ba', '1'), ('Sr', '7'), ('V', '8'), ('O', '24'), ('', '')] when i␣
 ↪= 0
        matches = re.findall(r"(\D*)(\d*)", dft_calc["COMPOSITION"].iloc[i])

        # Proceed only when site is not NaN, note that most materials do not
        # have 3 elements in their A-site/B-site
        if type(dft_calc[site].iloc[i]) == str:

            # Looping through the list of tuples yielded above
            # to populate all sites
            for j in range(len(matches)):
                # When j = 0, i = 0, matches[j] = ('Ba', '1')
                # When j = 1, i = 0, matches[j] = ('Sr', '7')
                if matches[j][0] == dft_calc[site].iloc[i]:
                    nums.append(int(matches[j][1]))
                    continue
                else:
                    continue

        # Appends NaN when site is unfilled
        else:
            nums.append(np.nan)
    return nums
```

[15]:
```
num_sites = []

# Loops through all sites
# Each iteration populates a site
# Results into a list of lists
for i in site_names:
    num_sites.append(num_of_sites(i))

# The first element in num_sites consists of
```

```
# 196 rows corresponding to the number of the A1 atoms
# in the composition
num_sites[0][0:20]
```

[15]: [1, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2, 2]

[16]:
```
# Generating names for new columns
# ['NUM_A1', 'NUM_A2', 'NUM_A3', 'NUM_B1', 'NUM_B2', 'NUM_B3']
num_col_names = ["NUM_" + i for i in suffixes]
num_col_names

# Populating dft_calc with list of lists created earlier
for i in range(6):
    dft_calc[num_col_names[i]] = num_sites[i]
    df[num_col_names[i]] = num_sites[i]
```

Below shows the new columns for Ba1Sr7V8O24.

[17]:
```
dft_calc[num_col_names].iloc[0]
```

[17]:
```
NUM_A1    1.0
NUM_A2    7.0
NUM_A3    NaN
NUM_B1    8.0
NUM_B2    NaN
NUM_B3    NaN
Name: 0, dtype: float64
```

0.3.1 Adding structural parameters

Structural parameters unique to perovskite crystals such as Goldschmidt tolerance factor (t) and octahedral factor (μ) are added to the dataset. The t of a material is a dimensionless number that is calculated from the ratio of the ionic radii of the elements comprising a perovskite crystals. It is expressed mathematically as

$$t = \frac{r_A + r_O}{\sqrt{2}(r_B + r_O)}$$

where r_A is the radius of the A cation, r_B is the radius of the B cation, and r_O is the radius of the anion (oxygen, in this case, which is equal to 1.4).

The ideal and stable structure for a perovskite crystal is cubic which as $t = 1$. - > 1: Hexagonal or tetragonal (A too big or B too small) - 0.9-1: Cubic (A and B ideal) - 0.71-0.9: Orthorhombic/rhombohesral (A too small)

For perovskites with multiple ions in each site such as the ones in the dataset, the radius for each ion type should be averaged with respect to composition. The composition averaged radius of site S can be calculated using the formula below:

$$r_S = \sum_{i=0}^{2} \frac{n_i}{n_{tot}} r_i$$

where n_i is the number of the i-th ion, and n_{tot} is the total number of atoms in site S (always 8 in this case).

Much like t, μ is also widely used to predict crystal stability. It is expressed as:

$$\mu = \frac{r_B}{r_X}.$$

When $\mu > 0.41$, the perovskite structure is said to be stable.

The bond length between the A, B, and X ions are also added as a new feature where:

$$AB = r_A + r_B$$

$$AO = r_A + r_O$$

$$BO = r_B + r_O$$

[18]:

```
shan_rad_col_names = [i + "_SHANRAD" for i in suffixes]
shan_rad_col_names

for i in range(len(shan_rad_col_names)):
    df[shan_rad_col_names[i]] = df[site_names[i]]

    if i < 3:
        df[shan_rad_col_names[i]] = df[shan_rad_col_names[i]].
 ↪replace(a_shan_rad_sites, a_shan_rad)
    else:
        df[shan_rad_col_names[i]] = df[shan_rad_col_names[i]].
 ↪replace(b_shan_rad_sites, b_shan_rad)
```

[19]:

```
# Generating names for new columns indicating ionic radii of
# ions in different sites
# ['A1_IONIC_RADIUS',
#  'A2_IONIC_RADIUS',
#  'A3_IONIC_RADIUS',
#  'B1_IONIC_RADIUS',
#  'B2_IONIC_RADIUS',
#  'B3_IONIC_RADIUS']

ionic_radius_names = [i for i in dft_calc if "_IONIC_RADIUS" in i]
ionic_radius_names

# Empty list for Goldschmidt tolerance factor
gs = []
```

```python
# Empty list for Goldschmidt tolerance factor
gs_shan = []

# Empty list for octahedral factor
of = []

# Empty list for octahedral factor
of_shan = []

# Empty list for A-B bond length
ab = []

# Empty list for A-O bond length
ao = []

# Empty list for B-O bond length
bo = []

# Empty list for A ions with highest composition
a_max = []

# Empty list for B ions with highest composition
b_max = []

# Loops through all rows of dft_calc
for i in range(dft_calc.shape[0]):

    # Takes list of ionic radii of A ions (r_i when S=A)
    ionic_radii_list_a = list(dft_calc[ionic_radius_names[0:3]].iloc[i].
 ↪dropna())

    # Takes list of Shannon radii of A ions (r_shan_i when S=A)
    shannon_radii_list_a = list(df[shan_rad_col_names[0:3]].iloc[i].dropna())

    # Takes list of number of A ions (n_i when S=A)
    num_list_a = list(dft_calc[num_col_names[0:3]].iloc[i].dropna())

    # Adding which A ion has highest composition to a_max
    a_max.append(dft_calc[site_names[np.argmax(num_list_a)]].iloc[i])

    # Takes list of ionic radii of B ions (r_i when S=B)
    ionic_radii_list_b = list(dft_calc[ionic_radius_names[3:6]].iloc[i].
 ↪dropna())

    # Takes list of Shannon radii of B ions (r_i when S=B)
    shannon_radii_list_b = list(df[shan_rad_col_names[3:6]].iloc[i].dropna())
```

```
# Takes list of number of A ions (n_i when S=A)
num_list_b = list(dft_calc[num_col_names[3:6]].iloc[i].dropna())

# Adding which A ion has highest composition to a_max
b_max.append(dft_calc[site_names[np.argmax(num_list_b)+3]].iloc[i])

sam_list_a = len(ionic_radii_list_a)
sam_list_b = len(ionic_radii_list_b)

sam_list_a_shan = len(shannon_radii_list_a)
sam_list_b_shan = len(shannon_radii_list_b)

a_sum = 0

# Calculating composition-averaged radius when S=A
for j in range(sam_list_a):
    a = ionic_radii_list_a[j] * num_list_a[j]
    a_sum = a_sum + a

b_sum = 0

# Calculating composition-averaged radius when S=B
for k in range(sam_list_b):
    b = ionic_radii_list_b[k] * num_list_b[k]
    b_sum = b_sum + b
    # Calculating octahedral factor
    of_ = (b_sum/8)/1.4

a_sum_shan = 0

# Calculating composition-averaged radius when S=A
for l in range(sam_list_a):
    a = shannon_radii_list_a[l] * num_list_a[l]
    a_sum_shan = a_sum_shan + a

b_sum_shan = 0

# Calculating composition-averaged radius when S=B
for m in range(sam_list_b):
    b = shannon_radii_list_b[m] * num_list_b[m]
    b_sum_shan = b_sum_shan + b
    # Calculating octahedral factor
    of_shan_ = (b_sum_shan/8)/1.4

# Calculating t
gs_tf = ((a_sum/8) + 1.4)/(np.sqrt(2)*((b_sum/8)+1.4))
```

```
    # Calculating t based on Shannon radius
    gs_shan_ = ((a_sum_shan/8) + 1.4)/(np.sqrt(2)*((b_sum_shan/8)+1.4))

    # Appending t
    gs.append(gs_tf)
    # Appending t based on Shannon radius
    gs_shan.append(gs_shan_)
    # Appending octahedral factor
    of.append(of_)
    # Appending octahedral factor based on Shannon radius
    of_shan.append(of_shan_)
    # Appending AB
    ab.append((a_sum_shan/8)+(b_sum_shan/8))
    # Appending AO
    ao.append((a_sum_shan/8)+1.4)
    # Appending BO
    bo.append((b_sum_shan/8)+1.4)

df["GOLDSCHMIDT_TF"] = gs
df["OCTAHEDRAL_FACTOR"] = of
df["A_B"] = ab
df["A_O"] = ao
df["B_O"] = bo
df["A_MAX"] = a_max
df["B_MAX"] = b_max
df["GOLDSCHMIDT_SHAN"] = gs_shan
df["OCTAHEDRAL_SHAN"] = of_shan
```

0.3.2 Adding elemental properties of majority ions

In the previous section, structural parameters such as t, μ, AB, AO, and BO were added to the dataframe. A previously undiscussed feature was also added, namely `A_MAX` and `B_MAX` which represent which ions have the highest total number in their respective sites. For instance, `A_MAX` and `B_MAX` for Ba1Sr7V8O24 are Sr (7) and V (8), respectively. These new columns will be used as keys as the elemental properties for each ion are merged into the dataset.

[20]:
```
# Preparing new column names
placeholder_df_amax = elemental_prop.copy()
placeholder_df_amax.columns = "A_MAX_" + placeholder_df_amax.columns.values
a_max_names = list(placeholder_df_amax.columns)

placeholder_df_bmax = elemental_prop.copy()
placeholder_df_bmax.columns = "B_MAX_" + placeholder_df_bmax.columns.values
b_max_names = list(placeholder_df_bmax.columns)
```

[21]:

```
df = pd.merge(df, placeholder_df_amax, how="inner", left_on="A_MAX",␣
↪right_on="A_MAX_SYMBOL")
df = df.drop(columns=["A_MAX_SYMBOL"])
a_max_names.remove("A_MAX_SYMBOL")
```

[22]:
```
df = pd.merge(df, placeholder_df_bmax, how="inner", left_on="B_MAX",␣
↪right_on="B_MAX_SYMBOL")
df = df.drop(columns=["B_MAX_SYMBOL"])
b_max_names.remove("B_MAX_SYMBOL")
```

0.3.3 Adding composition averaged properties, maximum, minimum, and range

Considering that the alloying elements and number of elements in the A- and B-site vary, new features can be added to capture the stoichiometric diversity of each perovskite crystal. Below, the following features are added: - Composition averaged properties of all ions in each site. - Maximum value of properties in each site - Minimum value of properties in each site - Range of values of properties in each site

[23]:
```
a_prop_names = []
b_prop_names = []

a_wt_avg_names = []
b_wt_avg_names = []

# For loop creating (1) column names for each property "triplet"
# a_prop_names[0] for example yields
# ['A1_IONIC_RADIUS', 'A2_IONIC_RADIUS', 'A3_IONIC_RADIUS']
# and (2) names for new features corresponding to
# composition averages of each property "triplet"
for i in elemental_prop.columns[1:82]:
    triplet_props_a = [j + "_" + i for j in suffixes[0:3]]
    triplet_props_b = [j + "_" + i for j in suffixes[3:6]]

    a_prop_names.append(triplet_props_a)
    b_prop_names.append(triplet_props_b)

    a_wt_avg_names.append("A_WT_AVG_" + i)
    b_wt_avg_names.append("B_WT_AVG_" + i)
```

[24]:
```
df[shan_rad_col_names[0:3]]
```

[24]:

	A1_SHANRAD	A2_SHANRAD	A3_SHANRAD
0	1.61	1.440	NaN
1	1.61	1.440	NaN
2	1.44	1.340	NaN
3	1.44	1.083	NaN

```
4           1.44      1.107        NaN
…           …         …         …
1921        1.61        NaN        NaN
1922        1.61        NaN        NaN
1923        1.61        NaN        NaN
1924        1.61        NaN        NaN
1925        1.61        NaN        NaN

[1926 rows x 3 columns]
```

[25]:

```
# Creating names for new columns corresponding to the maximum property
a_max_all_names = ["ALL_MAX_A_" + i for i in elemental_prop.columns[1:82]]
b_max_all_names = ["ALL_MAX_B_" + i for i in elemental_prop.columns[1:82]]

# Creating names for new columns corresponding to the minimum property
a_min_all_names = ["ALL_MIN_A_" + i for i in elemental_prop.columns[1:82]]
b_min_all_names = ["ALL_MIN_B_" + i for i in elemental_prop.columns[1:82]]

# Creating names for new columns corresponding to the range
a_range_names = ["RANGE_A_" + i for i in elemental_prop.columns[1:82]]
b_range_names= ["RANGE_B_" + i for i in elemental_prop.columns[1:82]]
```

[26]:

```
for j in range(81):
    a_vals = []
    b_vals = []
    a_max_alls = []
    b_max_alls = []
    a_min_alls = []
    b_min_alls = []
    a_ranges = []
    b_ranges = []

    for i in range(dft_calc.shape[0]):

        num_list_a = list(dft_calc[num_col_names[0:3]].iloc[i].dropna())
        num_list_b = list(dft_calc[num_col_names[3:6]].iloc[i].dropna())

        len_a = len(num_list_a)
        len_b = len(num_list_b)

        a_properties = list(dft_calc[a_prop_names[j]].iloc[i].dropna())
        b_properties = list(dft_calc[b_prop_names[j]].iloc[i].dropna())

        a_max_all = max(a_properties)
        b_max_all = max(b_properties)

        a_min_all = min(a_properties)
```

```
        b_min_all = min(b_properties)

        a_range = a_max_all - a_min_all
        b_range = b_max_all - b_min_all

        a = np.sum(np.multiply(a_properties, num_list_a))/(8)
        b = np.sum(np.multiply(b_properties, num_list_b))/(8)

        a_vals.append(a)
        b_vals.append(b)
        a_max_alls.append(a_max_all)
        b_max_alls.append(b_max_all)
        a_min_alls.append(a_min_all)
        b_min_alls.append(b_min_all)
        a_ranges.append(a_range)
        b_ranges.append(b_range)

    df[a_wt_avg_names[j]] = a_vals
    df[b_wt_avg_names[j]] = b_vals

    df[a_max_all_names[j]] = a_max_alls
    df[b_max_all_names[j]] = b_max_alls

    df[a_min_all_names[j]] = a_min_alls
    df[b_min_all_names[j]] = b_min_alls

    df[a_range_names[j]] = a_ranges
    df[b_range_names[j]] = b_ranges
```

[27]:
```
a_shan_rad_comp_avgd = []
b_shan_rad_comp_avgd = []

# Adding maximum, minimum, range column for Shannon radius

df["A_MAX_SHAN_RAD"] = df[shan_rad_col_names[0:3]].max(axis=1)
df["A_MIN_SHAN_RAD"] = df[shan_rad_col_names[0:3]].min(axis=1)
df["A_RANGE_SHAN_RAD"] = df["A_MAX_SHAN_RAD"] - df["A_MIN_SHAN_RAD"]

df["B_MAX_SHAN_RAD"] = df[shan_rad_col_names[3:6]].max(axis=1)
df["B_MIN_SHAN_RAD"] = df[shan_rad_col_names[3:6]].min(axis=1)
df["B_RANGE_SHAN_RAD"] = df["B_MAX_SHAN_RAD"] - df["B_MIN_SHAN_RAD"]

# Adding composition averaged Shannon radii

for i in range(df.shape[0]):
    a_shan_rad_list = list(df[shan_rad_col_names[0:3]].iloc[i].dropna())
    b_shan_rad_list = list(df[shan_rad_col_names[3:6]].iloc[i].dropna())
```

```
    num_col_names = ["NUM_" + i for i in suffixes]

    a_nums = list(df[num_col_names[0:3]].iloc[i].dropna())
    b_nums = list(df[num_col_names[3:6]].iloc[i].dropna())

    a_comp_avgd = np.sum(np.multiply(a_shan_rad_list, a_nums))/8
    b_comp_avgd = np.sum(np.multiply(b_shan_rad_list, b_nums))/8

    a_shan_rad_comp_avgd.append(a_comp_avgd)
    b_shan_rad_comp_avgd.append(b_comp_avgd)

df["A_SHAN_RAD_COMP_AVGD"] = a_shan_rad_comp_avgd
df["B_SHAN_RAD_COMP_AVGD"] = b_shan_rad_comp_avgd
```

0.3.4 Adding average, difference, and ratio of A and B majority ions

[28]:
```
# Preparing new column names
placeholder_df_ab_avg = elemental_prop.iloc[:, 1:40].copy()
placeholder_df_ab_avg.columns = "AB_AVG_" + placeholder_df_ab_avg.columns.values
ab_avg_list_names = placeholder_df_ab_avg.columns

placeholder_df_diff = elemental_prop.iloc[:, 1:40].copy()
placeholder_df_diff.columns = "DIFF_" + placeholder_df_diff.columns.values
ab_diff_list_names = placeholder_df_diff.columns

placeholder_df_ratio = elemental_prop.iloc[:, 1:40].copy()
placeholder_df_ratio.columns = "RATIO_" + placeholder_df_ratio.columns.values
ratio_list_names = placeholder_df_ratio.columns
```

[29]:
```
# Adding average columns
for i in range(len(ab_avg_list_names)):
    df[ab_avg_list_names[i]] = (df[a_max_names[i]] + df[b_max_names[i]])/2

df["A_MAX_SHAN_RAD"] = df["A_MAX"]
df["B_MAX_SHAN_RAD"] = df["B_MAX"]

# Adding average of Shannon radius
df["A_MAX_SHAN_RAD"] = df["A_MAX_SHAN_RAD"].replace(a_shan_rad_sites,␣
  ↪a_shan_rad)
df["B_MAX_SHAN_RAD"] = df["B_MAX_SHAN_RAD"].replace(b_shan_rad_sites,␣
  ↪b_shan_rad)
df["AB_SHANRAD_AVG"] = (df["A_MAX_SHAN_RAD"] + df["B_MAX_SHAN_RAD"])/2
```

[30]:
```
# # Adding difference columns
# for i in range(len(ab_diff_list_names)):
```

```
#     df[ab_diff_list_names[i]] = df[a_max_names[i]] - df[b_max_names[i]]

# Adding difference of Shan rad between A and B
df["AB_SHANRAD_DIFF"] = df["A_MAX_SHAN_RAD"] - df["B_MAX_SHAN_RAD"]
```

[31]:
```
# Taking ratio of A and B
for i in range(len(ratio_list_names)):
        df[ratio_list_names[i]] = np.divide(df[a_max_names[i]],
                                            df[b_max_names[i]],
                                            out=np.
 ↪zeros_like(df[a_max_names[i]]), # Ensuring no division by zero
                                            where = df[b_max_names[i]]!=0)

# Taking ratio of A and B
df["AB_SHAN_RA_RATIO"] = np.divide(df["A_MAX_SHAN_RAD"],
                                   df["B_MAX_SHAN_RAD"],
                                   out=np.zeros_like(df["B_MAX_SHAN_RAD"]), #␣
 ↪Ensuring no division by zero
                                   where = df["B_MAX_SHAN_RAD"]!=0)
```

[32]:
```
# Calculating t and octahedral factor based on new comp avgd rations

df["GOLDSCHMIDT_COMP_AVGD"] = (df["A_SHAN_RAD_COMP_AVGD"] + 1.4)/(np.
 ↪sqrt(2)*(df["B_SHAN_RAD_COMP_AVGD"]+1.4))
df["OCTAHEDRAL_COMP_AVGD"] = df["B_SHAN_RAD_COMP_AVGD"] / 1.4
```

[33]:
```
# df.to_csv("data/calculated_features.csv")
df = pd.read_csv("data/calculated_features.csv")
```

[34]:
```
df
```

[34]:
```
      COMPOSITION A_SITE_1 A_SITE_2 A_SITE_3 B_SITE_1 B_SITE_2 B_SITE_3  \
0      Ba1Sr7V8O24       Ba       Sr      NaN        V      NaN      NaN
1      Ba2Sr6V8O24       Ba       Sr      NaN        V      NaN      NaN
2      Sr4Ca4V8O24       Sr       Ca      NaN        V      NaN      NaN
3      Sr4Dy4V8O24       Sr       Dy      NaN        V      NaN      NaN
4      Sr4Gd4V8O24       Sr       Gd      NaN        V      NaN      NaN
…             …        …        …        …        …        …        …
1921     Ba8Pd8O24       Ba      NaN      NaN       Pd      NaN      NaN
1922     Ba8Pt8O24       Ba      NaN      NaN       Pt      NaN      NaN
1923     Ba8Re8O24       Ba      NaN      NaN       Re      NaN      NaN
1924     Ba8Rh8O24       Ba      NaN      NaN       Rh      NaN      NaN
1925     Ba8Ru8O24       Ba      NaN      NaN       Ru      NaN      NaN

     X_SITE  NUM_ELEMS  ENERGY_ABOVE_HULL  …  RATIO_CTE  RATIO_SP_HEAT_CAP  \
0         0          4          29.747707  …   2.678571           0.613497
1         0          4          42.133507  …   2.678571           0.613497
```

```
2         0         4          66.990941  …   2.678571          0.613497
3         0         4         183.475519  …   2.678571          0.613497
4         0         4         290.221571  …   2.678571          0.613497
…      …        …                  …  …     …                …
1921      0         3          90.406834  …   1.745763          0.836066
1922      0         3         156.627930  …   2.340909          1.569231
1923      0         3         158.218632  …   3.322581          1.489051
1924      0         3         114.525327  …   2.512195          0.842975
1925      0         3          56.673837  …   3.218750          0.857143

      RATIO_THERMAL_COND  RATIO_CONDUCTIVITY  RATIO_HEAT_OF_FUSION  \
0               0.114984            1.250000              0.359649
1               0.114984            1.250000              0.359649
2               0.114984            1.250000              0.359649
3               0.114984            1.250000              0.359649
4               0.114984            1.250000              0.359649
…                    …                  …                    …
1921            0.256267            0.280000              0.478495
1922            0.256983            0.297872              0.407426
1923            0.384134            0.482759              0.242360
1924            0.122667            0.121739              0.368107
1925            0.157265            0.187919              0.313871

      RATIO_HEAT_OF_VAP  RATIO_ELECTRONEGATIVITY  AB_SHAN_RA_RATIO  \
0              0.306470                 0.582822          2.482759
1              0.306470                 0.582822          2.482759
2              0.306470                 0.582822          2.482759
3              0.306470                 0.582822          2.482759
4              0.306470                 0.582822          2.482759
…                   …                        …                 …
1921           0.356471                 0.404545          2.617886
1922           0.274660                 0.390351          2.576000
1923           0.198275                 0.468421          2.555556
1924           0.283009                 0.390351          2.683333
1925           0.246931                 0.404545          2.596774

      GOLDSCHMIDT_COMP_AVGD  OCTAHEDRAL_COMP_AVGD
0                  1.021823              0.414286
1                  1.029412              0.414286
2                  0.996378              0.414286
3                  0.950487              0.414286
4                  0.954773              0.414286
…                      …                    …
1921               1.056274              0.439286
1922               1.051057              0.446429
1923               1.048469              0.450000
1924               1.064196              0.428571
```

```
1925                1.053659              0.442857

[1926 rows x 933 columns]
```

0.4 Univariate analysis

[35]:
```python
def histogram_w_avg(bins, feature, target):

    # Duplicate dataframe
    df_sample = df_.copy()
    binned_feature = feature + "_BINNED"
    avg_target = "AVG_"+target

    labels = np.linspace(df[feature].min(), df[feature].max(), bins)

    # Binning
    df_sample[binned_feature] = pd.cut(df_sample[feature], bins, labels=labels)
    avg_score = df_sample.groupby(binned_feature)[target].mean().
 ↪rename(avg_target).reset_index()
    count = df_sample.groupby(binned_feature)[target].count().rename("COUNT").
 ↪reset_index()

    # Create figure with secondary y-axis
    fig = make_subplots(specs=[[{"secondary_y": True}]])

    fig.add_trace(
        go.Bar(
            x=count[binned_feature],
            y=list(count["COUNT"]),
            name = "Count",
            showlegend=False
            ),
            secondary_y=False,
    )

    fig.add_trace(
        go.Scatter(
            x=avg_score[binned_feature],
            y=avg_score[avg_target],
            name="Average "+target,
            connectgaps=True,
            showlegend=False),
            secondary_y=True,
    )

    # Add figure title
```

```
    fig.update_layout(
        title_text="Average " + target + " with " + feature,
        width=600,
        height=450
    )

    # Set x-axis title
    fig.update_xaxes(title_text=feature)

    # Set y-axes titles
    fig.update_yaxes(title_text="Count", secondary_y=False)
    fig.update_yaxes(title_text="Average "+target, secondary_y=True)

    # Updating layout
    fig.update_layout

    fig.show()
```

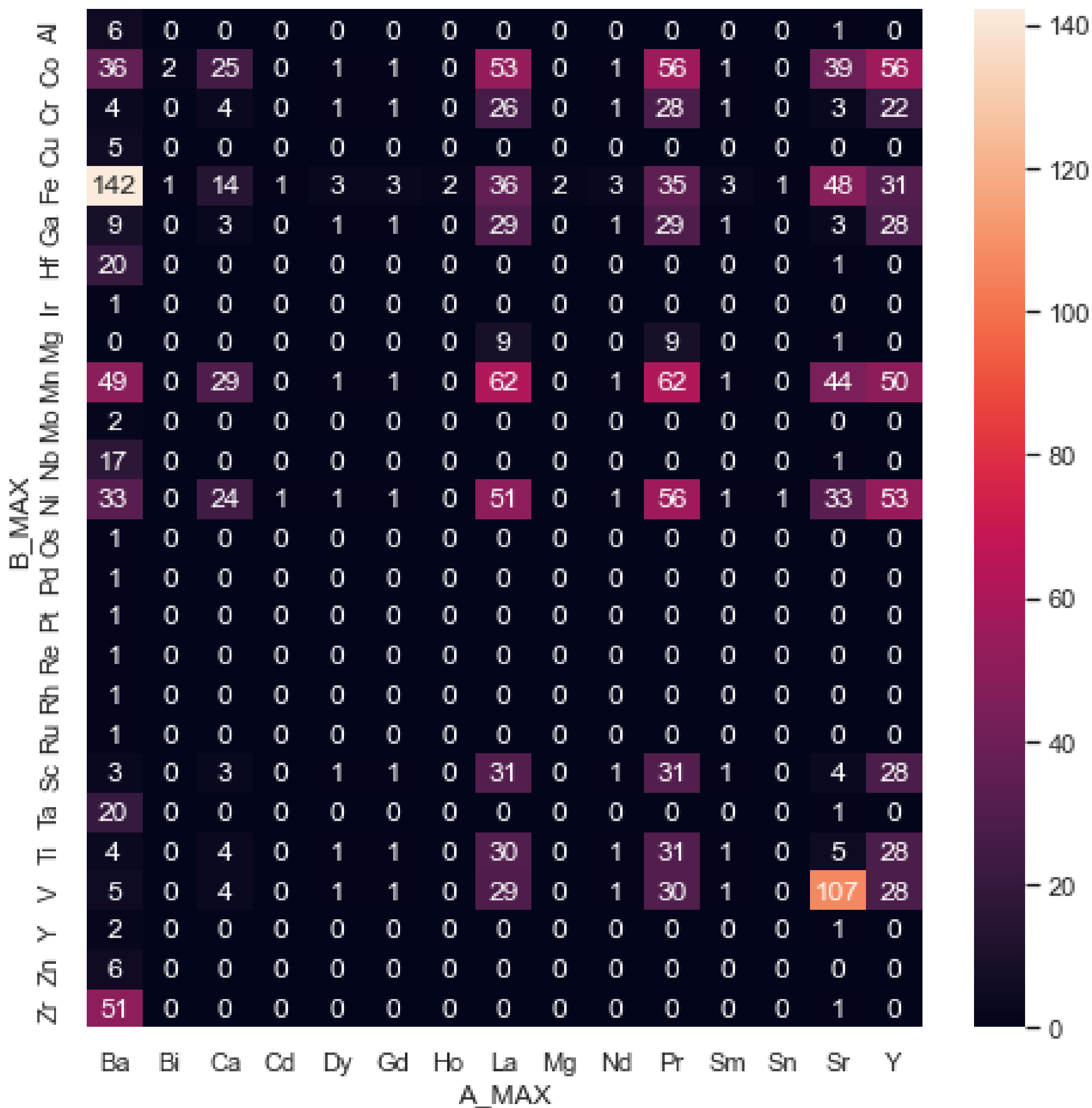
0.4.1 Distribution of ions

[36]:
```
matrix_count = df.groupby(["B_MAX", "A_MAX"]).size().unstack(fill_value=0)
```

Below shows a heatmap of the frequency distribution of the majority elements. Some insights: - Ba and Sr are the most common A cations - Fe is the most common B cation - Pr, Dy, Gd, and Ho are the only rare earth elements, and they are all A cations - The most common alkaline earth elements are Ba and Ca - V, Cr, Ti, Ga, and Sc are moderately represented as B cations - Bi, Cd, Mg, Ce, and Er are the least represented A cations

[37]:
```
sns.heatmap(matrix_count, annot=True, fmt="d")
```

[37]: <AxesSubplot:xlabel='A_MAX', ylabel='B_MAX'>

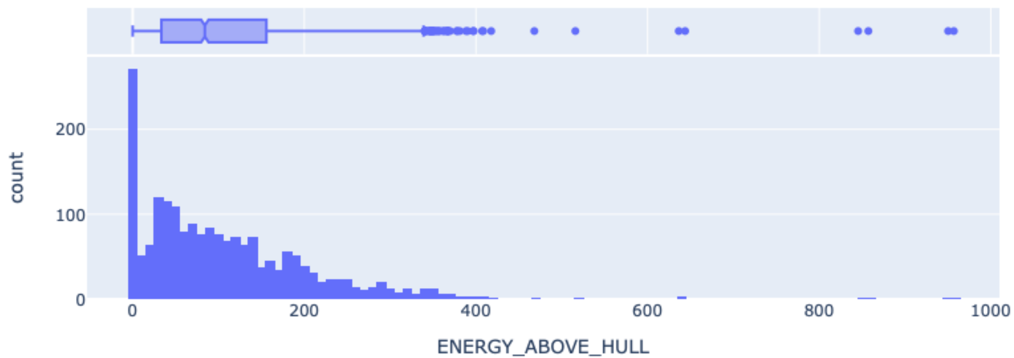


0.4.2 Energy above hull (E_{hull})

The histogram below shows the distribution of E_{hull} for all perovskites in the dataset. As can be seen, there are are several outliers in the target that have to be removed from the dataset. Below, `E_{hull}` is filtered to only keep rows with z-scores less than 3.

Nevertheless, E_{hull} has a left skew with majority of the dataset composed of low E_{hull} perovskites.

[38]:
```
fig = px.histogram(df, x="ENERGY_ABOVE_HULL",
                   marginal="box", # or violin, rug
                   hover_data=df.columns)
fig.show()
```



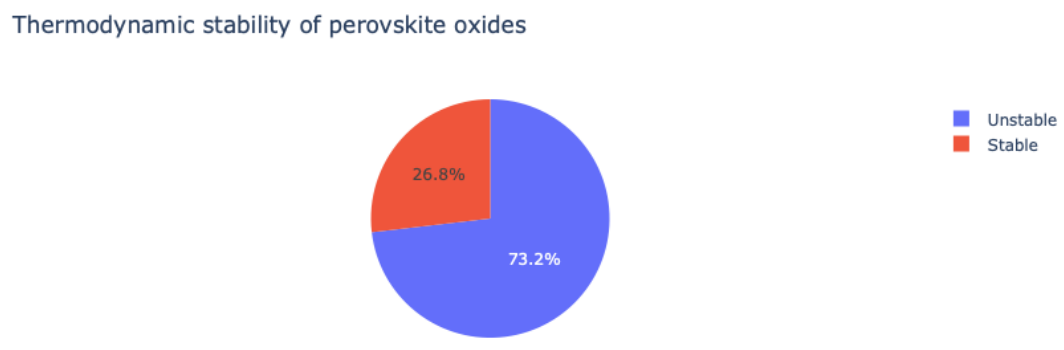
[39]:
```
df = df[abs(zscore(df["ENERGY_ABOVE_HULL"])<3)]
```

After removing outliers, the dataset is composed of 1915 perovskite crystals 26.8% of which are stable, and 73.2% are unstable. The "stability" of these perovskite crystals are determined using a threshold set by Wu et al. in their 2013 work when they found that 80% of the crystals in their dataset that have $E_{hull} < 36.0$ meV/atom are thermodynamically stable.

[40]:
```
df.shape[0]
```

[40]: 1915

[41]:
```
df_ = df.copy()

df_["IS_UNSTABLE"] = df_["ENERGY_ABOVE_HULL"]>36
e_above_hull = df_.groupby(df_["IS_UNSTABLE"])["COMPOSITION"].count().
  ↪rename("COUNT")
pie = px.pie(values=e_above_hull, names=["Stable", "Unstable"],␣
  ↪title="Thermodynamic stability of perovskite oxides")
pie.show()
```



0.4.3 Goldschmidt tolerance factor (t) and stability

After the determination of crystal structure based on Goldschmidt factor, the ratio of stable and unstable crystals per structure is plotted in the bar graph below. Interestingly, crystals that are predicted to be orthorhombic/rhombohedral are most likely to be stable, while hexagonal/tetragonal crystals are least likely. Cubic crystals are somewhere in between these two when it was discussed earlier that cubic crystals, in theory, are the most thermodynamically stable lattice structure for perovskites.

The high stable rate for orthorhombic/rhombohedral crystals can be attributed to the fact that they are underrepresented in the dataset.

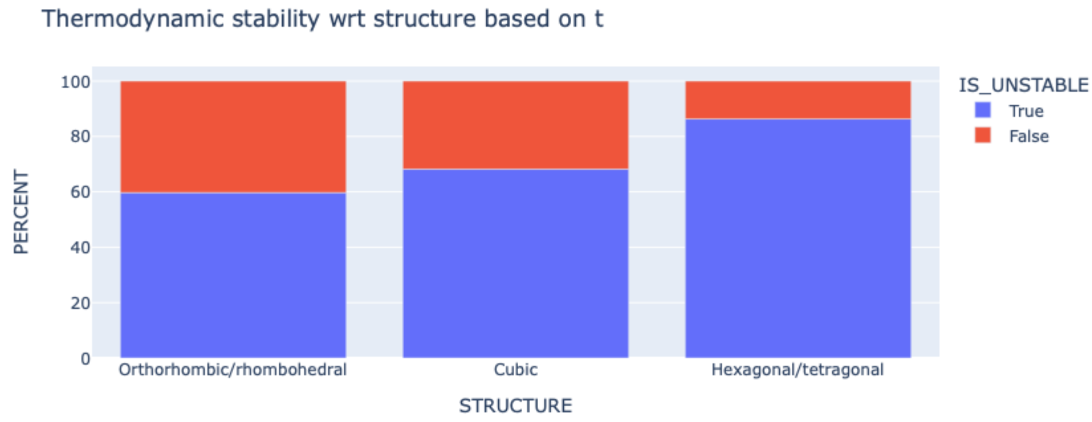
[42]:
```
goldschmidt = [i for i in df.columns if "GOLD" in i]
goldschmidt
```

[42]:
```
['GOLDSCHMIDT_TF', 'GOLDSCHMIDT_SHAN', 'GOLDSCHMIDT_COMP_AVGD']
```

[43]:
```
df_["STRUCTURE"] = pd.cut(x=df_["GOLDSCHMIDT_COMP_AVGD"],
                          bins=[0.71, 0.9, 1, 10],
                          labels=["Orthorhombic/rhombohedral", "Cubic",␣
 ↪"Hexagonal/tetragonal"])
```

[44]:
```
grouped_structures = df_.groupby("STRUCTURE")["IS_UNSTABLE"].
 ↪value_counts(normalize=True).mul(100).rename('PERCENT').reset_index()

fig = px.bar(grouped_structures,
             x="STRUCTURE",
             y="PERCENT",
             color="IS_UNSTABLE")

fig.update_layout(title_text="Thermodynamic stability wrt structure based on t")
```

```
fig.show()
```

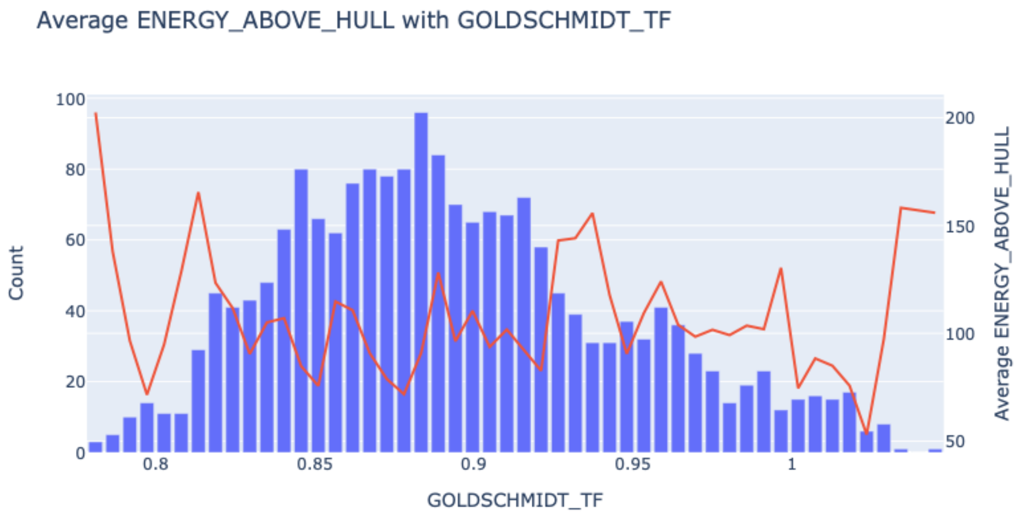


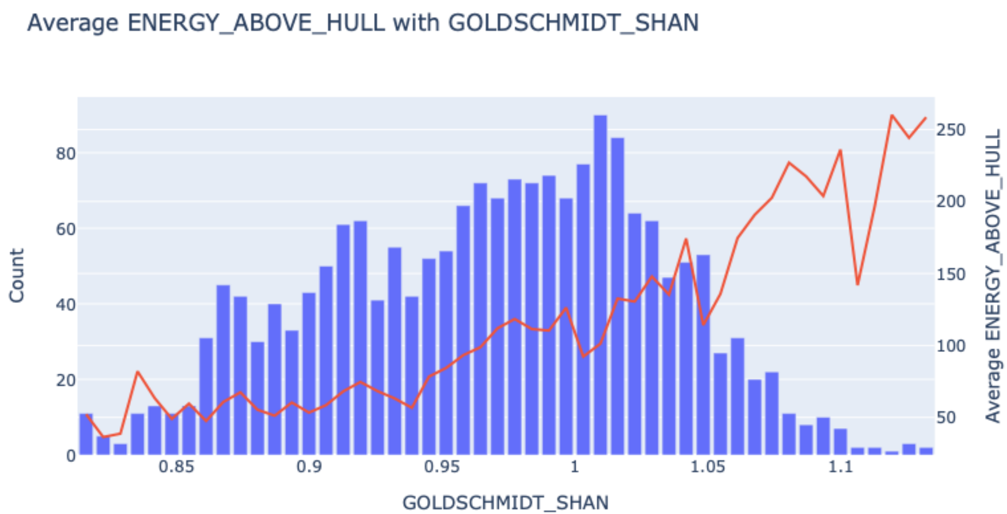
[45]:
```
df_["STRUCTURE"].value_counts()
```

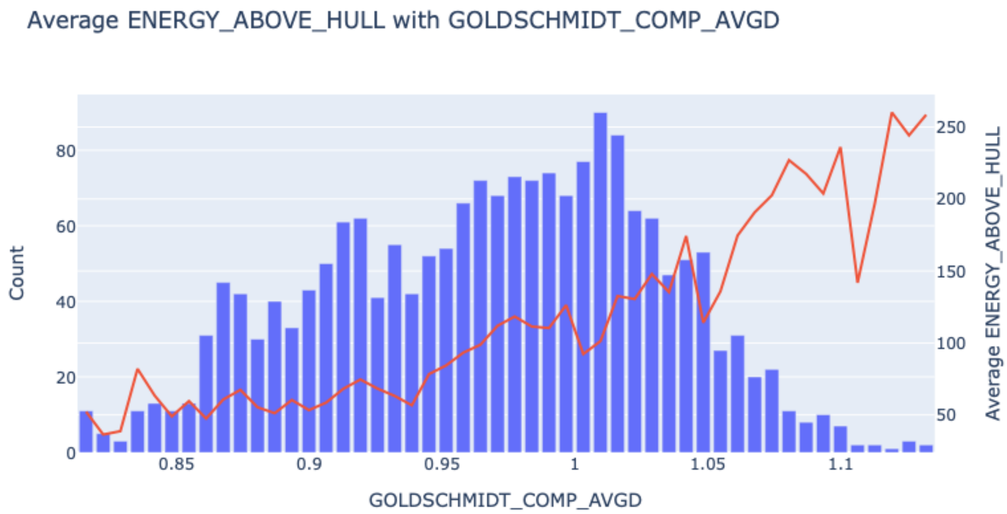
[45]:
```
Cubic                        947
Hexagonal/tetragonal         665
Orthorhombic/rhombohedral    303
Name: STRUCTURE, dtype: int64
```

The plot below shows the average E_{hull} with respect to t, superimposed on the histogram of t. As can be seen, t of the perovskites have a relatively normal distribution. Interestingly, the average E_{hull} steadily increases with t. This is consistent with earlier discussion stating that $t > 1.0$ causes thermodynamic instability.

[46]:
```
for i in goldschmidt:
    histogram_w_avg(50, i, "ENERGY_ABOVE_HULL")
```


Average ENERGY_ABOVE_HULL with GOLDSCHMIDT_TF
Count
Average ENERGY_ABOVE_HULL
GOLDSCHMIDT_TF
0
20
40
60
80
100
50
100
150
200
0.8
0.85
0.9
0.95
1


Average ENERGY_ABOVE_HULL with GOLDSCHMIDT_SHAN
Count
Average ENERGY_ABOVE_HULL
GOLDSCHMIDT_SHAN
0
20
40
60
80
50
100
150
200
250
0.85
0.9
0.95
1
1.05
1.1



0.4.4 Octahedral factor (μ) and stability

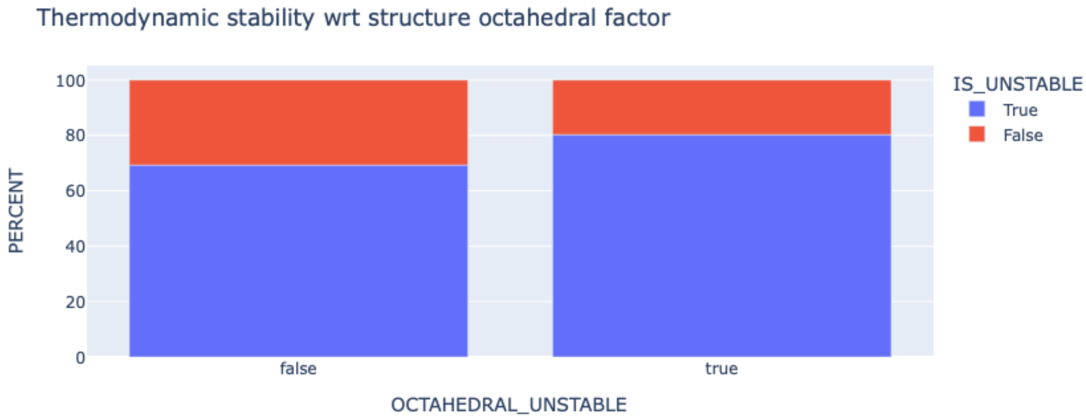
The stability based on octahedral factor was determined for each perovskite crystal. As can be seen below, crystals predicted to be stable based on their octahedral factor are more likely to be unstable. This is perhaps due to the assumption of mono-ionic crystals.

[47]:
```
octahedral = [i for i in df.columns if "OCTAHEDRAL" in i]
octahedral
```

[47]:
```
['OCTAHEDRAL_FACTOR', 'OCTAHEDRAL_SHAN', 'OCTAHEDRAL_COMP_AVGD']
```

[48]:
```
df_["OCTAHEDRAL_UNSTABLE"] = df_["OCTAHEDRAL_COMP_AVGD"] < 0.41
```

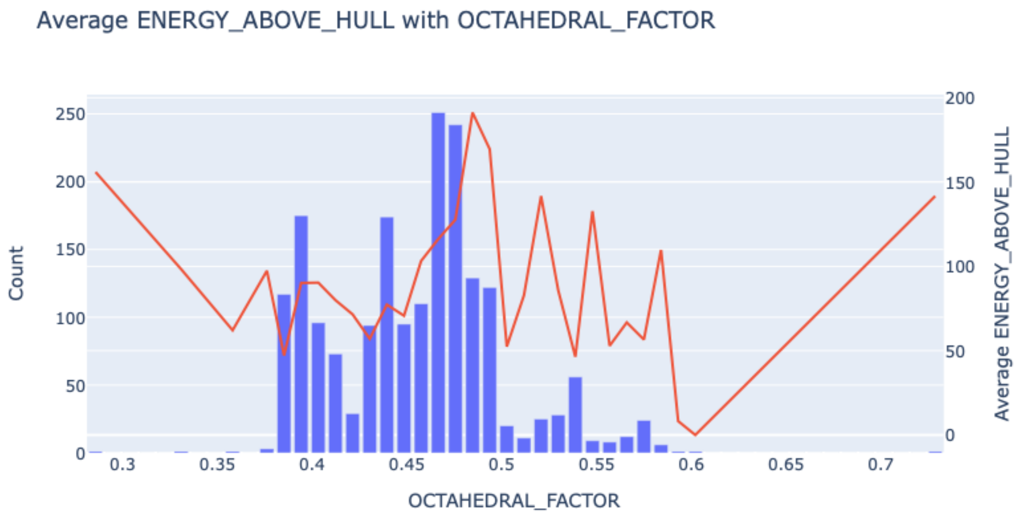
[49]:
```
grouped_structures = df_.groupby("OCTAHEDRAL_UNSTABLE")["IS_UNSTABLE"].
 ↪value_counts(normalize=True).mul(100).rename('PERCENT').reset_index()

fig = px.bar(grouped_structures,
             x="OCTAHEDRAL_UNSTABLE",
             y="PERCENT",
             color="IS_UNSTABLE")

fig.update_layout(title_text="Thermodynamic stability wrt structure octahedral␣
 ↪factor")

fig.show()
```



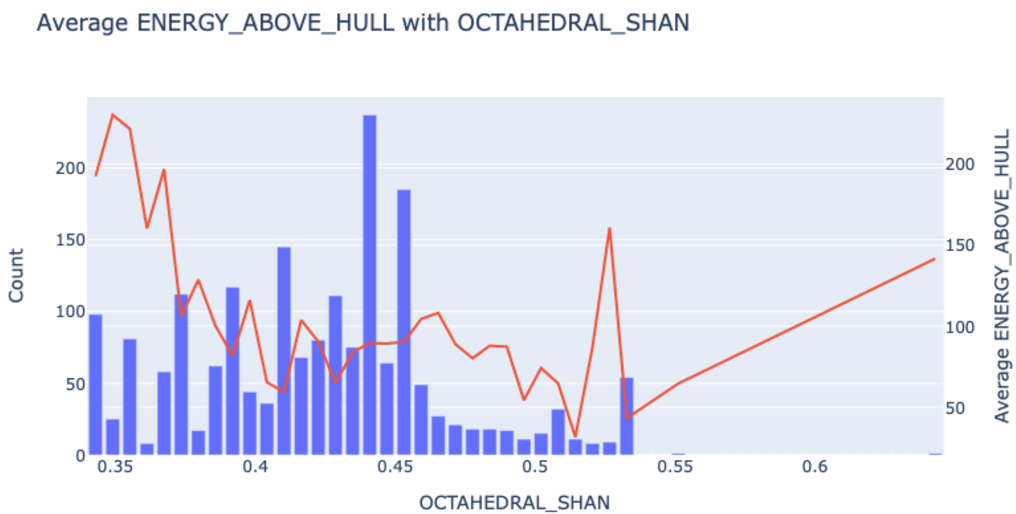
The plot below shows the average E_{hull} with respect to μ, superimposed on the histogram of μ. Interestingly, the average E_{hull} stays steady with μ. Moreover, among all features that signify the μ, `OCTAHEDRAL_FACTOR` has the most noise. This might be due to the fact that, unlike `OCTAHEDRAL_SHAN` and `OCTAHEDRAL_COMP_AVGD`, `OCTAHEDRAL_FACTOR` was calculated from the ionic radii of the ions, and not their Shannon radii.

The ionic radii of an ion is its radius in its monatonic version as an ionic crystal structure. The ionic radii of Sr for example, is its radius if it where an ionic crystal made up of only Sr. As mentioned previously, perovskite crystals are made up of multiple ions with varying oxidation states, and thus, varying coordination numbers as well. The Shannon radius is able to reflect these as it is the radius of ions depending on their oxidation states and coordination numbers. Thus, calculations based on the Shannon radii as opposed to the ionic radii are able to reflect the ionic diversity of the perovskite crystals.

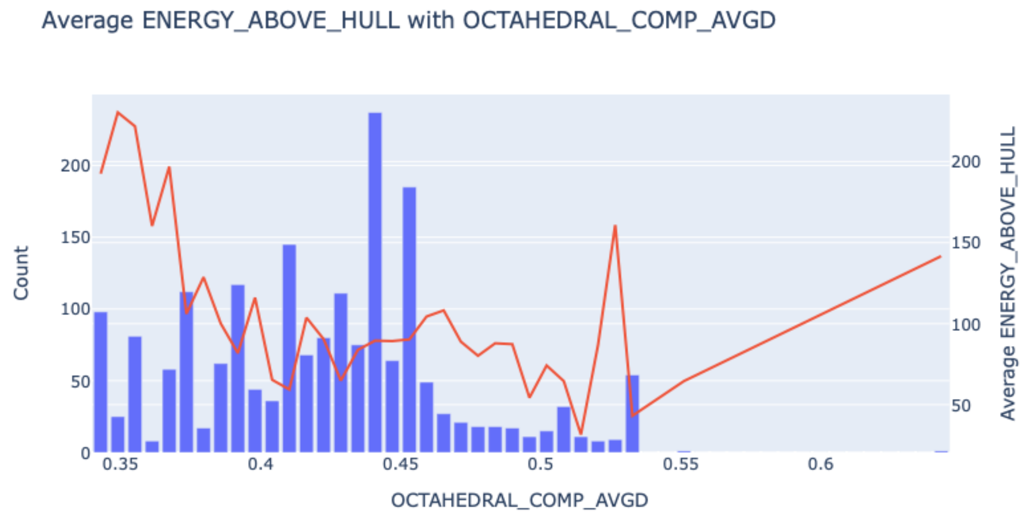
[50]:
```python
for i in octahedral:
    histogram_w_avg(50, i, "ENERGY_ABOVE_HULL")
```



Average ENERGY_ABOVE_HULL with OCTAHEDRAL_FACTOR
Count
Average ENERGY_ABOVE_HULL
OCTAHEDRAL_FACTOR



Average ENERGY_ABOVE_HULL with OCTAHEDRAL_SHAN
Count
Average ENERGY_ABOVE_HULL
OCTAHEDRAL_SHAN



0.4.5 Atomic weight of A cation with stability

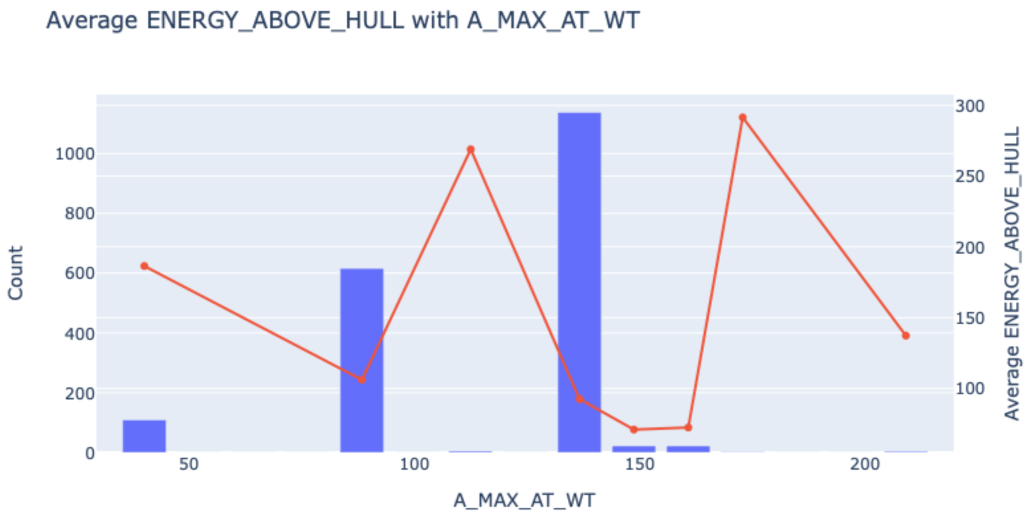
Zhang et al. has shown that the stability of perovskites increase gradually as A becomes heavier i.e. E_{hull} decreases with atomic weight of A [3]. Below we show a gradual decrease in average E_{hull} as atomic weight of A increases. Below, the average E_{hull} is plotted against both `A_MAX_AT_WT` and `A_WT_AVG_AT_WT` which corresponds to the atomic weights of the majority ion in the A-site, and the composition averaged atomic weight of all A-site ions, respectively.

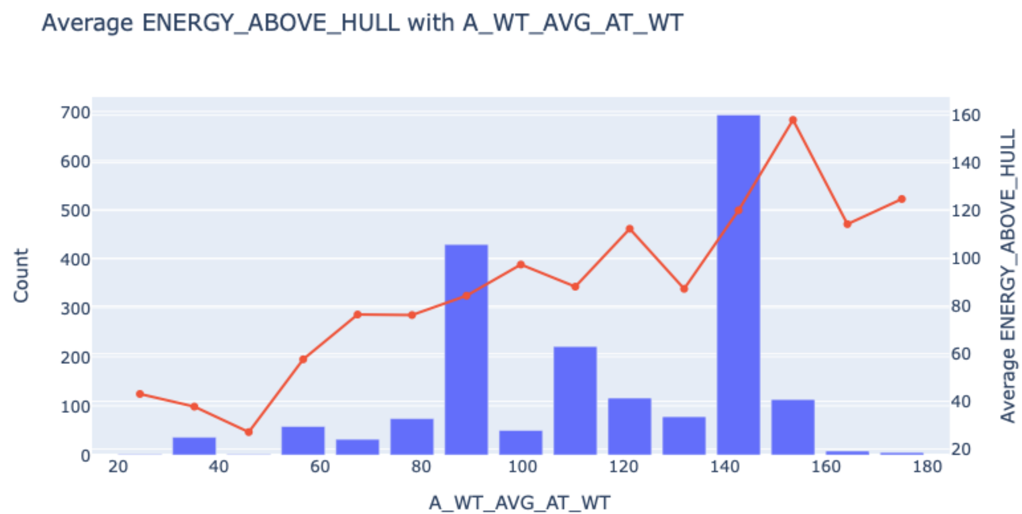
As can be seen, both have a general upward trend with increasing atomic weight. However, the composition averaged atomic weight shows a smoother plot. This is because composition average is much better at representing the combination of different elements in the A-site of the perovskite as opposed to `A_MAX_AT_WT` which only represents one ion in a crystal that is composed of multiple ions that are tightly coordinated with each other.

[51]:
```
a_wt = [i for i in df.columns if "_AT_WT" in i and "B" not in i][0:2]
a_wt
```

[51]: `['A_MAX_AT_WT', 'A_WT_AVG_AT_WT']`

[52]:
```
for i in a_wt:
    histogram_w_avg(15, i, "ENERGY_ABOVE_HULL")
```





0.4.6 Atomic packing factor and Sun factor and stability

The atomic packing factor (η) of a crystal quantifies the density of ions within the unit cell of a crystal. It can be calculated using the following formula:

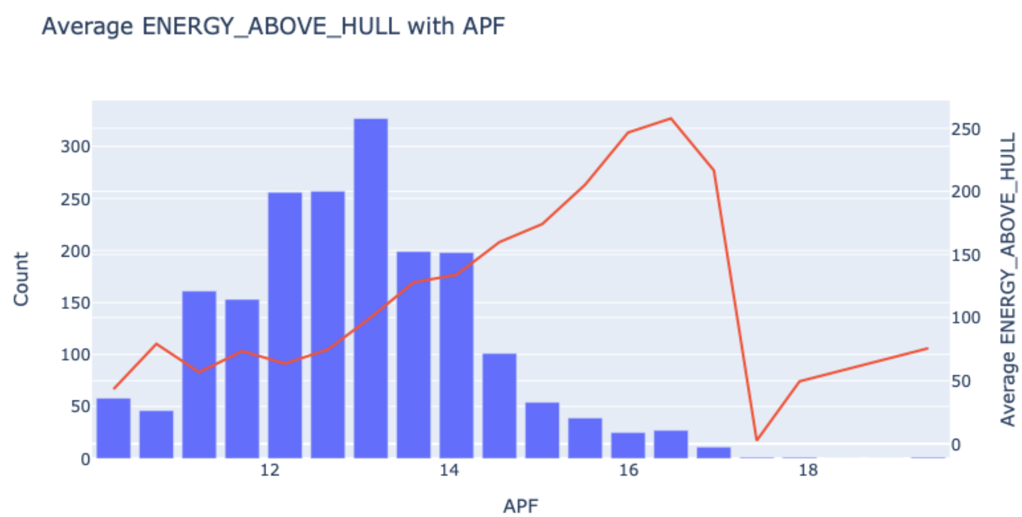
$$\eta = \frac{\frac{M_A}{\rho_A} + \frac{M_B}{\rho_B} + 3 \cdot \frac{M_X}{\rho_X}}{a^3}$$

where M is for the molar mass of the ion, ρ is the density of the ion, and a is the lattice parameter of the unit cell which is equal to the bond length between B and O.

As can be seen below, average E_{hull} steadily increases with respect to APF.

[53]:
```
df["APF"] = (np.divide(df["A_MAX_AT_WT"],
                       df["A_MAX_DENSITY"],
                       out=np.zeros_like(df["A_MAX_DENSITY"]),
                       where = df["A_MAX_DENSITY"]!=0) +
             np.divide(df["B_MAX_AT_WT"],
                       df["B_MAX_DENSITY"],
                       out=np.zeros_like(df["B_MAX_DENSITY"]),
                       where = df["B_MAX_DENSITY"]!=0) + (3*22.4))/(df["B_O"])**3

df_["APF"] = df["APF"]
```

[54]:
```
histogram_w_avg(20, "APF", "ENERGY_ABOVE_HULL")
```



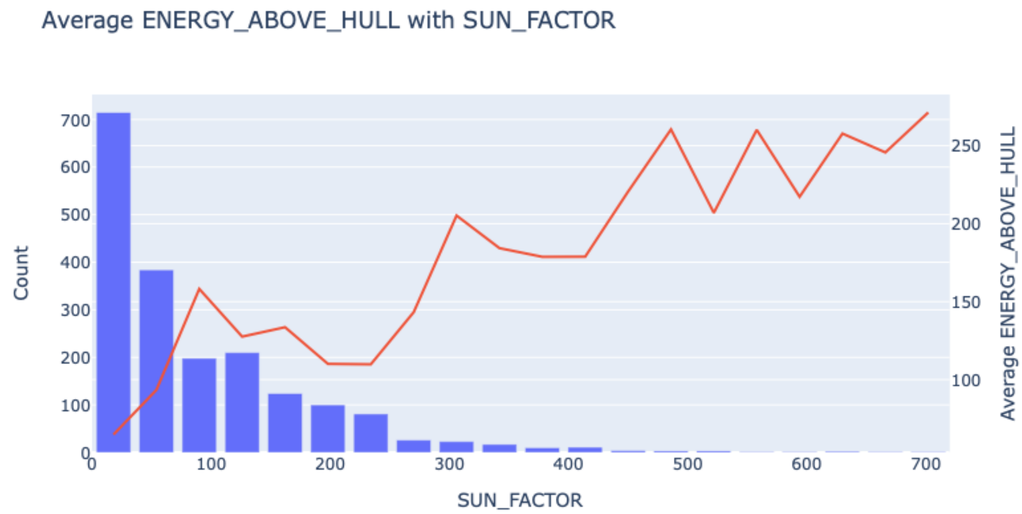
The Sun factor (SF) is a quantity introduced in the 2017 paper of Sun et al [4]. It can be derived from the APF, t, and μ as shown below:

$$SF = (\mu + t)^{\eta}$$

Sun et al. has shown that E_{hull} varies directly with SF which is also shown on the below plot. One can see below that E_{hull} increases with SF.

[55]:
```
df["SUN_FACTOR"] = (df["GOLDSCHMIDT_COMP_AVGD"] +␣
  ↪df["OCTAHEDRAL_COMP_AVGD"])**df["APF"]
df_["SUN_FACTOR"] = df["SUN_FACTOR"]
```

[56]:
```
histogram_w_avg(20, "SUN_FACTOR", "ENERGY_ABOVE_HULL")
```



0.5 Machine learning modeling

Below, several candidate models are tested on their RMSE.

[57]:
```
# Imputing NaN values in the dataset with zero
df = df.replace(np.nan, 0)

# Removing string-type features as well as ENERGY_ABOVE_HULL and FORMATION␣
↪ENERGY
string_columns_non_y = [i for i in df.columns if df[i].dtypes != "object" and i␣
↪not in ["ENERGY_ABOVE_HULL", "FORMATION_ENERGY"]]

X = df[string_columns_non_y]
y = df["ENERGY_ABOVE_HULL"]

# Train test split
X_train, X_test, y_train, y_test = train_test_split(X, y,
                                                    test_size=0.20,
                                                    random_state=42)
```

[58]:
```
scaler = StandardScaler()
X_train_ = scaler.fit_transform(X_train)
```

[59]:
```
lr_i = LinearRegression()
svr_i = SVR()
rf_i = RandomForestRegressor()
mlp_i = MLPRegressor()
```

```
xgb_i = XGBRegressor()

estimator_list = [lr_i, svr_i, rf_i, mlp_i, xgb_i]
estimator_names = ["Linear regression",
                   "SVM",
                   "Random forest regressor",
                   "Multi-layer perceptron",
                   "XGBoost"]
```

[60]:
```
def model_selection(estimator, name):

    if name == "XGBoost":
        input = X_train
    else:
        input = X_train_

    scores = cross_validate(estimator,
                            input,
                            y_train,
                            cv=5,
                            scoring="neg_mean_squared_error")

    print("")
    print(f"{name} Mean Scores")
    print(f"RMSE: {(-scores['test_score'].mean())**(1/2.0)}")
    print("")

    return (-scores['test_score'].mean())**(1/2.0)
```

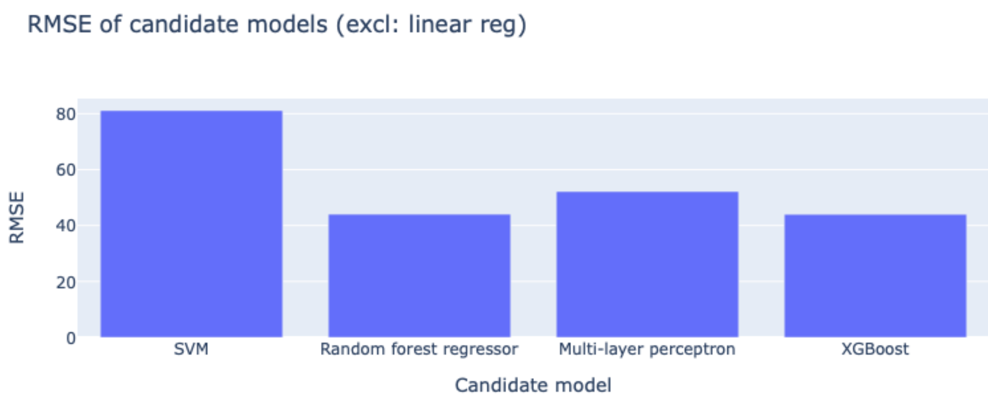
As can be seen below, XGBoost performed the best among all the candidate models with an RMSE of 43.504.

[61]:
```
model_scores = []

for i in range(len(estimator_list)):
    model_scores.append(model_selection(estimator_list[i], estimator_names[i]))
```

```
Linear regression Mean Scores
RMSE: 83257715981977.86


SVM Mean Scores
RMSE: 81.1075629243277


Random forest regressor Mean Scores
RMSE: 44.05873452985197
```

```
Multi-layer perceptron Mean Scores
RMSE: 52.16194065769254


XGBoost Mean Scores
RMSE: 44.01093331955984
```

[62]:
```
fig = px.bar(x=estimator_names[1:],
             y=model_scores[1:],
             title="RMSE of candidate models (excl: linear reg)")
fig.update_yaxes(title_text="RMSE")
fig.update_xaxes(title_text="Candidate model")
fig.show()
```



[63]:
```
xgb_i.fit(X_train, y_train)
f_i = xgb_i.feature_importances_
```

[64]:
```
df_fi = pd.DataFrame(list(zip(X.columns, f_i)), columns=["FEATURE",␣
 ↪"IMPORTANCE"])
df_fi = df_fi.sort_values("IMPORTANCE", ascending=False)
```

0.5.1 Hyperparameter tuning

[65]:
```
class MostImportFeaturesOnly(BaseEstimator, TransformerMixin):
    def __init__(self, f, n=None):
        self.n = n
        self.f = f
    def fit(self, X, y=None):
```

```python
        return self
    def transform(self, X, y=None):
        top = list((self.f).sort_values("IMPORTANCE").tail(self.n).index)
        return X.iloc[:, top]
```

[66]:
```python
# Doing gridsearch that searches through all possible number of features
# on top of other XGBoost hyperparameter takes too much time

# Determining optimum number of features first, then
# optimizing XGBoost hyperparameters

pipeline_best_num_features = Pipeline(
    [
        ("most_imp", MostImportFeaturesOnly(f=df_fi)),
        ("xgbr", XGBRegressor())
    ]
)
params_bnf = {'most_imp__n': np.arange(0,X_train.shape[1],5)}

gs_bnf = GridSearchCV(estimator=pipeline_best_num_features,
                      param_grid=params_bnf,
                      scoring='neg_mean_squared_error',
                      verbose=1)

gs_bnf.fit(X_train, y_train)

print("XGBoost Regression Report")
print("Best number of feature:", gs_bnf.best_params_)
print("Lowest RMSE: ", (-gs_bnf.best_score_)**(1/2.0))
```

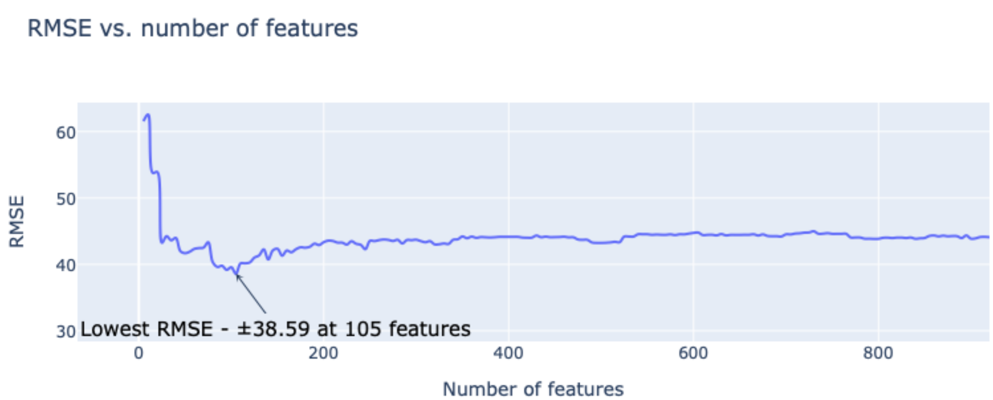
```
Fitting 5 folds for each of 185 candidates, totalling 925 fits
XGBoost Regression Report
Best number of feature: {'most_imp__n': 105}
Lowest RMSE:  38.58726423174326
```

[67]:
```python
gs_bnf_filename = "gs_bnf.pkl"
joblib.dump(gs_bnf, gs_bnf_filename)

gs_bnf = joblib.load("gs_bnf.pkl")
```

[68]:
```python
bnf = gs_bnf.best_params_['most_imp__n']
best_rmse = (-gs_bnf.best_score_)**(1/2.0)
```

[69]:
```python
num_feature = np.arange(0,X_train.shape[1],5)
mean_rmse = ((-gs_bnf.cv_results_["mean_test_score"])**(1.0/2))

fig = go.Figure()
```

```python
fig.add_trace(
    go.Scatter(x=num_feature,
               y=mean_rmse,
               line_shape="spline")
)

fig.update_layout(
        title_text="RMSE vs. number of features"
)

# Set x-axis title
fig.update_xaxes(title_text="Number of features")

# Set y-axes titles
fig.update_yaxes(title_text="RMSE")

annotation = {
    'x': bnf,
    'y': best_rmse,
    'ax': 30,
    'ay': 40,
    'text': f'Lowest RMSE - ±{round(best_rmse,2)} at {bnf} features',  # text
    'showarrow': True,  # would you want to see arrow
    'arrowhead': 3,  # which type for arrowhead
    'font': {'size': 15, 'color': 'black'},  # font style
}
fig.add_annotation(annotation)
fig.show()
```



[70]:
```
pipeline = Pipeline(
    [
        ("most_imp", MostImportFeaturesOnly(f=df_fi)),
        ("xgbr", XGBRegressor())
    ]
)

params = {
            'most_imp__n': np.arange(95,115,5),
            'xgbr__max_depth': [3,6,10],
            'xgbr__learning_rate': [0.01, 0.05, 0.1],
            'xgbr__n_estimators': [100, 500, 1000],
            'xgbr__colsample_bytree': [0.3, 0.7]
        }

gs = GridSearchCV(estimator=pipeline,
                  param_grid=params,
                  scoring='neg_mean_squared_error',
                  verbose=1)

gs.fit(X_train, y_train)
```

```
Fitting 5 folds for each of 216 candidates, totalling 1080 fits
```

[70]:
```
GridSearchCV(estimator=Pipeline(steps=[('most_imp',
                                        MostImportFeaturesOnly(f=
FEATURE  IMPORTANCE
126      B_MAX_AT_RAD    0.410524
870         AB_AVG_CTE    0.075651
174  B_MAX_ND_VALENCE    0.061857
108  B_MAX_BCC_ENERGY    0.050108
902    RATIO_ATOM_RAD    0.030229
..                ...         ...
505  ALL_MAX_B_PERIOD    0.000000
506  ALL_MIN_A_PERIOD    0.000000
507  ALL_MIN_B_PERIOD    0.000000
508    RANGE_A_PERIOD    0.000000
525   RANGE_B_VALENCE    0…
                                                     n_estimators=100,
                                                     n_jobs=None,
                                                     num_parallel_tree=None,
                                                     predictor=None,
                                                     random_state=None,
                                                     reg_alpha=None,
                                                     reg_lambda=None, …))]),
             param_grid={'most_imp__n': array([ 95, 100, 105, 110]),
                         'xgbr__colsample_bytree': [0.3, 0.7],
```

```
                         'xgbr__learning_rate': [0.01, 0.05, 0.1],
                         'xgbr__max_depth': [3, 6, 10],
                         'xgbr__n_estimators': [100, 500, 1000]},
             scoring='neg_mean_squared_error', verbose=1)
```

[71]:

```
gs_filename = "gs.pkl"
joblib.dump(gs, gs_filename)

gs = joblib.load("gs.pkl")

print("Best parameters:", gs.best_params_)
print("Lowest RMSE: ", (-gs.best_score_)**(1/2.0))
```

```
Best parameters: {'most_imp__n': 105, 'xgbr__colsample_bytree': 0.3,
'xgbr__learning_rate': 0.05, 'xgbr__max_depth': 6, 'xgbr__n_estimators': 1000}
Lowest RMSE:  37.62030166663581
```

[72]:

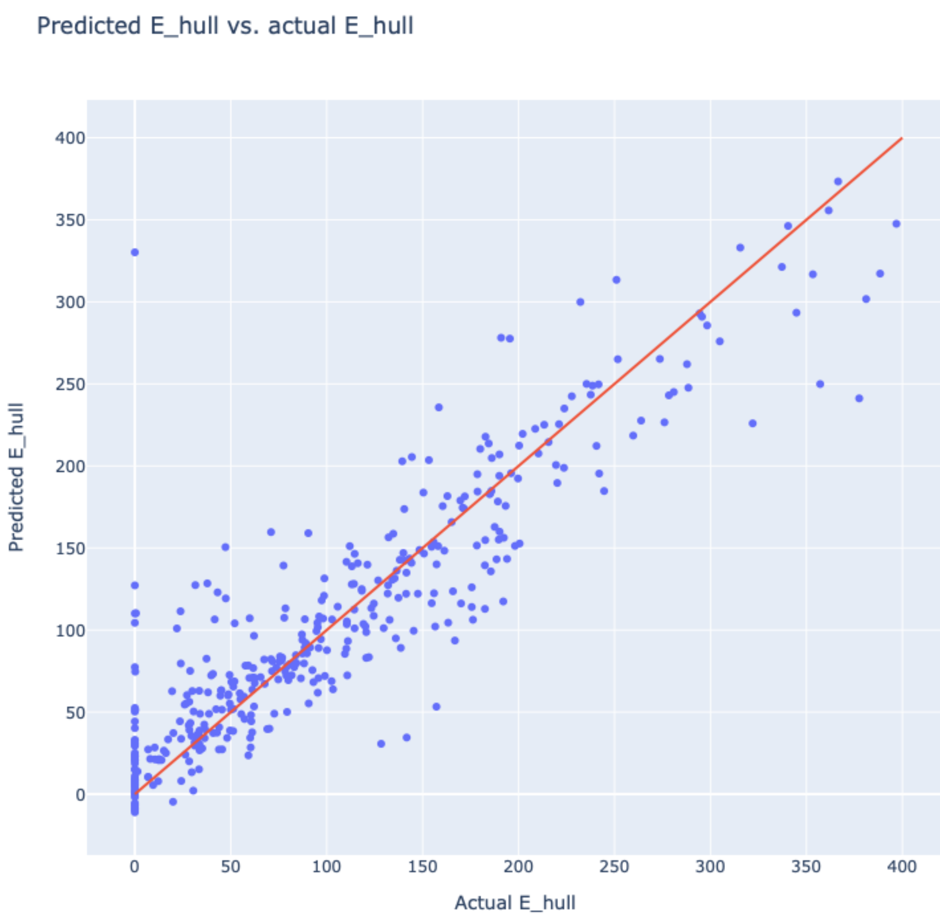
```
xgb_fin = gs.best_estimator_
y_pred = xgb_fin.predict(X_test)
rmse_fin = np.sqrt(mean_squared_error(y_test, y_pred))
print(f"RMSE of XGBRegressor on X_test: {rmse_fin}")
```

```
RMSE of XGBRegressor on X_test: 38.44647626967033
```

[73]:

```
fig = make_subplots(specs=[[{"secondary_y": True}]])

fig.add_trace(
    go.Scatter(
        x=y_test,
        y=y_pred,
        mode="markers",
        name="Predictions",
        showlegend=False
    ),

)

fig.add_trace(
    go.Scatter(
        x=np.linspace(0, 400, num=1000),
        y=np.linspace(0, 400, num=1000),
        line_shape="spline",
        name="Perfect model",
        showlegend=False
    ),
)
```

```python
# Add figure title
fig.update_layout(
        title_text="Predicted E_hull vs. actual E_hull",
        width=750,
        height=750
    )

# Set x-axis title
fig.update_xaxes(title_text="Actual E_hull")

# Set y-axes titles
fig.update_yaxes(title_text="Predicted E_hull", secondary_y=True)
fig.update_yaxes(title_text="Predicted E_hull", secondary_y=False)

fig.show()
```



References [1] Jacobs, Ryan, et al. “Material discovery and design principles for stable, high activity perovskite cathodes for solid oxide fuel cells.” Advanced Energy Materials 8.11 (2018): 1702708.

[2] Ward, Logan, et al. “A general-purpose machine learning framework for predicting properties of inorganic materials.” npj Computational Materials 2.1 (2016): 1-7.

[3] Zhang, Tao, Zenghua Cai, and Shiyou Chen. “Chemical trends in the thermodynamic stability and band gaps of 980 halide double perovskites: A high-throughput first-principles study.” ACS Applied Materials & Interfaces 12.18 (2020): 20680-20690.

[4] Sun, Qingde, and Wan-Jian Yin. “Thermodynamic stability trend of cubic perovskites.” Journal of the American Chemical Society 139.42 (2017): 14905-14908.

[5] Wu, Yabi, et al. “First principles high throughput screening of oxynitrides for water-splitting photocatalysts.” Energy & environmental science 6.1 (2013): 157-168.