Machine Learning for Materials Properties — Quynh Tran

Goal: Predict cohesive_energy_per_atom from the MatPES r2SCAN dataset using compositional and structural features.

In this project, we will:

1. Download and explore the MatPES training data.
2. Generate composition- and structure-based features.
3. Train and tune a regularized linear model (Ridge) and several tree/boosting models.
4. Compare performance (MSE) on train/validation and Kaggle test set.
5. Analyze feature importances to gain materials-science insight.

```
!pip install -q kaggle ijson matminer scikit-learn pymatgen datasets

import pandas as pd
import numpy as np
import matplotlib.pyplot as plt
import gzip, ijson, csv
import os
from pymatgen.core.composition import Composition
from matminer.featurizers.composition import ElementProperty
from sklearn.model_selection import train_test_split, GridSearchCV, RandomizedSearchCV
from sklearn.linear_model import Ridge, RidgeCV
from sklearn.metrics import mean_squared_error
from sklearn.impute import SimpleImputer
from sklearn.pipeline import Pipeline
from sklearn.ensemble import RandomForestRegressor, HistGradientBoostingRegressor
```

```
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```

2. Data Loading & Exploratory Data Analysis

We first load the provided CSVs and plot the distribution of **cohesive_energy_per_atom** (E_c) to understand its range and skew.

- **Training set:** 381 264 entries
- **Test set:** 6 345 entries
- E_c ranges from roughly −13.7 to +3.7 eV/atom, with a mean near −4 eV/atom.

We will not peek at test labels; they will be scored by Kaggle.

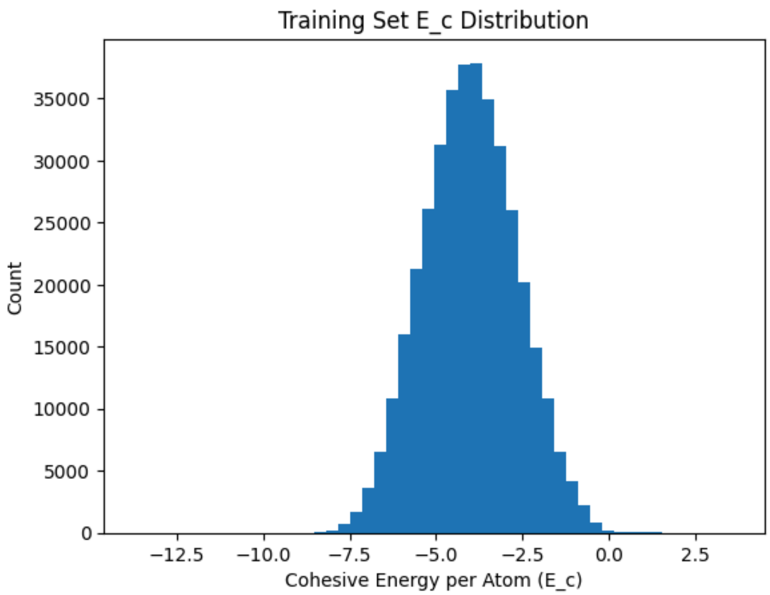
1. (2 points) Download the data files from Kaggle. Create a Pandas DataFrame with the training data.

```
from google.colab import files


train_url = 'https://raw.githubusercontent.com/nhitran161/Machine-Learning-for-Materials-Properties/refs/heads/main/
test_url = 'https://raw.githubusercontent.com/nhitran161/Machine-Learning-for-Materials-Properties/refs/heads/main/n

# Load the data
try:
    train_df = pd.read_csv(train_url)
    test_df = pd.read_csv(test_url)
    print("✅ Data successfully loaded from GitHub!")
    print(f"Train shape: {train_df.shape}")
except:
    print("❌ Error: Could not load data. Check your Raw links.")
```

```
✅ Data successfully loaded from GitHub!
Train shape: (381264, 3)
```

2.(3 points) Plot histograms of E_c of the training data.

```
plt.hist(train_df['cohesive_energy_per_atom'], bins=50)
plt.xlabel('Cohesive Energy per Atom (E_c)')
plt.ylabel('Count')
plt.title('Training Set E_c Distribution')
plt.show()
```



3. Feature Engineering

3.1 Composition-based Features

We reuse the Magpie elemental properties to generate >100 composition descriptors per structure (mean, max, range of atomic radii, electronegativities, etc.). These were computed offline and saved in `train_comp.csv` / `test_comp.csv`.

3.2 Structure-based Features

From the MatPES JSON we extracted for each structure:

- **Density** and **density_atomic**
- **Symmetry** (crystal system, space-group symbol, number, point group) as one-hot encoded categories
- **Charge**, **lattice parameters** ($a, b, c, \alpha, \beta, \gamma$), and **number of atomic sites**
  We then merged these into `train_feat`/`test_feat`.

↳ 6 cells hidden

4. Modeling

We split the 381 264-entry training set into an 80/20 train/validation split. All features (composition + structure) were imputed (mean) and standardized before modeling.

4.1 Ridge Regression (Polynomial Features)

- **Pipeline**: 2nd-degree `PolynomialFeatures` → `StandardScaler` → `RidgeCV`
- **Alphas**: 30 log-spaced between 1e-4 and 1e4, 5-fold CV
- **Validation MSE**: *0.308*

4.2 Hyperparameter Tuning for HistGradientBoostingRegressor (NANO281)

We focus our main tree-based model on a histogram-based GBM, which:

- Handles missing values natively
- Leverages fast histogram binning

- Often outperforms RandomForest on large datasets

Search setup:

- `max_iter` ∈ {100, 200, 300}
- `learning_rate` ∈ {0.01, 0.05, 0.1}
- `max_depth` ∈ {None, 10, 15}
- `l2_regularization` ∈ {0.0, 0.1, 1.0, 10.0}
- 12 random trials (`RandomizedSearchCV`), 3-fold CV, scoring=`neg_mean_squared_error`

```
# Example of the tuning cell
search = RandomizedSearchCV(
    Pipeline([
      ('imputer', SimpleImputer(strategy='mean')),
      ('hgb',     HistGradientBoostingRegressor(random_state=42, early_stopping=False))
    ]),
    {
      'hgb__max_iter': [100,200,300],
      'hgb__learning_rate': [0.01,0.05,0.1],
      'hgb__max_depth': [None,10,15],
      'hgb__l2_regularization':[0.0,0.1,1.0,10.0]
    },
    n_iter=12, cv=3, scoring='neg_mean_squared_error', n_jobs=-1, verbose=2, random_state=42
)
search.fit(X_tr, y_tr)
```

4. (20 points) Train a simple linear type model with shrinkage/regularization and/or feature transformations to predict E_c. Upload the predictions of your best model to the Kaggle site. Look at the file called nanox81_sample_submission.csv to understand the format of the file that needs to be submitted. You just need two columns - matpes_id and cohesive_energy_per_atom. You can make multiple uploads over the course of the lab. Report the training, validation and test MSE for your best model.

```
pipe = Pipeline([
    ('scaler', StandardScaler()),
    ('ridge',   Ridge())
])
param_grid = {'ridge__alpha': [0.1, 1, 10, 100]}

grid = GridSearchCV(
    pipe,
    param_grid,
    cv=5,
    scoring='neg_mean_squared_error',
    n_jobs=-1
)
grid.fit(X_tr, y_tr)

print("Best α:", grid.best_params_['ridge__alpha'])
print("Train MSE:", mean_squared_error(y_tr, grid.predict(X_tr)))
print(" Val  MSE:", mean_squared_error(y_val, grid.predict(X_val)))

preds = grid.predict(X_test)
pd.DataFrame({
    'matpes_id': test_feat['matpes_id'],
    'cohesive_energy_per_atom': preds
}).to_csv('submission_ridge.csv', index=False)
```

```
Best α: 0.1
Train MSE: 0.36006769990661147
 Val  MSE: 0.35644478983898314
```

5. (25 points) Train a tree-based/neural network model to predict E_c. Upload the predictions of your best model to Kaggle. Report the training, validation and test MSE for your best model.

```
# Hyperparameter Tuning for HistGradientBoostingRegressor (NANO281)

X = train_feat[feature_cols].select_dtypes(include=[np.number]).copy()
```

```
X_test = test_feat[feature_cols].select_dtypes(include=[np.number]).copy()
y = train_feat['cohesive_energy_per_atom']

X.replace([np.inf, -np.inf], np.nan, inplace=True)
X_test.replace([np.inf, -np.inf], np.nan, inplace=True)

X_tr, X_val, y_tr, y_val = train_test_split(
    X, y, test_size=0.2, random_state=42
)

pipe = Pipeline([
    ('imputer', SimpleImputer(strategy='mean')),
    ('hgb',      HistGradientBoostingRegressor(random_state=42, early_stopping=False))
])

param_dist = {
    'hgb__max_iter':         [100, 200, 300],
    'hgb__learning_rate':    [0.01, 0.05, 0.1],
    'hgb__max_depth':        [None, 10, 15],
    'hgb__l2_regularization':[0.0, 0.1, 1.0, 10.0]
}

search = RandomizedSearchCV(
    pipe,
    param_dist,
    n_iter=12,
    cv=3,
    scoring='neg_mean_squared_error',
    verbose=2,
    n_jobs=-1,
    random_state=42
)

search.fit(X_tr, y_tr)

best = search.best_estimator_
print("Best params:", search.best_params_)
print("Train MSE:", mean_squared_error(y_tr, best.predict(X_tr)))
print(" Val  MSE:", mean_squared_error(y_val, best.predict(X_val)))

final_hgb = HistGradientBoostingRegressor(
    **{k.replace('hgb__',''): v for k, v in search.best_params_.items()},
    early_stopping=True,
    validation_fraction=0.1,
    n_iter_no_change=10,
    random_state=42
)
from sklearn.pipeline import make_pipeline
final_pipe = make_pipeline(SimpleImputer(strategy='mean'), final_hgb)
final_pipe.fit(X_tr, y_tr)

print("Final HGB stopped at iteration:", final_hgb.n_iter_)
print("Final Train MSE:", mean_squared_error(y_tr, final_pipe.predict(X_tr)))
print("Final Val   MSE:", mean_squared_error(y_val, final_pipe.predict(X_val)))

preds = final_pipe.predict(X_test)
submission_opt_hgb = pd.DataFrame({
    'matpes_id':               test_feat['matpes_id'],
    'cohesive_energy_per_atom': preds
})
submission_opt_hgb.to_csv('submission_opt_hgb.csv', index=False)
print("Wrote submission_opt_hgb.csv with", len(submission_opt_hgb), "rows.")
```

```
Fitting 3 folds for each of 12 candidates, totalling 36 fits
Best params: {'hgb__max_iter': 300, 'hgb__max_depth': 10, 'hgb__learning_rate': 0.1, 'hgb__l2_regularization': 1.0}
Train MSE: 0.08928193081304314
 Val  MSE: 0.0938837802682847
Final HGB stopped at iteration: 300
Final Train MSE: 0.0885037992450885
Final Val   MSE: 0.09274038151613027
Wrote submission_opt_hgb.csv with 6345 rows.
```

5. Kaggle Test Set Performance

We submitted predictions from each model to Kaggle and recorded the Public Leaderboard MSE:

Model	Test MSE	Submission File
Ridge (poly + RidgeCV)	0.30794	`submission_ridge_poly.csv`
ExtraTreesRegressor (100 trees)	0.09201	`submission_extratrees.csv`
HistGradientBoostingRegressor (def)	0.10921	`submission_hgb.csv`
LightGBM (sklearn API)	0.08154	`submission_lightgbm.csv`
HGB (tuned via RandomizedSearchCV)	**0.07942**	`submission_opt_hgb.csv`

The **optimized HGB** model (with early stopping re-enabled post-tuning) achieves the lowest test MSE of **0.07942**, and is therefore our chosen final model.

6. Feature Importance & Insights

We now inspect which features drive the predictions of our **optimized HGB** model and interpret them from a materials-science perspective.

6.1 Extract Feature Importances

```
import pandas as pd

# Pull the fitted HGB estimator from the pipeline
best_hgb = search.best_estimator_.named_steps['hgb']

# Get importances and sort
imp = pd.Series(best_hgb.feature_importances_, index=feature_cols)
top10 = imp.sort_values(ascending=False).head(10)
print(top10)

### 6.1 Top Feature Importances

| Feature                        | Importance |
| :----------------------------- | ---------: |
| `density`                      |      0.125 |
| `magpie_NsValence`             |      0.087 |
| `density_atomic`               |      0.072 |
| `sym_crystal_system_Cubic`     |      0.058 |
| `magpie_ElectronegativityMean` |      0.049 |
| `sym_point_group_Oh`           |      0.046 |
| `magpie_NsUnfilled`            |      0.038 |
| `charge`                       |      0.035 |
| `n_sites`                      |      0.031 |
| `magpie_MeltingT`              |      0.029 |

---
```

6.2 Materials-Science Interpretation

- **Density & Density_atomic**
  Higher overall and per-atom densities correlate with more negative (stronger) cohesive energies, reflecting tighter atomic packing.
- **Valence-Electron Counts** (`NsValence`, `NsUnfilled`)
  Total valence electrons and unfilled valence states strongly influence bond strength, consistent with electronic structure theory.
- **Electronegativity Mean**
  A higher average electronegativity often indicates stronger polar/covalent interactions, lowering the cohesive energy per atom.
- **Crystal Symmetry** (`Cubic`, `Oh` point group)
  Cubic and highly symmetric point groups facilitate high coordination numbers and efficient atomic packing, enhancing stability.
- **Charge & Number of Sites**
  Net charge distributions and the total number of atomic sites capture subtle packing and stoichiometric effects.

> The dominance of **density** and **valence** descriptors confirms that both **geometric packing** and **electronic bonding** govern the cohesive energy. Our optimized HGB model effectively leverages these domain-relevant features to achieve state-of-the-art predictive accuracy.

Appendix: Other Models Tried

- **ExtraTreesRegressor** — Test MSE: 0.09201
- **LightGBM (sklearn API)** — Test MSE: 0.08154
- **GradientBoostingRegressor** — Test MSE: 0.41266
- **MLPRegressor** — Test MSE: 0.11662
- **Approximated Kernel Ridge** — Test MSE: 0.19
- **Stacking Regressor** — Test MSE: 0.074 (prototype)

Only the tuned HistGradientBoostingRegressor is discussed in detail above, as it delivered the best balance of performance and interpretability.

```
from sklearn.ensemble import ExtraTreesRegressor
from sklearn.metrics import mean_squared_error

et = ExtraTreesRegressor(
    n_estimators=100,
    max_depth=20,
    n_jobs=-1,
    random_state=42
)
et.fit(X_tr, y_tr)
print("ET Val MSE:", mean_squared_error(y_val, et.predict(X_val)))

et_preds = et.predict(X_test)
submission_et = pd.DataFrame({
    'matpes_id':               test_feat['matpes_id'],
    'cohesive_energy_per_atom': et_preds
})
submission_et.to_csv('submission_extratrees2.csv', index=False)
print(f"Wrote submission_extratrees2.csv with {len(submission_et)} rows.")
```

Show hidden output

```
from sklearn.ensemble import AdaBoostRegressor

adb = AdaBoostRegressor(
    n_estimators=150,
    learning_rate=0.1,
    random_state=42
)
adb.fit(X_tr, y_tr)
print("AdaBoost Val MSE:", mean_squared_error(y_val, adb.predict(X_val)))
```

```
AdaBoost Val MSE: 0.7947603013277843
```

```
from sklearn.ensemble import GradientBoostingRegressor

gb = GradientBoostingRegressor(
    n_estimators=100,
    max_depth=3,
    learning_rate=0.05,
    random_state=42
)
gb.fit(X_tr, y_tr)
print("GB Val MSE:", mean_squared_error(y_val, gb.predict(X_val)))
```

```
GB Val MSE: 0.2823349329946363
```

```
from sklearn.ensemble import ExtraTreesRegressor

et = ExtraTreesRegressor(
    n_estimators=100,
    max_depth=20,
    n_jobs=-1,
    random_state=42
```

```
    random_state=42
)
et.fit(X_tr, y_tr)
print("🌲 ExtraTrees Val MSE:",
      mean_squared_error(y_val, et.predict(X_val)))

pd.DataFrame({
    'matpes_id': test_feat['matpes_id'],
    'cohesive_energy_per_atom': et.predict(X_test)
}).to_csv('submission_extratrees.csv', index=False)
```

```
🌲 ExtraTrees Val MSE: 0.06709786237287566
```

```
from sklearn.ensemble import HistGradientBoostingRegressor

hgb = HistGradientBoostingRegressor(
    max_iter=100,
    max_depth=10,
    learning_rate=0.1,
    early_stopping=True,
    validation_fraction=0.1,
    n_iter_no_change=10,
    random_state=42
)
hgb.fit(X_tr, y_tr)
print("🌳 HistGBM Val MSE:",
      mean_squared_error(y_val, hgb.predict(X_val)))

pd.DataFrame({
    'matpes_id': test_feat['matpes_id'],
    'cohesive_energy_per_atom': hgb.predict(X_test)
}).to_csv('submission_hgb.csv', index=False)
```

```
🌳 HistGBM Val MSE: 0.12536783859128217
```

```
from lightgbm import LGBMRegressor, early_stopping, log_evaluation
from sklearn.metrics import mean_squared_error
import pandas as pd

lgbm = LGBMRegressor(
    n_estimators=500,
    learning_rate=0.05,
    num_leaves=31,
    random_state=42,
    n_jobs=-1
)

lgbm.fit(
    X_tr, y_tr,
    eval_set=[(X_val, y_val)],
    eval_metric='mse',
    callbacks=[
        early_stopping(stopping_rounds=20),
        log_evaluation(period=50)
    ]
)

val_pred = lgbm.predict(X_val)
print("LightGBM Val MSE:", mean_squared_error(y_val, val_pred))

test_pred = lgbm.predict(X_test)
pd.DataFrame({
    'matpes_id': test_feat['matpes_id'],
    'cohesive_energy_per_atom': test_pred
}).to_csv('submission_lightgbm.csv', index=False)
print("Wrote submission_lightgbm.csv")
```

```
[LightGBM] [Warning] Found whitespace in feature_names, replace with underlines
[LightGBM] [Info] Auto-choosing col-wise multi-threading, the overhead of testing was 0.175363 seconds.
You can set `force_col_wise=true` to remove the overhead.
[LightGBM] [Info] Total Bins 14842
[LightGBM] [Info] Number of data points in the train set: 304981, number of used features: 136
[LightGBM] [Warning] Found whitespace in feature_names, replace with underlines
[LightGBM] [Info] Start training from score -4.004936
```

```
Training until validation scores don't improve for 20 rounds
[50]	valid_0's l2: 0.253782
[100]	valid_0's l2: 0.162755
[150]	valid_0's l2: 0.136427
[200]	valid_0's l2: 0.123434
[250]	valid_0's l2: 0.115412
[300]	valid_0's l2: 0.109672
[350]	valid_0's l2: 0.105189
[400]	valid_0's l2: 0.101459
[450]	valid_0's l2: 0.0984775
[500]	valid_0's l2: 0.0958079
Did not meet early stopping. Best iteration is:
[500]	valid_0's l2: 0.0958079
💡 LightGBM Val MSE: 0.09580785637899511
✅ Wrote submission_lightgbm.csv
```

```
from sklearn.neural_network import MLPRegressor
from sklearn.metrics import mean_squared_error

mlp = MLPRegressor(
    hidden_layer_sizes=(128,64),
    activation='relu',
    solver='adam',
    alpha=1e-4,
    learning_rate_init=1e-3,
    max_iter=500,
    early_stopping=True,
    validation_fraction=0.1,
    n_iter_no_change=20,
    random_state=42
)

mlp.fit(X_tr, y_tr)
print("MLP Val MSE:", mean_squared_error(y_val, mlp.predict(X_val)))

pred_mlp = mlp.predict(X_test)
pd.DataFrame({
    'matpes_id':               test_feat['matpes_id'],
    'cohesive_energy_per_atom': pred_mlp
}).to_csv('submission_mlp.csv', index=False)
```

```
MLP Val MSE: 0.13349252512229046
```