Computational Search for New Quaternary Heusler Compounds, PSED Cluster 2015-2016

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RESEARCH OBJECTIVE

The objective of this PSED project is to develop a model combined with **Machine Learning (ML) algorithm and Density Functional Theory (DFT) calculation to discover new potential quaternary Heusler compounds (QHs)** among ~3,000,000 QHs by combinatorially-substituting elements.

- 73(elements)C_4x3 = ~3,000,000 QHs

Open Quantum Material Database (OQMD) → Develop ML model → Validate ML model → Perform prediction for searching new QHs. →

- Validate ML predicted stable phases by DFT
- Find best candidates among ~3,000,000 QHs

Search new stable quaternary Heuslers (QHs).

Rank	Attributes (#274)
1 2 3	mean., var., min. of effective coordination
4 5	max. number of d election and Mandeleev
6	range (max.-min.) of atomic weight
7	min. of bond length var.

Content	Number of compounds
ML prediction (Hull distance < 0.1 eV/atom)	827
In OQMD / stable phases in OQMD	762 / 204
Not in OQMD	65

- Important: Local chemistry (#valence electrons, atomic volume)
- Prediction (ML): 65 new QHs with potentially stable
- Need to perform DFT calculations for 65 QHs.
- Currently, we found **new stable phase (LiAlZnAg),** which is not included in the training data set (OQMD).

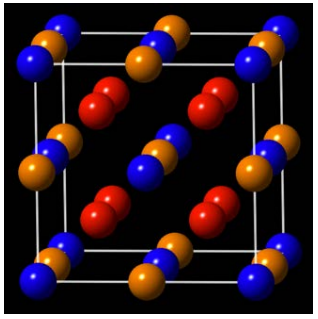
Development of Machine Learning (ML) model

#	Data set	Data type (eV/atom)	Total numbers	10CV, MAE (eV/atom)
A	Quaternary Heuslers	Formation energy	~90,000	0.0455
B	Ternary Heuslers	Formation energy	~180,000	0.0672
C	Ternary / 0.9 Quaternary	Formation energy	~260,000	0.0619
D	Quaternary Heuslers	Hull distance	~90,000	0.0442

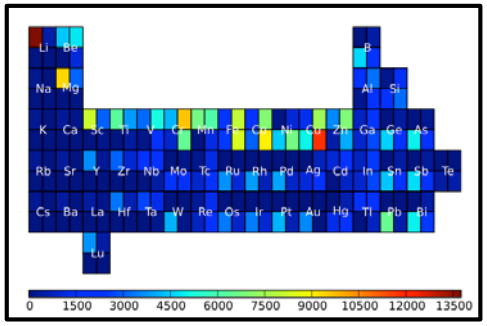
Algorithm	10CV, MAE (eV/atom)
REPTree	0.0673
Decision Stump	0.178
Random Forest	0.0442

- Best performance
  - Training set: QHs
  - Algorithm: Random Forest

Challenges and Difficulties



Chemical formula of Heusler compound: X_2YZ



The frequency of chosen elements in QH training data sets on four sites (X_2YZ)

- Dataset is biased towards compounds with 3d transition metals (conventional alloying elements) with Li.
- Issue: These are not representative of the entire search space.

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