Developing a sequential learning methodology in order to discover novel anti-perovskite and garnet-structured materials

Predictive Science & Engineering Design Project (2019-2020)

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RESEARCH GOALS

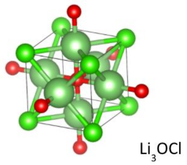
1. Discovery of new, stable antiperovskite structures
2. Discovery of new, stable garnet structures
3. Development of a scalable, reproducible computational framework that **accelerates the discovery of new materials**

Motivation

There aren't many known compounds with antiperovskite or garnet crystal structures. However, these two structure types have shown much promise of exceptional functionalities in recent research such as superionic conductivity, superconductivity, giant magnetoresistance, and negative thermal expansion.

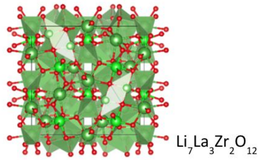
Key challenges:

1. **Few known compounds** within a structure-type
2. **Large search space**: millions of prototype compounds
3. **Computationally expensive** *ab initio* calculations



Li_3OCl

Anti-perovskites

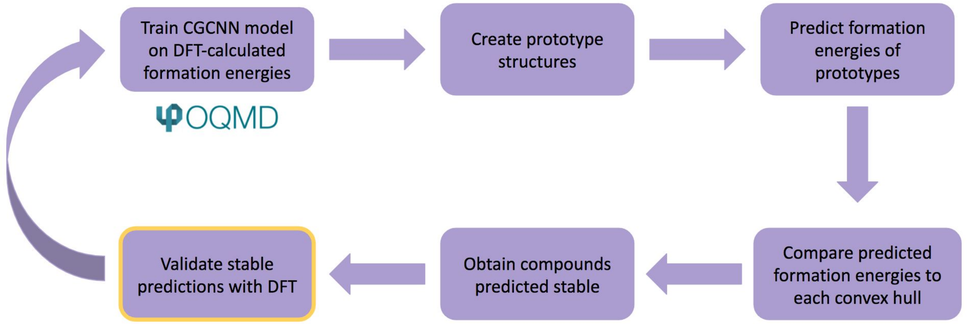


$Li_7La_3Zr_2O_{12}$

Garnets

METHOD OF RESEARCH

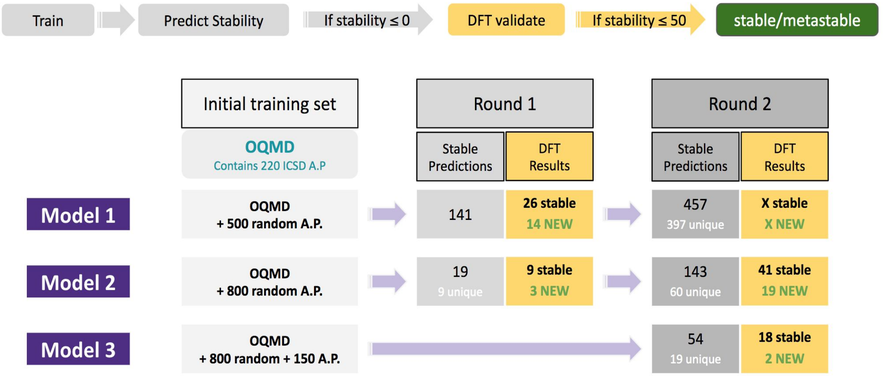
A sequential learning framework that combines high-throughput density functional theory with machine learning in a cycle of training, predicting, validating and retraining until convergence.



Tools used:

Open Quantum Materials Database (OQMD), **Crystal Graph Convolutional Neural Network** (CGCNN), **Vienna Ab Initio Simulation Package** (VASP)

ANTIPEROVSKITE RESULTS



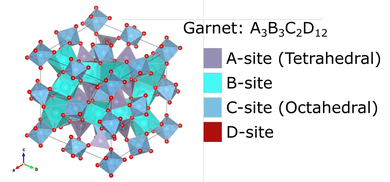
35 NEW compounds discovered

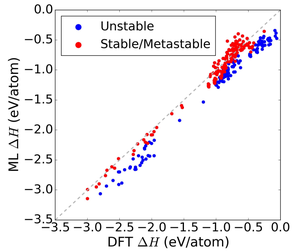
- 14 stable
- 21 metastable

Key Takeaways:

- CGCNN model has low MAE
- CGCNN depends on having structure-type in training set for higher accuracy.
- Multiple rounds of training and predicting are **vital**
- Multiple models trained on varying amounts of data are **vital**

GARNET RESULTS





Compound ($A_3B_3C_2D_{12}$)	**Stability** meV/atom	**Band Gap Energy** eV	**Competing Stable Phases in the OQMD**
$Mo_3Ca_3W_2N_{12}$	-62	1.0	$MoN + CaN_2 + W_3Ca_3Mo_2N_{12} + MoN_2$
$W_3Ca_3Mo_2N_{12}$	-50	0.8	$Mo_3Ca_3W_2N_{12} + Ca_3N_2 + W_3N_5 + CaN_2$
$Li_3Na_3Tl_2F_{12}$	-38	3.7	$LiF + NaF + TlF_3$
$Li_3Na_3In_2F_{12}$	-29	5.7	$LiInF_4 + LiF + Na_3InF_6$
$Nb_3Y_3W_2N_{12}$	-25	1.4	$YN + Nb_5N_6 + W_3N_5 + N$
$Ta_3Y_3W_2N_{12}$	-18	1.4	$N + W_3N_5 + Ta_3N_5 + YN$

153 NEW compounds discovered

- 110 stable
- 43 metastable

Key Takeaways:

- ML vs DFT Mean absolute error(MAE): **157** meV/atom
- ML success rate of **31%** very high (previous HT-study success rate ~10%)
- We recommend **6** compounds to be interesting and worth exploring more computationally and experimentally