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Faculty Advisors: Drs. Ankit Agrawal (EECS), Chris Wolverton (MSE), Peter Voorhees (MSE)

**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\text{(elements)}C_4\times3 = 3,000,000 \text{ QHs}**

![Diagram of Open Quantum Material Database (OQMD) Process]

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).**

<table>
<thead>
<tr>
<th>Rank</th>
<th>Attributes (#274)</th>
<th>Content</th>
<th>Number of compounds</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>mean., var., min. of effective coordination</td>
<td>ML prediction (Hull distance &lt; 0.1 eV/atom)</td>
<td>827</td>
</tr>
<tr>
<td>2</td>
<td>max. number of d electron and Mandeleev</td>
<td>In OQMD / stable phases in OQMD</td>
<td>762 / 204</td>
</tr>
<tr>
<td>3</td>
<td>range (max.-min.) of atomic weight</td>
<td>Not in OQMD</td>
<td>65</td>
</tr>
</tbody>
</table>

- 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**

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

**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**

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

**Chemical formula of Heusler compound: X_2YZ**

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