The Materials Genome Project: Materials Design with High-Throughput Ab Initio Computing

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**Materials science takes a long time to become materials technology**


- Teflon
- Velcro
- Titanium
- Polycarbonate
- GaAs
- Diamond-like Thin Films
- Amorphous soft magnets

Invented in lab → Li ion battery → Commercialized

First IBM PC → Google in 10 languages

*Materials Data from: Eagar, T.; King, M. Technology Review 1995*
The Materials Design problem...

**What delays materials advancements?**

**Lack of information.**
- properties of existing materials
- new materials prediction
- simultaneous property optimization

Often, ‘experience’ is the only guide.
Better data is needed for materials design.
Density Functional Theory provides materials data at the speed of computation

\[ i\hbar \frac{d\Psi(\{r_i\}; t)}{dt} = \hat{H} \Psi(\{r_i\}; t) \]

\[ H = \sum_{i=1}^{N_e} \nabla_i^2 + \sum_{i=1}^{N_e} V_{\text{nuclear}}(r_i) + \sum_{i=1}^{N_e} V_{\text{effective}}(r_i) \]

Total energy
Optimized structure
Magnetic ground state
Charge density
Band structure / DOS
Scaling to high-throughput DFT

GridEngine queue management

Computational resources
- Perl scripts:
  - identify converged runs
  - rerun failed jobs using modified convergence parameters
  - kill jobs which are not converging

AFLOW wrapper

VASP job

converged runs

intuition-guided selection of compounds

crystal structure prediction

Java backend

XML output
- parse into atomic data fragments

Postgresql Database
- ICSD data
- VASP data
- Analysis data
  - phase diagrams
  - structural features
  - application-specific data
    (e.g. battery voltage)

Java backend / frontend
- explore/analyze data
- generate VASP jobs

researcher knowledge
1. High-throughput DFT applied to Li ion-battery cathodes

2. The Materials Genome at MIT: a resource for materials informatics
Cathode is now the major barrier for better energy density, high rate, lower cost, safety

(Significant improvements needed in all areas...)
Voltage is one design criteria for batteries

- Voltage window between 3-4.5V

\[ V_{OC} = \frac{\mu_{anode} - \mu_{cathode}}{F} \]

integrate

\[ V_{avg} = \frac{\Delta G}{F\Delta x_{Li}} \]

\[ \Delta G \sim [ E(\text{Li Mn O2}) - + E(\text{MnO2}) ] \]

E (Li Mn O2) - [ E (MnO2) + E (Li) ]
How well can DFT predict voltage?

![Bar chart showing computed and experimental literature voltage results for various materials.](chart.png)
Li ion cathodes can be computationally optimized over multiple properties

- Safety is another largely computable property

Others include:
- thermodynamic stability
- intercalation
- volume change
- intrinsic rate capability

Optimize over multiple properties before time-consuming lab work

Ong, S. P., Jain, A., Hautier, G., Kang, B., & Ceder, G. 
*Electrochemistry Communications* (2010)
New materials for batteries discovered using high-throughput DFT

<table>
<thead>
<tr>
<th>Chemistry</th>
<th>Novelty</th>
<th>Potential for energy density improvement over LiFePO$_4$</th>
<th>Percent of experimental capacity already achieved in the lab</th>
</tr>
</thead>
<tbody>
<tr>
<td>Borate-related</td>
<td>Compound known (new electrochem.)</td>
<td>50% greater</td>
<td>~45%</td>
</tr>
<tr>
<td>Li$_3$ M (PO$_4$) (CO$_3$)</td>
<td>New (never reported)</td>
<td>40% greater</td>
<td>~45%</td>
</tr>
<tr>
<td>M=Fe, Mn, Co, ...</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Phosphate-related</td>
<td>New (never reported)</td>
<td>20% greater</td>
<td>~60%</td>
</tr>
</tbody>
</table>

![Graph showing voltage (V) vs. capacity (mAh/g)](image)
Data leads to design intuition

Voltage (V)

Eq. Oxygen Release $\mu O_2^-$
Charged State (eV)

more safe

less safe

Equ. Oxygen Release Temp., Charged State (eV)

oxides, phosphates, silicates, borates, sulfates
Remarks on Li ion battery cathode design

- Used fairly limited computational resources (~300 cores)
- Used fairly small experimental team (~3 full-time, 3 part-time members)

- Able to screen **20,000 materials** and experimentally synthesize **3 promising new cathode materials** for Li ion batteries
Outline

1. High-throughput DFT applied to Li ion-battery cathodes

2. The Materials Genome at MIT: a resource for materials informatics
Goals of the Materials Genome

- Provide a **public database** of electronic structure calculations for:

  Materials screening & design

  Structure Prediction
Goals of the Materials Genome

- Provide a **public database** of electronic structure calculations for:

Analysis

Data mining
Computations predict best performing metal sorbents

Pd shows best Hg adsorption in experiments

Pt shows second best Hg adsorption in experiments

Structure prediction methodology

**Step 1: Predict**
(Using data-mined correlations)

“Knowledge database”

**Step 2: Confirm**
(Using DFT computations)

Fischer, C. C., Tibbetts, K. J., Morgan, D., & Ceder, G.

Hautier, G., Fischer, C., Jain, A., Mueller, T., Ceder, G.
Chemistry of Materials (2010)

\[
\Psi^i \frac{d\Psi^i(r_i; t)}{dt} = \hat{H} \Psi^i(r_i; t)
\]

\[
H = \sum_{i=1}^{N_e} \nabla_i^2 + \sum_{i=1}^{N_e} V_{\text{nuclear}}(r_i) + \sum_{i=1}^{N_e} V_{\text{effective}}(r_i)
\]
New ternary oxides predicted

- Over 200 new ternary oxide compounds were predicted by our method.
- All predictions & oxidation range for synthesis in supplemental info of [1].

Can generate computed 0K, 0atm phase diagrams for almost all chemical systems.
Phase diagrams lead to ultrafast battery cathode

Full charge/discharge in 15 minutes

Full charge/discharge in 75 seconds

Kang, B., & Ceder, G.
Nature (2009)

Ong, S.P., Wang, L., Kang, B., & Ceder, G.
Chemistry of Materials (2008)
The Materials Genome: A tool for YOU to design materials in record-breaking time!

Materials Genome prototype web interface
Expected launch 2011
Thank you & Acknowledgements

- DFT automation & theory team
  - Chris Fischer
  - Shyue Ping Ong
  - Tim Mueller
  - Kristin Persson (LBNL)
  - Maria Chan (Argonne)

- Experimental Team
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  - Robert Doe
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  - Jae Chul Kim
  - Xiaofei Fang

- Ceder group

- Funding
  - Umicore
  - Bosch
  - DOE CSGF
BACKUP SLIDES
The Materials Genome: A Tool for Advancing Materials Informatics
Designing Stable Structures: Local Environment Statistics

Fe$^{2+}$ octahedral

Cu$^{2+}$ square planar

Relative frequency
K-point convergence

Energy difference distribution, 'default' and 'increased k-point' parameters

<table>
<thead>
<tr>
<th>Energy Difference Range</th>
<th>Number of Compounds</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5 - 5</td>
<td>(155)</td>
</tr>
<tr>
<td>5.0 - 10</td>
<td>(9)</td>
</tr>
<tr>
<td>10.0 - 15</td>
<td>(7)</td>
</tr>
<tr>
<td>15.0 - 20</td>
<td>(3)</td>
</tr>
<tr>
<td>20.0 - 25</td>
<td></td>
</tr>
<tr>
<td>25.0 - 30</td>
<td></td>
</tr>
<tr>
<td>30.0 - 35</td>
<td></td>
</tr>
<tr>
<td>35.0 - 40</td>
<td></td>
</tr>
<tr>
<td>40.0 - 45</td>
<td>(1)</td>
</tr>
</tbody>
</table>
Formation enthalpy accuracy

Energy difference distribution, 'default' vs. 'increased k-point' parameters

Number of compounds:
- 0 - 50: 49
- 50 - 100: 33
- 100 - 150: 29
- 150 - 200: 25
- 200 - 250: 12
- 250 - 300: 7
- 300 - 350: 7
- 350 - 400: 1
- 400 - 450: 3
- 450 - 500: 2

Absolute energy difference in formation enthalpy, meV/atom
IGCC (Integrated Gas Combustion Cycle) – efficient alternative to pulverized coal (PC) power

One Problem: hot syngas needs to be cooled to remove impurities, then reheated

3.6% estimated thermal efficiency loss can be regained if impurity sorption can occur above 440K

Nexant Inc., Environmental footprints and costs of coal-based integrated gasification combined cycle and pulverized coal technologies. 2006, Environmental Protection Agency.

Goal: find a metal capable of removing 100 ppbw Hg gas from a syngas stream at high T

Strategy: compute all formation enthalpies:

for all known binary Metal-Hg amalgams (along with oxidation energies)
Data mining
Substitution Probabilities

Hautier, G., Fischer, C., Ehlracher, V., Ceder, G.
(manuscript in preparation)