

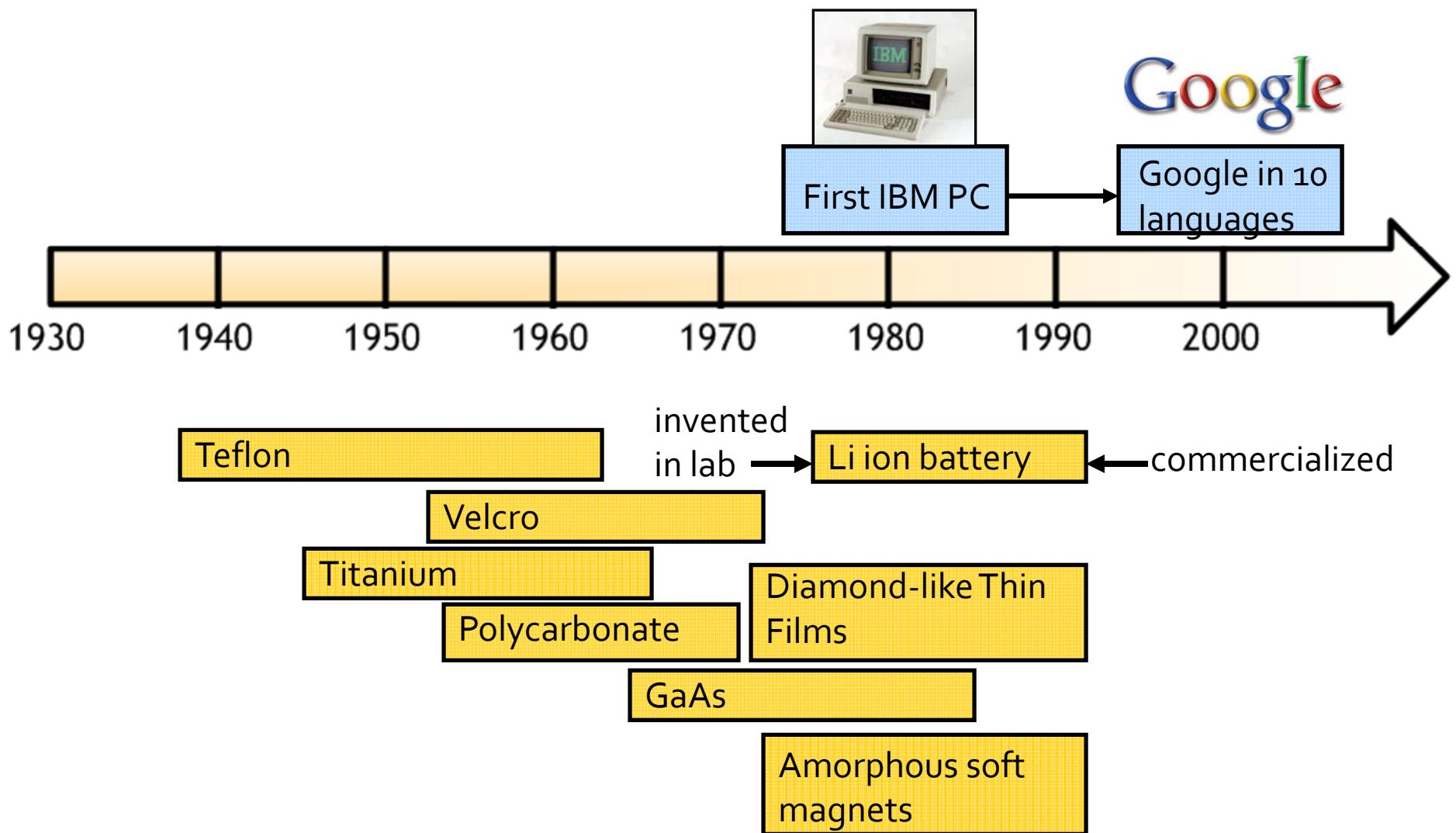
The Materials Genome Project: Materials Design with High-Throughput Ab Initio Computing

SciDAC 7/11/2010

Anubhav Jain
Geoffroy Hautier
Charles Moore
Gerbrand Ceder

*Massachusetts Institute of Technology
Cambridge, MA*

Materials science takes a long time to become materials technology



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

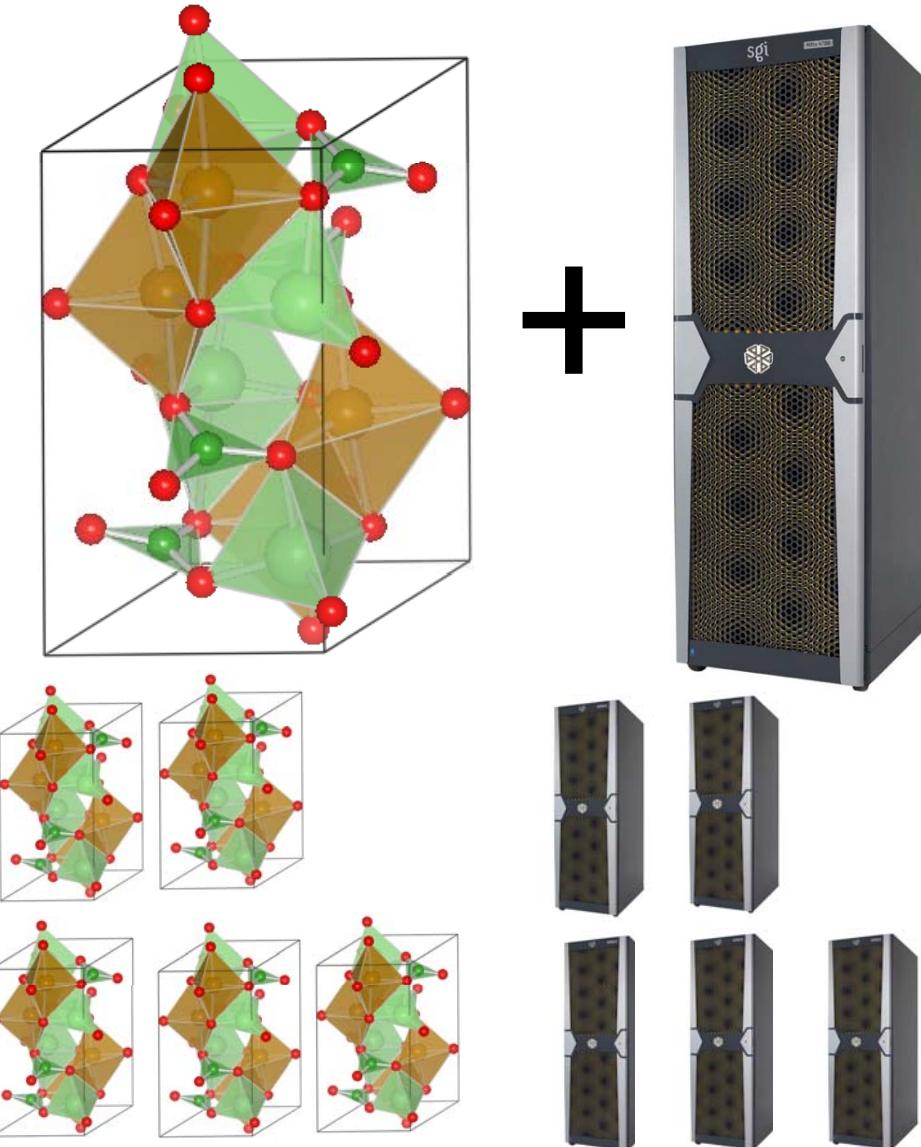
new materials prediction

simultaneous property optimization

Often, 'experience' is the only guide.

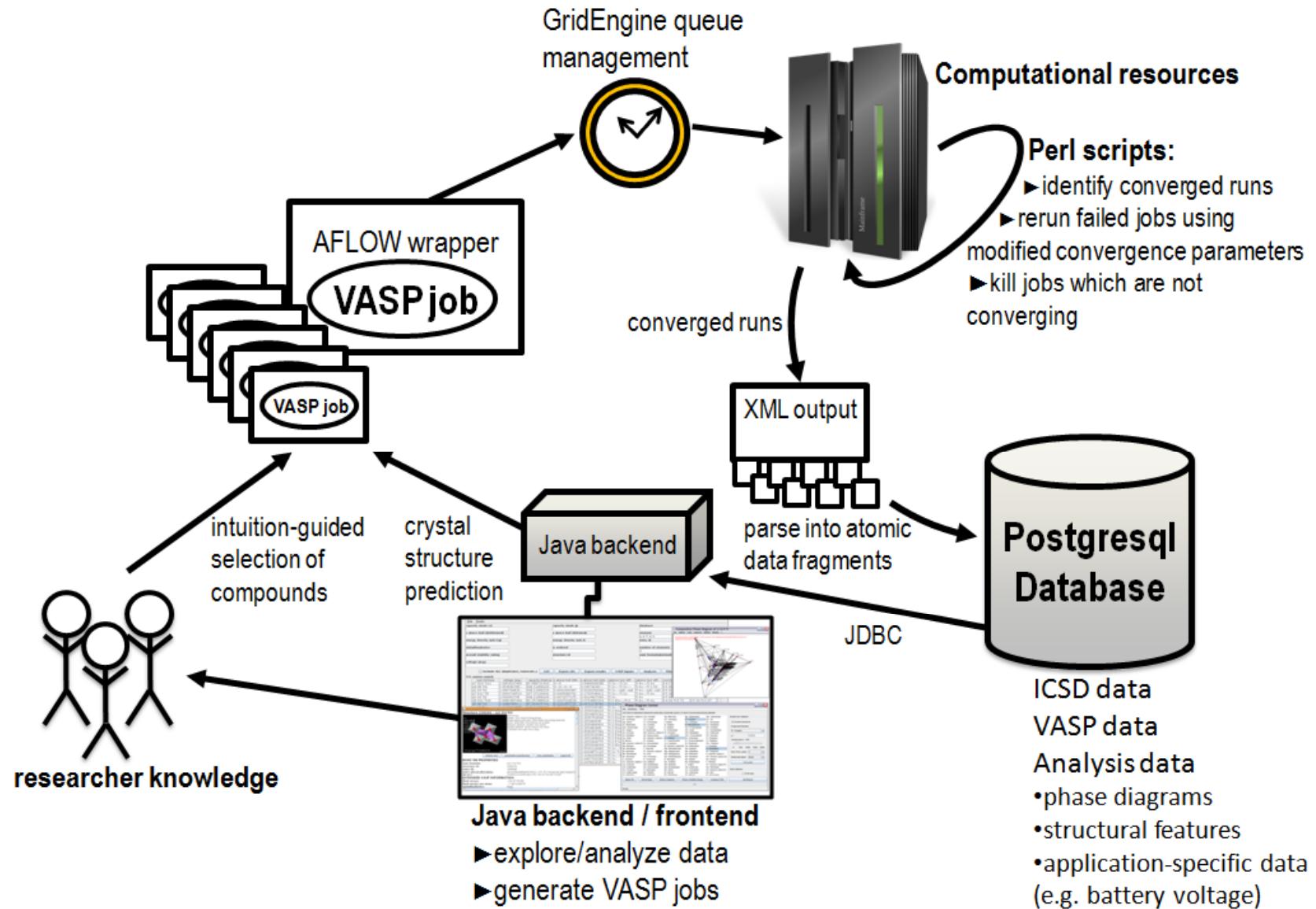
Better data is needed for materials design.

Density Functional Theory provides materials data at the speed of computation



Total energy
Optimized structure
Magnetic ground state
Charge density
Band structure / DOS

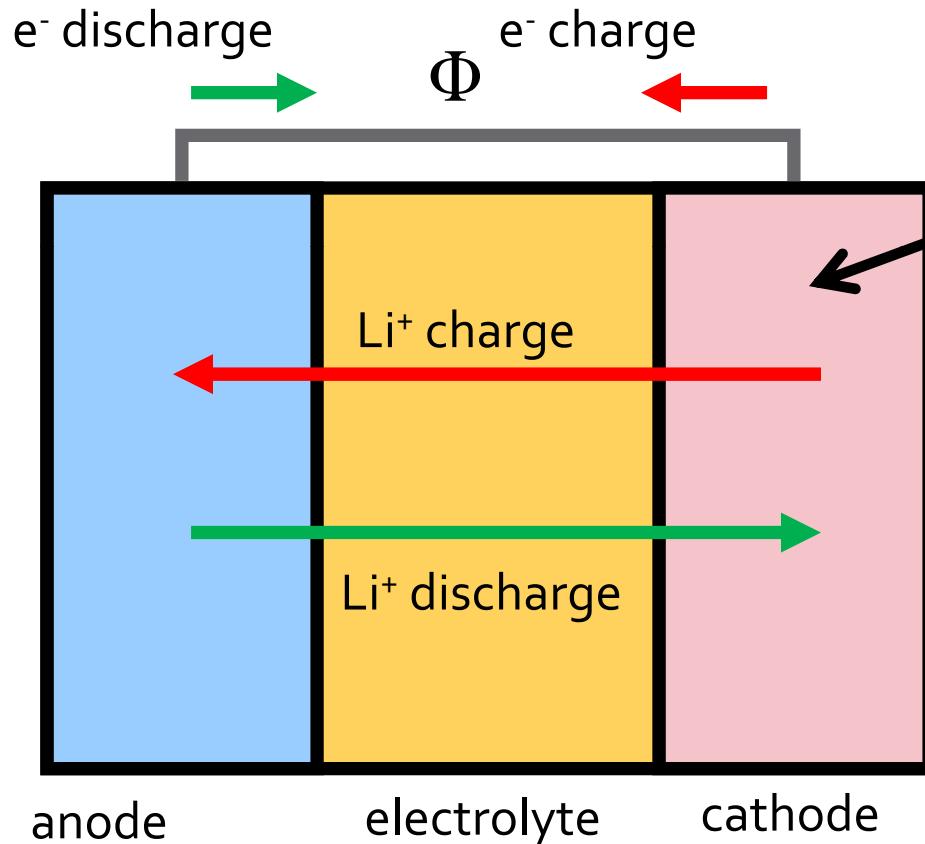
Scaling to high-throughput DFT



Outline

1. High-throughput DFT applied to Li ion-battery cathodes
2. The Materials Genome at MIT:
a resource for materials informatics

Lithium Ion Batteries



Cathode is now
the major barrier
for better energy
density, high rate,
lower cost, safety

(Significant
improvements
needed in all
areas...)

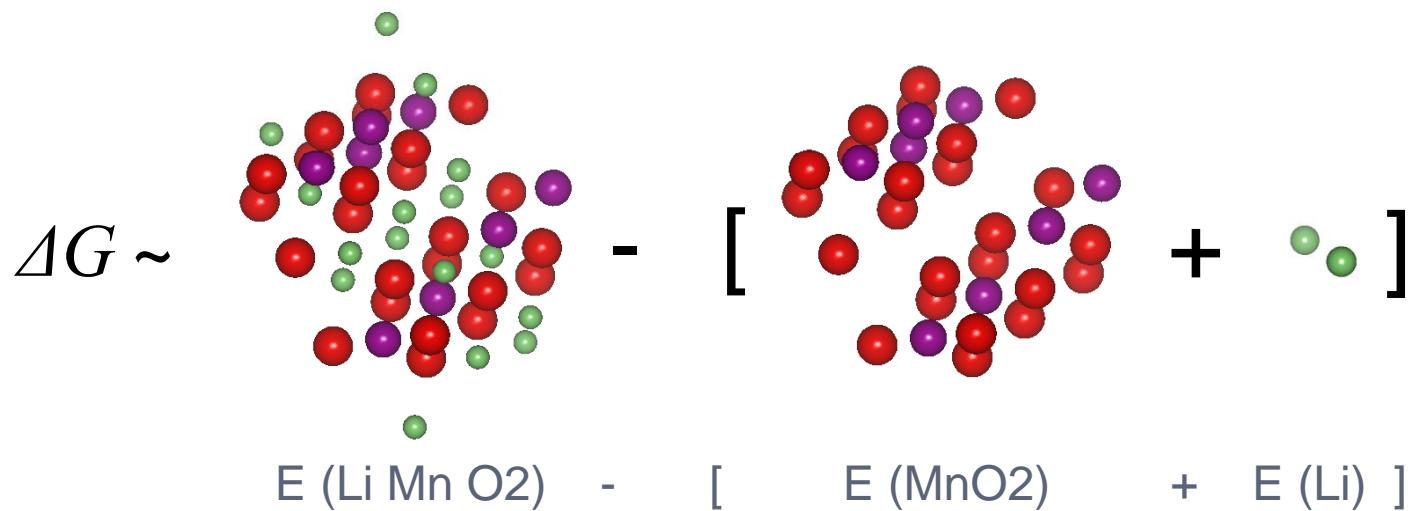
Voltage is one design criteria for batteries

- Voltage window between 3-4.5V

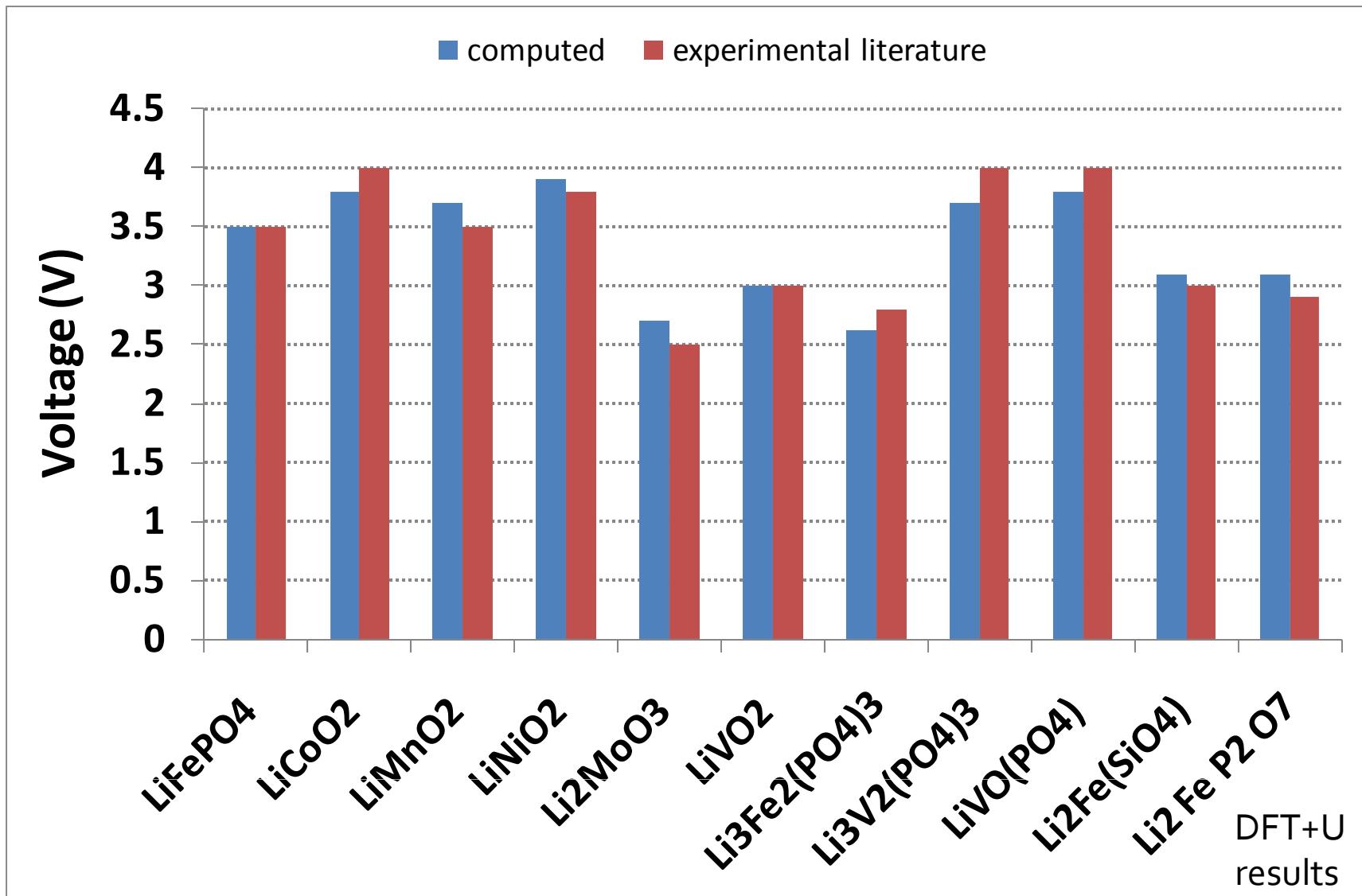
$$V_{OC} = \frac{\mu_{Li}^{anode} - \mu_{Li}^{cathode}}{F}$$

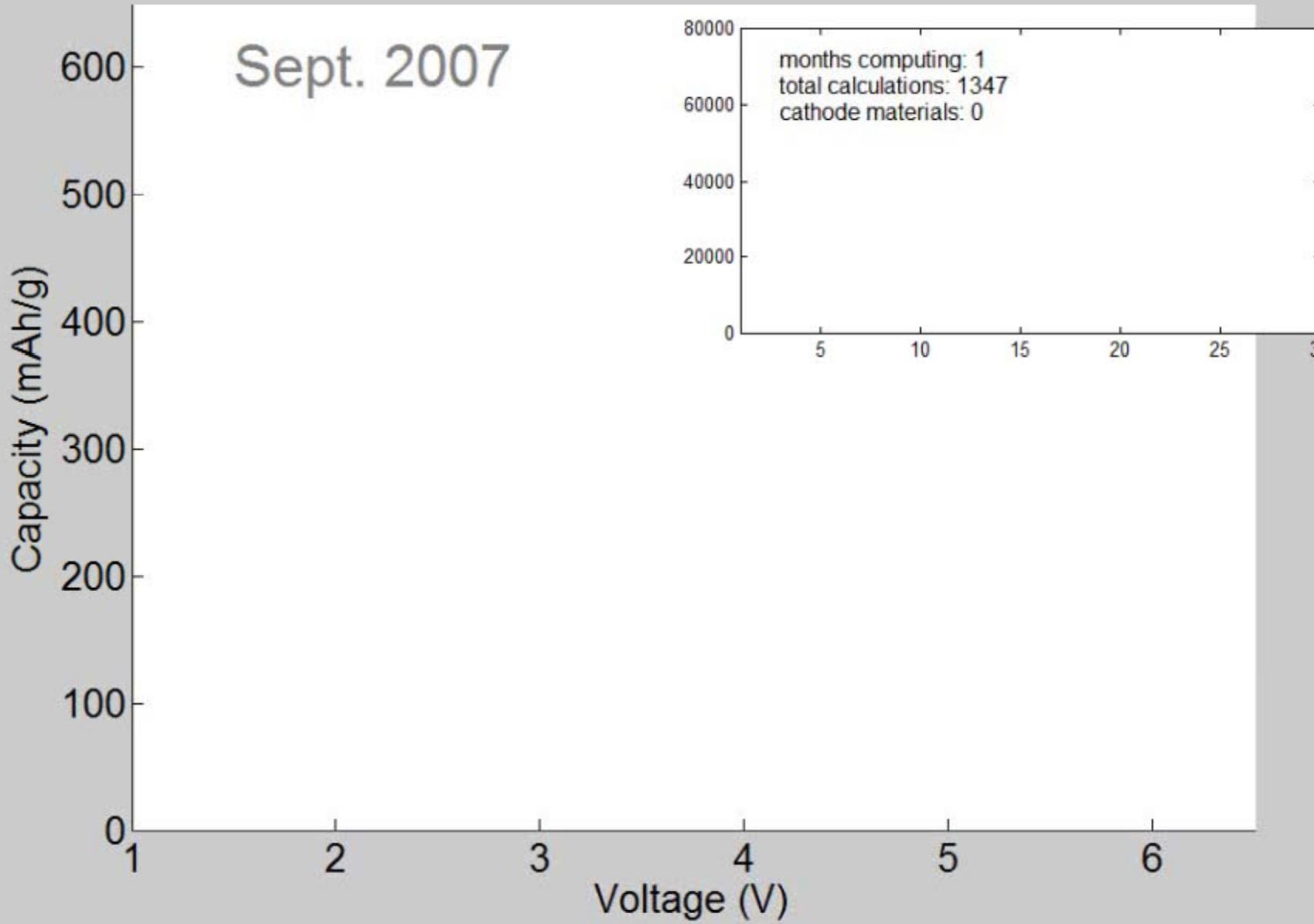
integrate 

$$V_{OC}^{avg} = \frac{\Delta G}{F\Delta x_{Li}}$$



How well can DFT predict voltage?

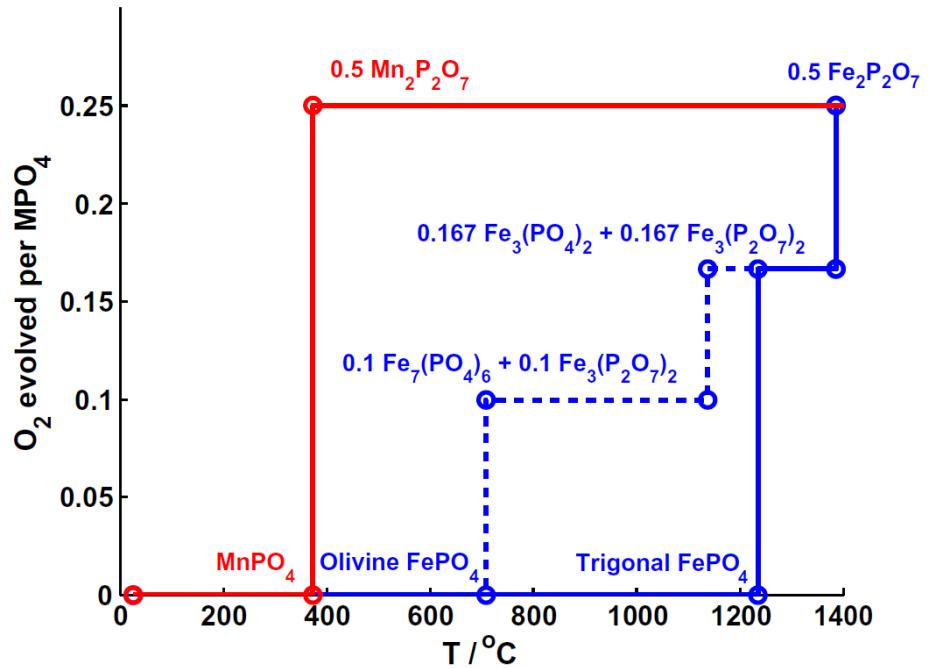




Li ion cathodes can be computationally optimized over multiple properties

- Safety is another largely computable property

Others include:
thermodynamic stability
intercalation
volume change
intrinsic rate capability

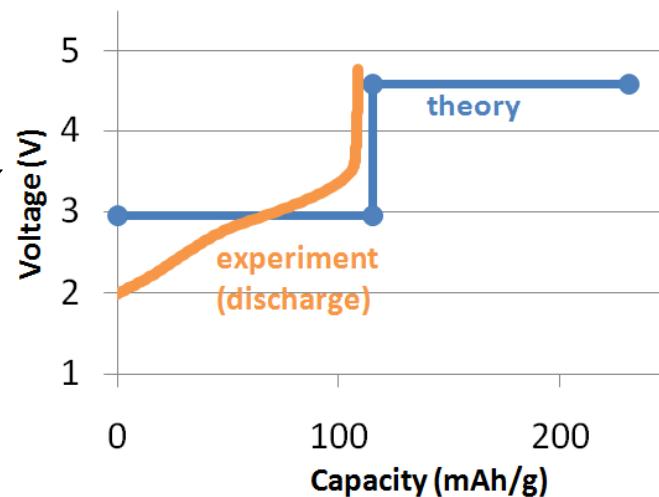


Ong, S. P., Jain, A., Hautier, G., Kang, B., & Ceder, G.
Electrochemistry Communications (2010)

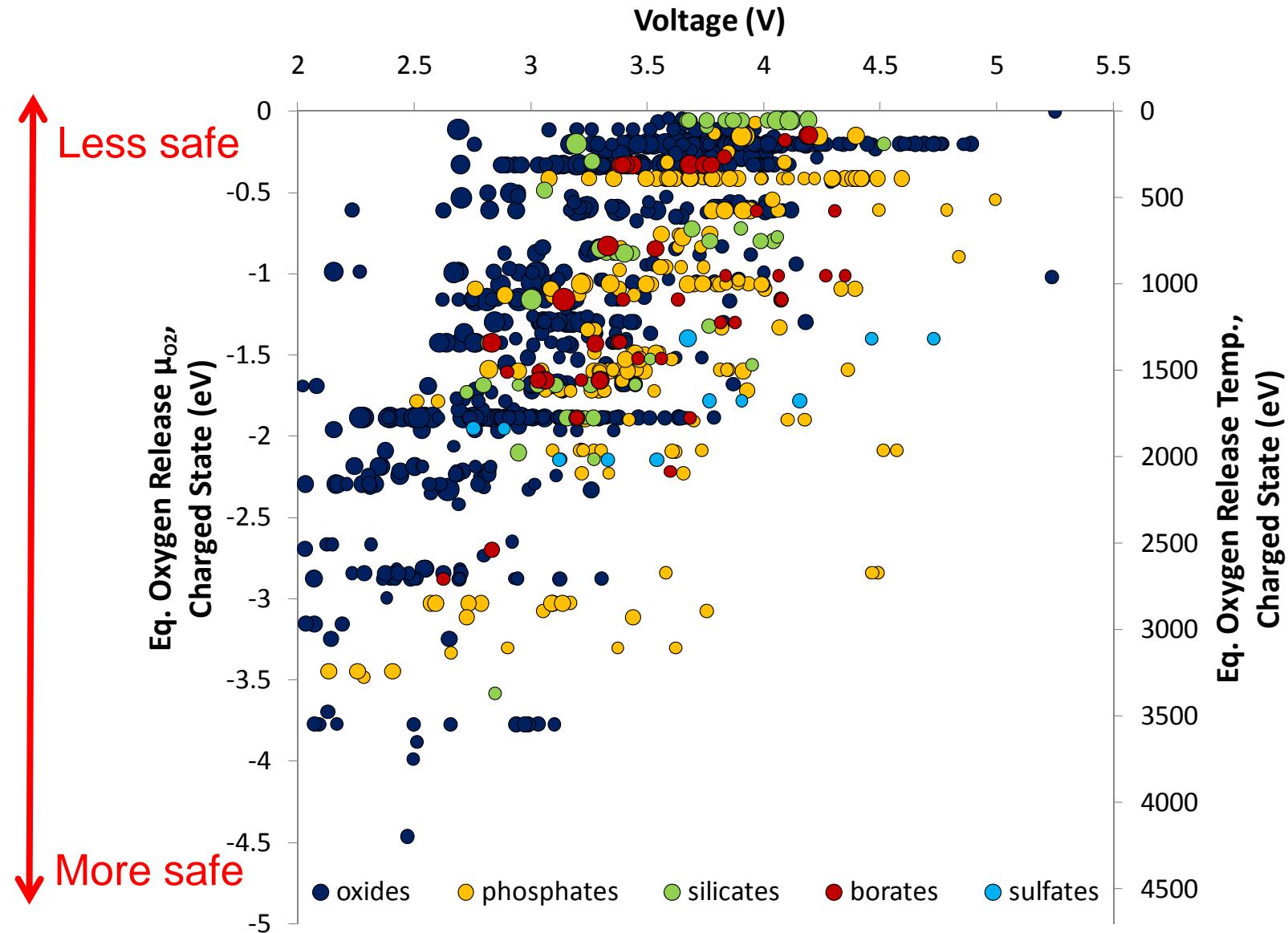
Optimize over multiple properties before time-consuming lab work

New materials for batteries discovered using high-throughput DFT

Chemistry	Novelty	Potential for energy density improvement over LiFePO ₄	Percent of experimental capacity already achieved in the lab
Borate-related	Compound known (new electrochem.)	50% greater	~45%
Li ₃ M (PO ₄) (CO ₃) M=Fe, Mn, Co, ...	New (never reported)	40% greater	~45%
Phosphate-related	New (never reported)	20% greater	~60%



Data leads to design intuition



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

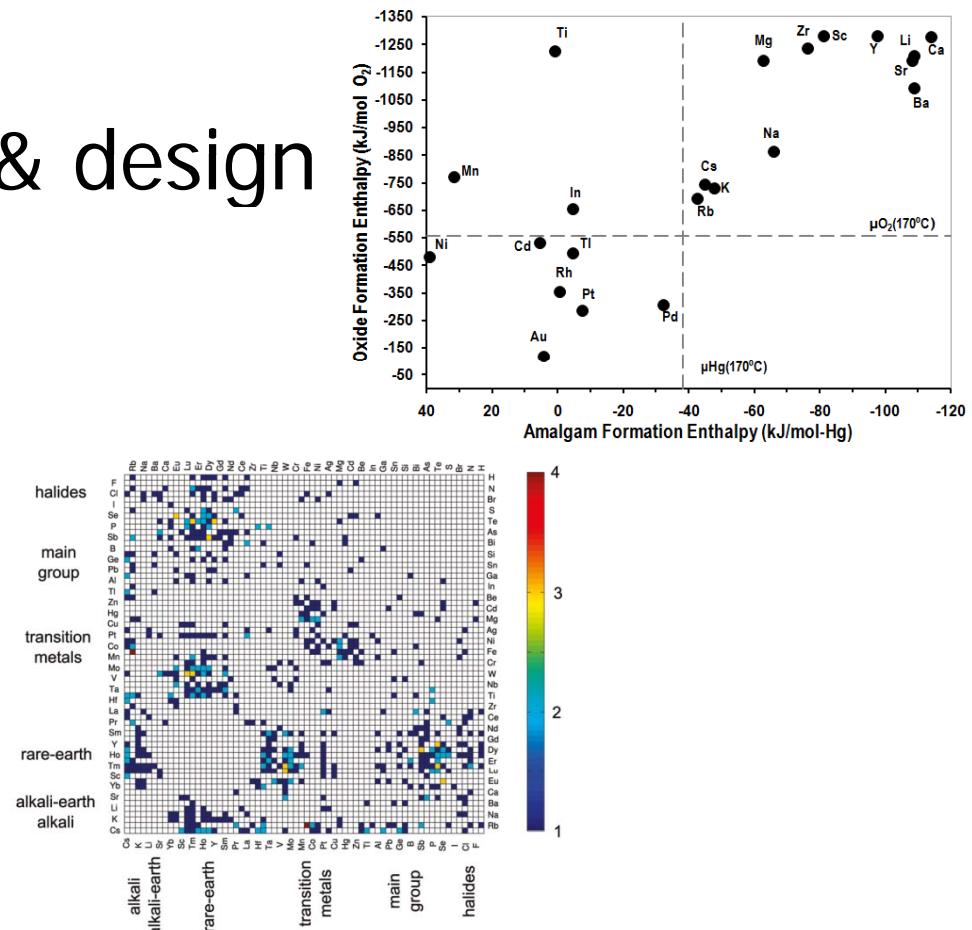
1. High-throughput DFT applied to Li ion-battery cathodes
2. **The Materials Genome at MIT:
a resource for materials informatics**

Goals of the Materials Genome

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

Materials screening & design

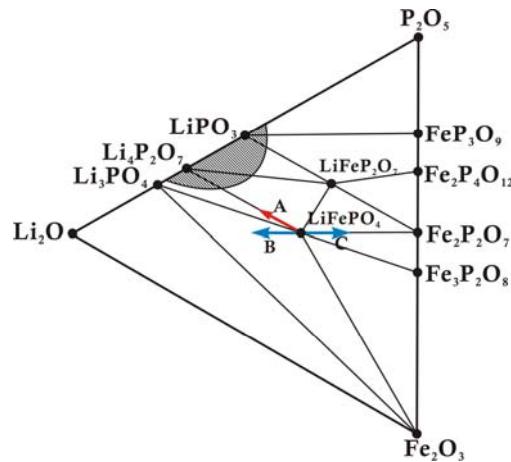
Structure Prediction



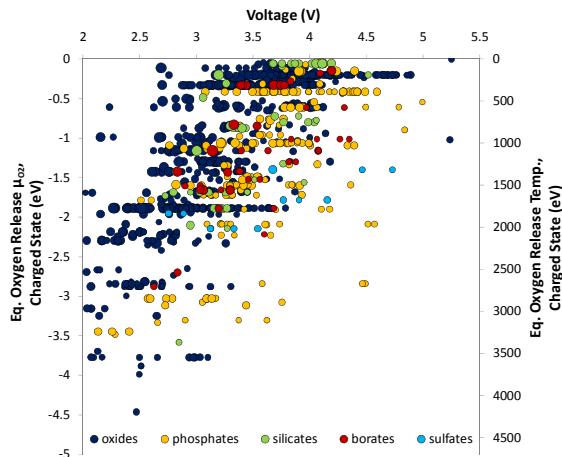
Goals of the Materials Genome

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

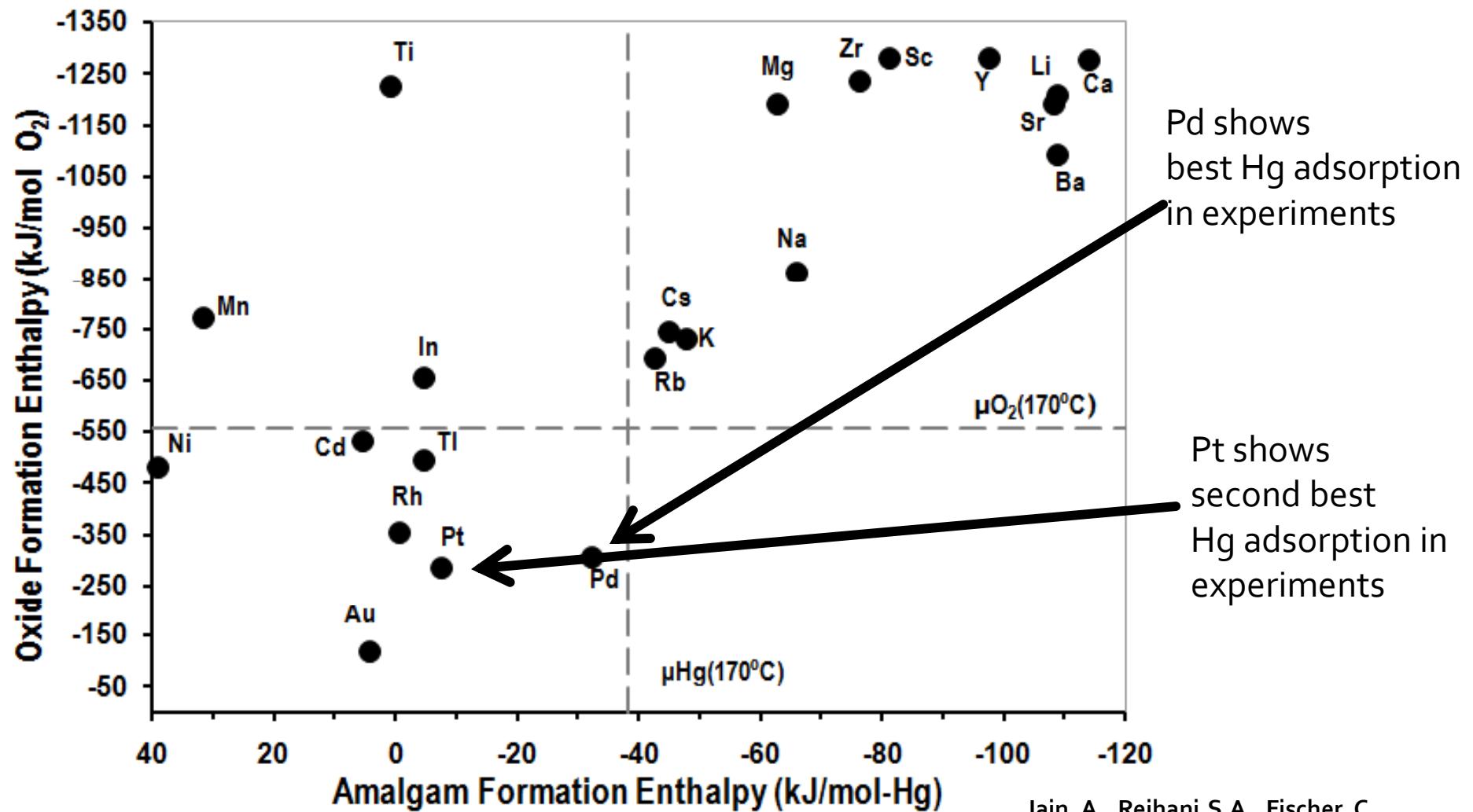
Analysis



Data mining



Computations predict best performing metal sorbents



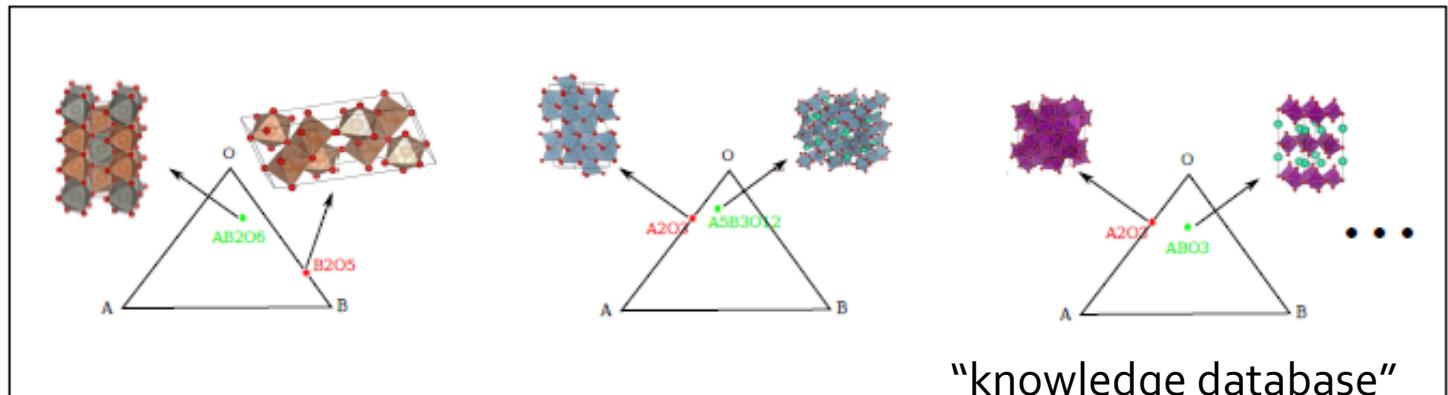
Pd shows
best Hg adsorption
in experiments

Pt shows
second best
Hg adsorption in
experiments

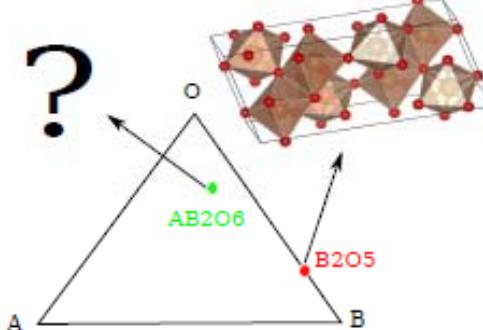
Jain, A., Reihani, S.A., Fischer, C.,
Couling, D., Ceder, G., Green, W.H.
Chemical Engineering Science (2010)
[in press]

Structure prediction methodology

Step 1: Predict
(using data-mined correlations)

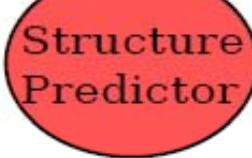


Partially unknown system



Statistical

Inference



Fischer, C. C., Tibbetts, K. J.,
Morgan, D., & Ceder, G.
Nature Materials (2006)

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

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

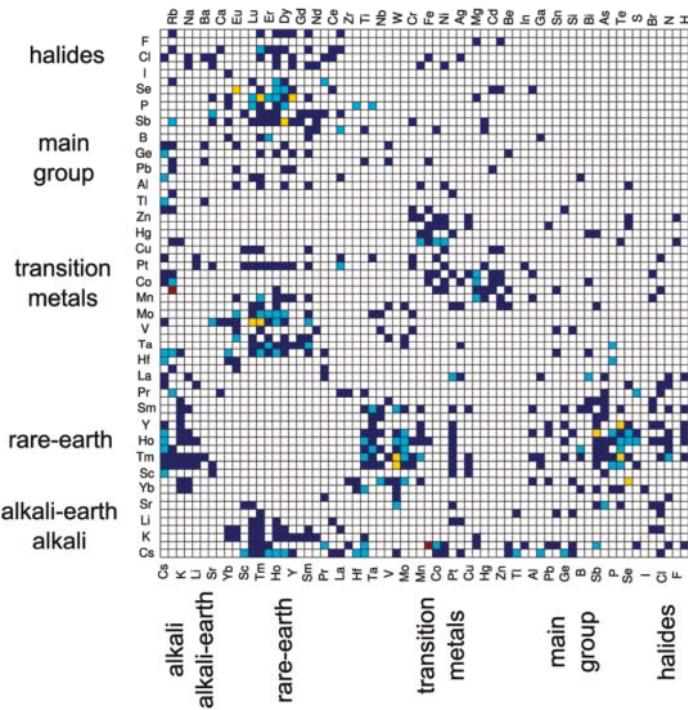
$$H = \sum_{i=1}^{N_e} \nabla_i^2 + \sum_{i=1}^{N_e} V_{nuclear}(r_i) + \sum_{i=1}^{N_e} V_{effective}(r_i)$$

Step 2: Confirm
(using DFT computations)

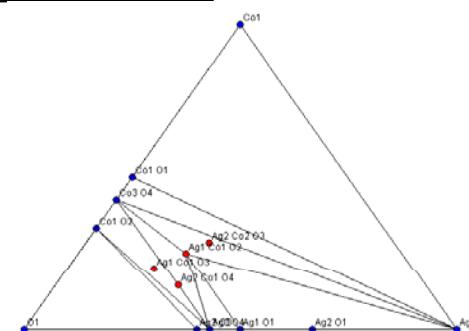
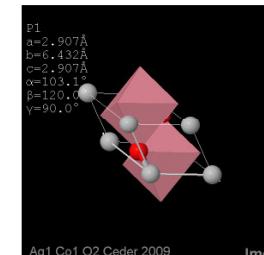


New ternary oxides predicted

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

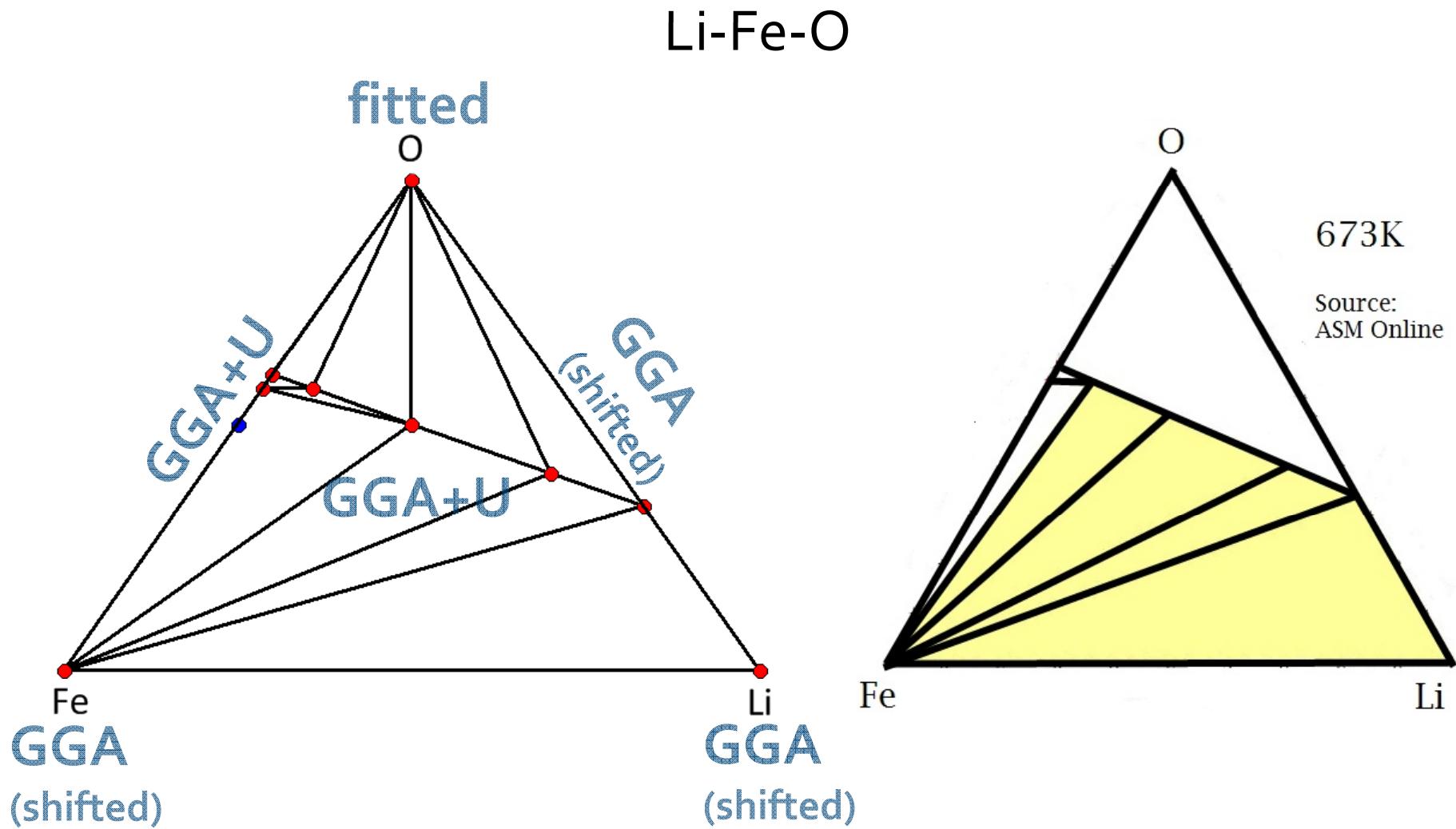


[structure id: 261981](#)
[chemical system: Ag-Co-O](#)
[formula: Ag₁Co₁O₂](#)
[based on: Ag₁In₁O₂ \(ICSD number: 91058\)](#)
[spacegroup: 166 \(R₃-m H\)](#)
[oxidation state: {Ag1.0+, Co3.0+}](#)
[minimum oxygen chemical potential: -0.44 eV/atom](#)
[maximum oxygen chemical potential: 0.289 eV/atom](#)
[temperature range stable in air: 0K - 386K](#)
[minimum energy decomposition: Co₃O₄\(198203\):0.583;Ag₁\(167256\):0.083;Ag₁O₁\(87771\):0.333](#)
[inverse distance to hull: 0.047 eV/atom](#)
[powder diffraction reference: 04-007-9668](#)

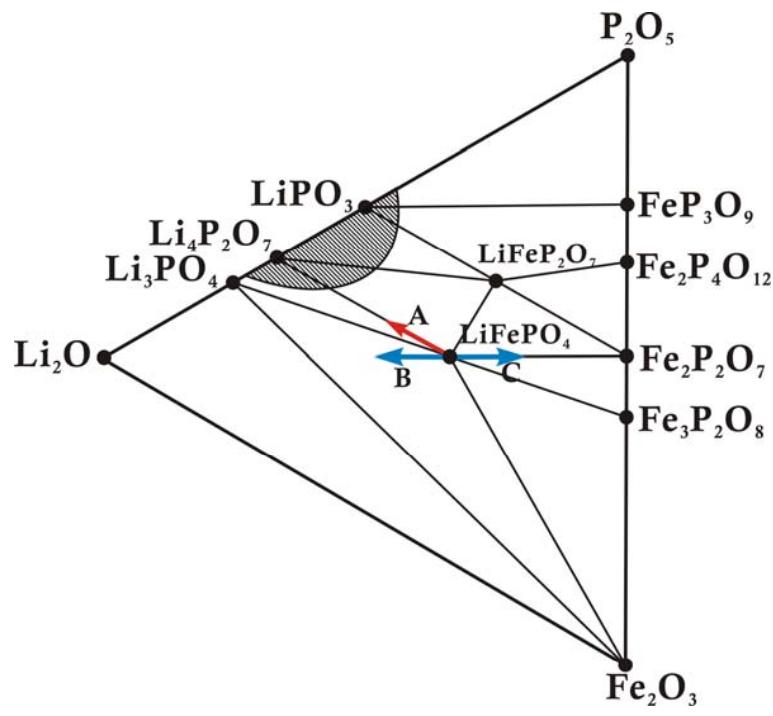


[1] Hautier, G., Fischer, C., Jain, A., Mueller, T., Ceder, G. *Chemistry of Materials* (2010)

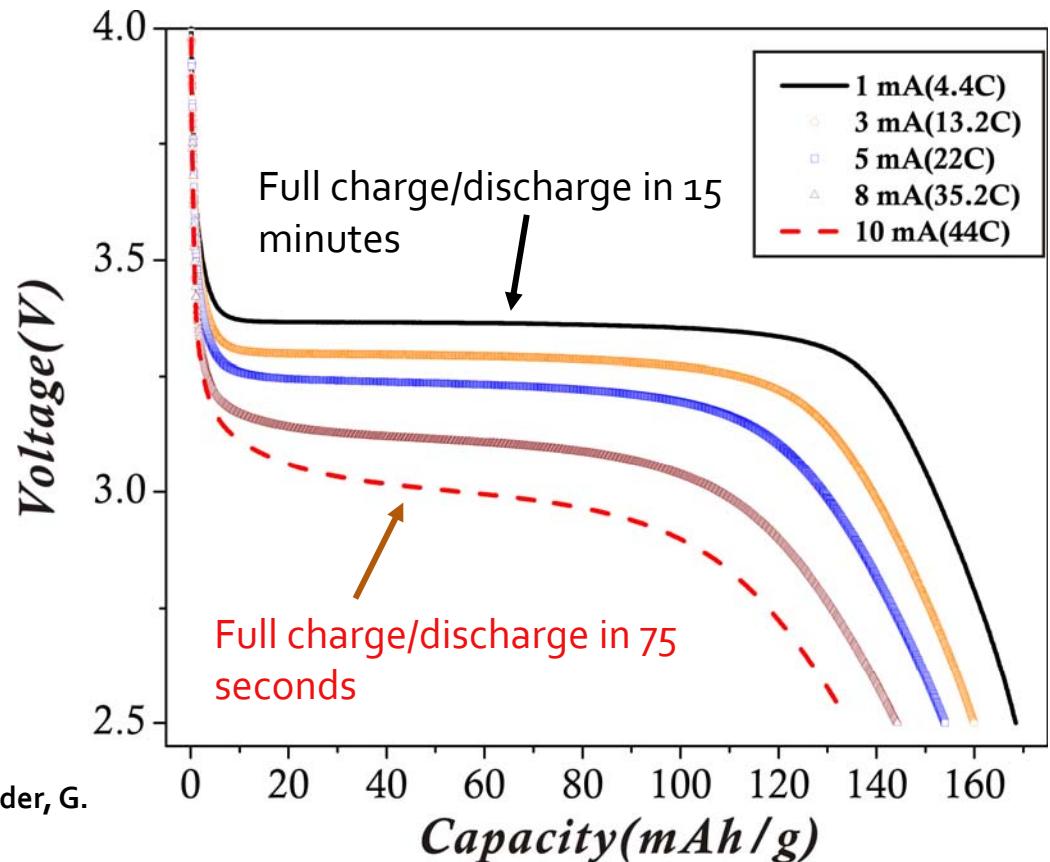
Can generate computed 0K, 0atm phase diagrams for almost all chemical systems



Phase diagrams lead to ultrafast battery cathode



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 Project

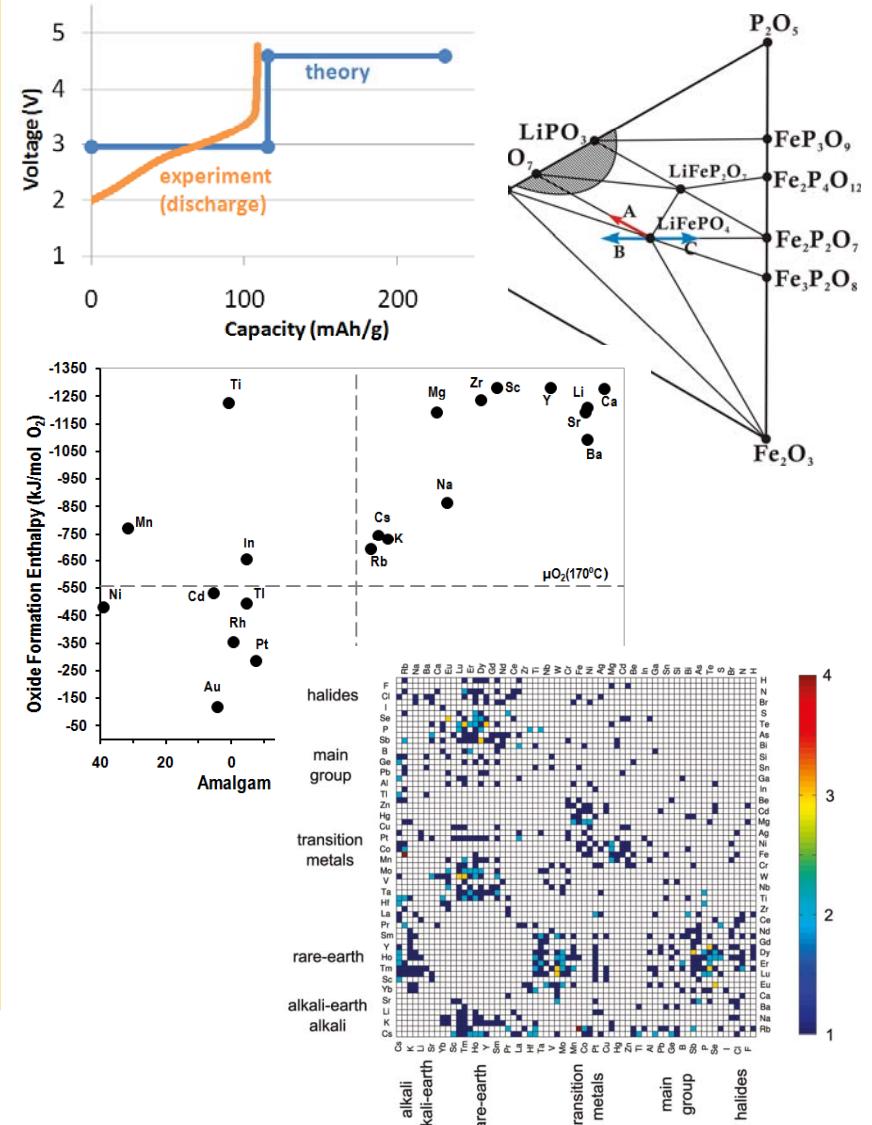
Number of Unique Elements: 2
Elements:

MATERIALS GENOME

Search Results

structure id	entry id	sum formula(normalized)	num sites (vasp)	spacegroup hnname	final energy per atom
5481	5481	Li ₂ O ₁	3	F m -3 m	null
8181	8181	Li ₁ O ₁	8	P -6	null
88069	87628	Li ₁ O ₁	8	P63/mmc	-4.8499175025
88070	87628	Li ₁ O ₁	8	P63/mmc	-4.8499175025
88071	87629	Li ₁ O ₁	8	P63/mmc	-4.8503746175
88072	87630	Li ₂ O ₁	3	Fm-3m	-4.77099761666667
88073	87630	Li ₂ O ₁	3	Fm-3m	-4.77099761666667
88074	87631	Li ₂ O ₁	3	Fm-3m	-4.7710484
88075	87632	Li ₁ O ₁	8	P63/mmc	-4.70763118625
88076	87632	Li ₁ O ₁	8	P63/mmc	-4.70763118625
88077	87633	Li ₁ O ₁	8	P63/mmc	-4.71186535875
94555	91931	Li ₁ O ₁	8	P63/mmc	-4.84987317625
94556	91931	Li ₁ O ₁	8	P63/mmc	-4.84987317625
94557	91932	Li ₁ O ₁	8	P63/mmc	-4.85019841375
98986	94850	Li ₁ O ₁	8	P63/mmc	-4.70742371375
98987	94850	Li ₁ O ₁	8	P63/mmc	-4.70742371375
98988	94851	Li ₁ O ₁	8	P63/mmc	-4.71162184875
104032	98174	Li ₂ O ₁	3	Fm-3m	-4.77110641333333
104033	98174	Li ₂ O ₁	3	Fm-3m	-4.77110641333333
104034	98175	Li ₂ O ₁	3	Fm-3m	-4.77117312
13949	13949	Li ₁ O ₁	8	P 63/m m c	null
352979	264100	Li ₂ O ₁	3	null	-4.77119122333333
352978	264099	Li ₂ O ₁	3	null	-4.77119122
352978	264099	Li ₂ O ₁	3	null	-4.77119122

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
 - Byoungwoo Kang
 - Hailong Chen
 - Robert Doe
 - Xiaohua Ma
 - Jae Chul Kim
 - Xiaofei Fang
- Ceder group
- Funding
 - Umicore
 - Bosch
 - DOE CSGF

BACKUP SLIDES

The Materials Genome: A Tool for Advancing Materials Informatics

File Tools

capacity (mah/c)

e above hull (delithiated)

energy density (wh/kg)

initialfinalnotes

overall stability rating

voltage (avg.)

Include ALL (duplicates, removals...) **GO!** Export cifs Export results VASP inputs Analysis Filter

511 entries match

sum formula	voltage (avg.)	capacity (mah/g)	e above hull (lithi...)	e above hull (deli...)	valence text diff...	valence text diff...	coordi...
Li ₇ O ₂ Ti ₁₁	2.6954207400...	83.688732410...	0--0	0.0490699288...	Ti:3+--Ti:3+	Ti:4+--Ti:4+	0:4.53
Li ₄ O ₈ Ti ₄	2.654583785...	308.21958136...	0---0--0	0.345742464...	Ti:3+--Ti:3+--...	Ti:4+--Ti:3.50...	0:5.86
Li ₂ O ₄ Ti ₂	1.605799815...	308.21958136...	0.0041093487...	0.0016269349...	Ti:3+--null--null	Ti:4+--null--null	0:5.89
Li ₇ O ₁₂ Ti ₅	2.6956740533...	167.25190943...	0.0066086702...	0.1703648421...	Ti:3+--Ti:3+	Ti:4+--Ti:4+	0:5.98
Li ₂ O ₄ Ti ₂	1.593053745	308.21958136...	0.0071577175...	0.0014427366...	Ti:3+	Ti:4+	0:5.88
Li ₈ O ₃₂ Ti ₁₅	2.8483487449...	83.252488010...	0.0097028528...	0.0734882034...	Ti:3.43+--Ti:3...	Ti:4+--Ti:4+	0:4.69 Ti:5.72 ...
Li ₅ O ₈ Ti ₃	2.10807855...	87.350475791...	0.0191455715...	0.0572278783...	Ti:3.67+--...	Ti:4+--Ti:4+	0:5.30 Ti:5.67 ...
Li ₁ O ₂₀ Ti ₁	1.0005500575...	308.21958136...	0.07411011504...	0.1555395083...	Ti:3+--...	Ti:4+--Ti:4+	0:4.69 Ti:5.72 ...

Composition Phase Diagram of Li-O-P-Ti

File Options Tools Inspector Method Window ?

missing license, cannot find = C:\Documents and Settings\stanubhai\Desktop\src\javaview\register at www.javaview.org

Phase Diagram Center

Ctrl-Click to (de)select elements (note that commonly used Li, P and O are preselected by default)

AA : Generic cation A	Cm : Curium	Hg : Mercury	Np : Neptunium	Sm : Samarium
Ac : Actinium	Co : Cobalt	Ho : Holmium	O : Oxygen	Sn : Tin
Ag : Silver	Cr : Chromium	I : Iodine	Os : Osmium	Sr : Strontium
Al : Aluminum	Cs : Cesium	In : Indium	P : Phosphorus	Ta : Tantalum
Am : Americium	Cu : Copper	Ir : Iridium	Pa : Protactinium	Tb : Terbium
Ar : Argon	D : Deuterium	K : Potassium	Pb : Lead	Tc : Technetium
As : Arsenic	DD : Generic cation D	Kr : Krypton	Pd : Palladium	Tell : Tellurium
At : Astatine	Dy : Dysprosium	La : Lanthanum	Pm : Promethium	Th : Thorium
Au : Gold	EE : Generic cation E	L : Lithium	Po : Polonium	Ti : Titanium
B : Boron	Er : Erbium	Lu : Lawrencium	Pr : Praseodymium	Tl : Thallium
BB : Generic cation B	Es : Einsteinium	M : Lutetium	Pt : Platinum	Tm : Thulium
Ba : Barium	Eu : Europium	M : Generic cation M	Pu : Plutonium	U : Uranium
Be : Beryllium	F : Fluorine	Md : Mendelevium	Ra : Radium	V : Vanadium
Bi : Bismuth	FF : Generic cation F	Mg : Magnesium	Rb : Rubidium	W : Tungsten
Bk : Berkelium	Fe : Iron	Mn : Manganese	Re : Rhenium	X : Generic anion X
Br : Bromine	Fm : Fermium	Mo : Molybdenum	Rh : Rhodium	XX : Generic anion XX
C : Carbon	Fr : Francium	N : Nitrogen	Rs : Radon	Xe : Xenon
CC : Generic cation C	Ga : Gallium	Na : Sodium	Ru : Ruthenium	Y : Yttrium
Ca : Calcium	Gd : Gadolinium	Nb : Niobium	S : Sulfur	YY : Generic anion YY
Cd : Cadmium	Ge : Germanium	Nd : Neodymium	Sb : Antimony	Yb : Ytterbium
Ce : Cerium	H : Hydrogen	Ne : Neon	Sc : Scandium	ZZ : Generic anion ZZ
Cf : Californium	He : Helium	Ni : Nickel	Se : Selenium	Zn : Zinc
Cl : Chlorine	Hf : Hafnium	No : Nobelium	Si : Silicon	Zr : Zirconium

Grand Can. Options

Grand Canonical

Projected Element

O : Oxygen

$\mu = 0.0000$

Temperature = 300

Part. Pres. (atm) 1

Reducing Agent None

Crit. μ list

Entry Options

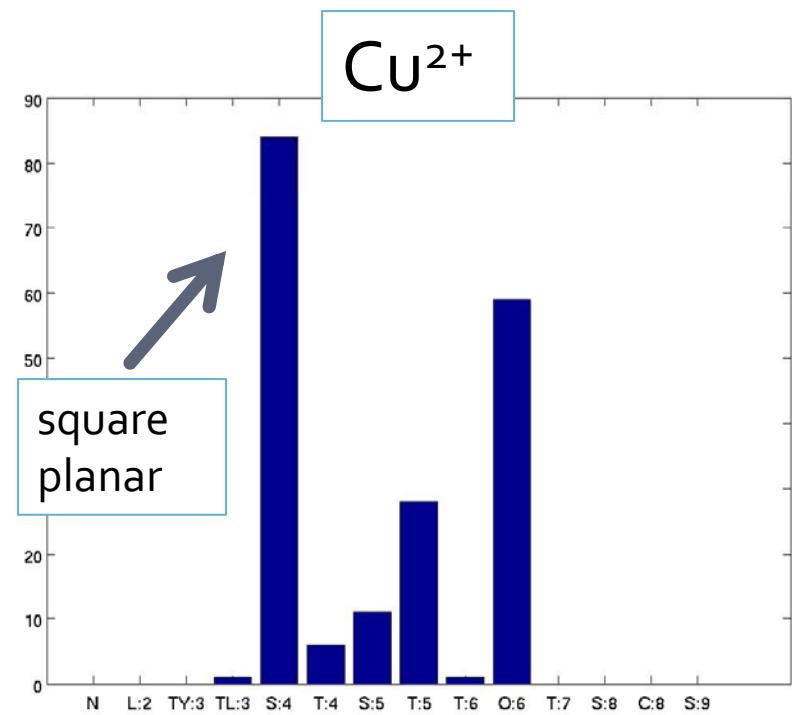
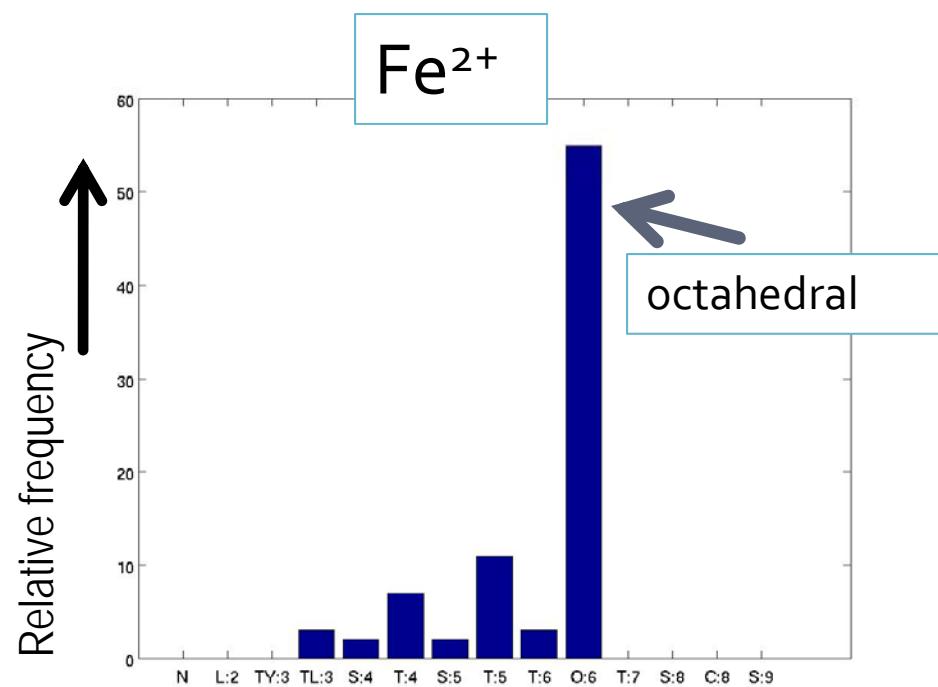
ICSD only

Make PD Reset Data Redox Analysis Phase Stability Range Compare PDs Get Report

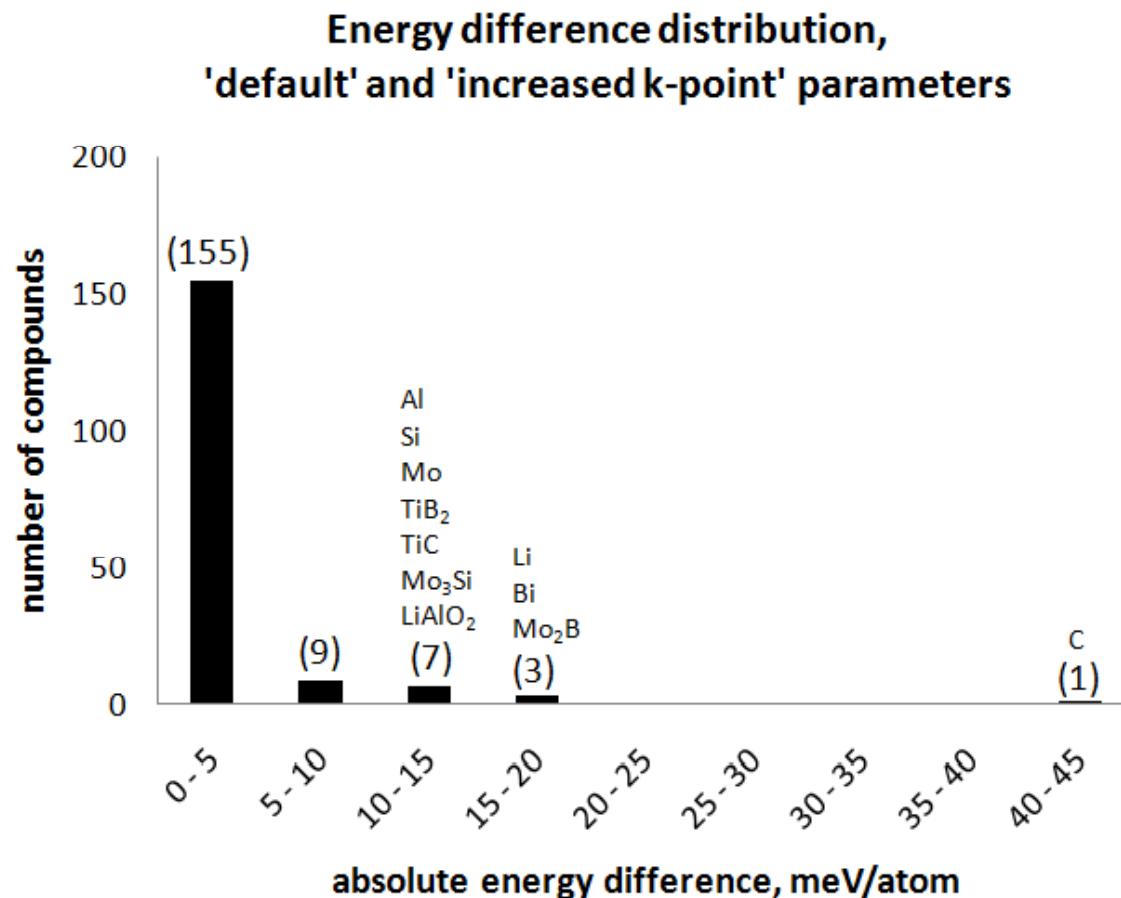
0%

Ready

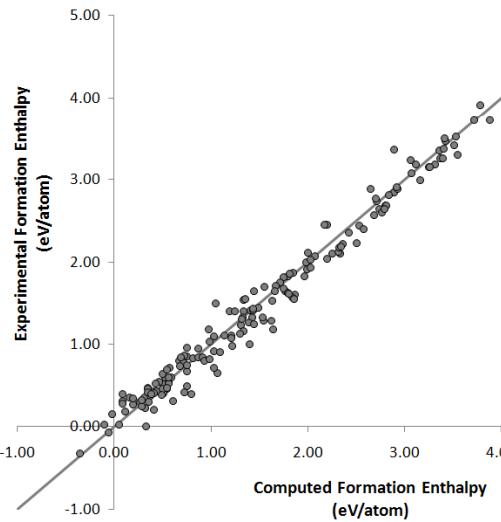
Designing Stable Structures: Local Environment Statistics



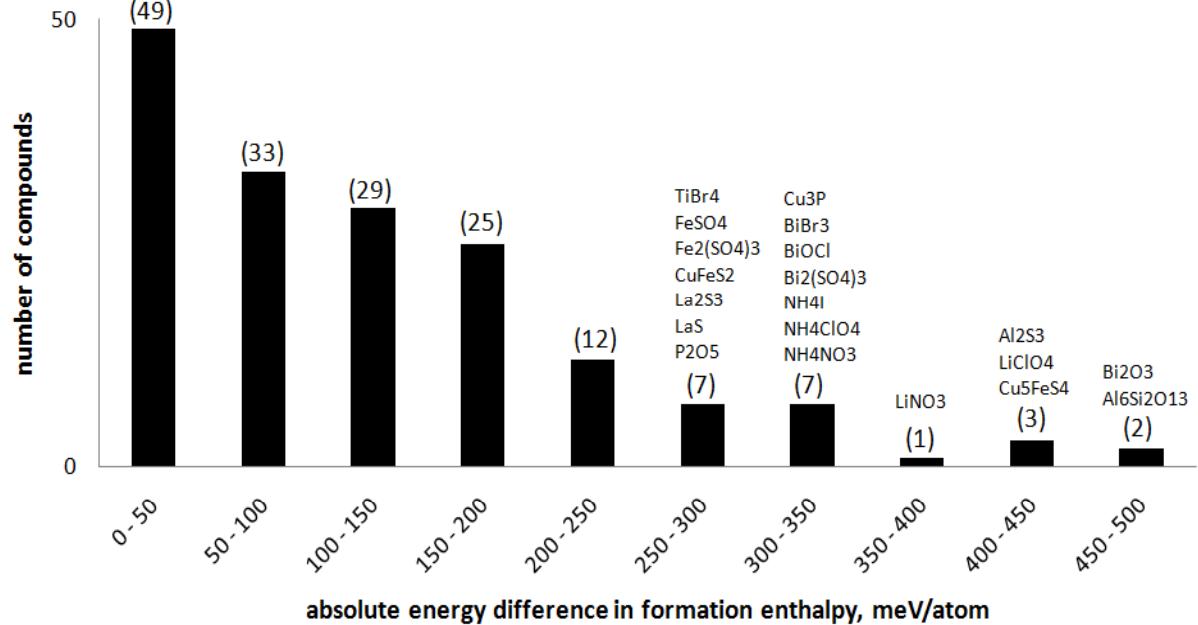
K-point convergence



Formation enthalpy accuracy



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



Improving coal gasification efficiency by removing trace Hg gas at high T

- 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

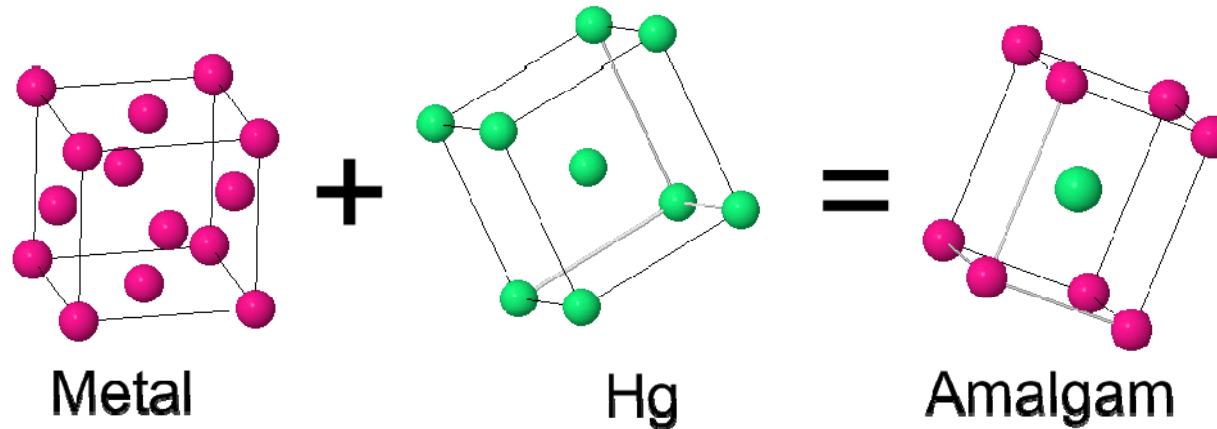
Nexant Inc., *Environmental footprints and costs of coal-based integrated gasification combined cycle and pulverized coal technologies.* 2006, Environmental Protection Agency.

Schlather, J. and B. Turk. *Comparison of a New Warm-Gas Desulfurization Process versus Traditional Scrubbers for a Commercial IGCC Power Plant.* 2007.

Improving coal gasification efficiency by removing trace Hg gas at high T

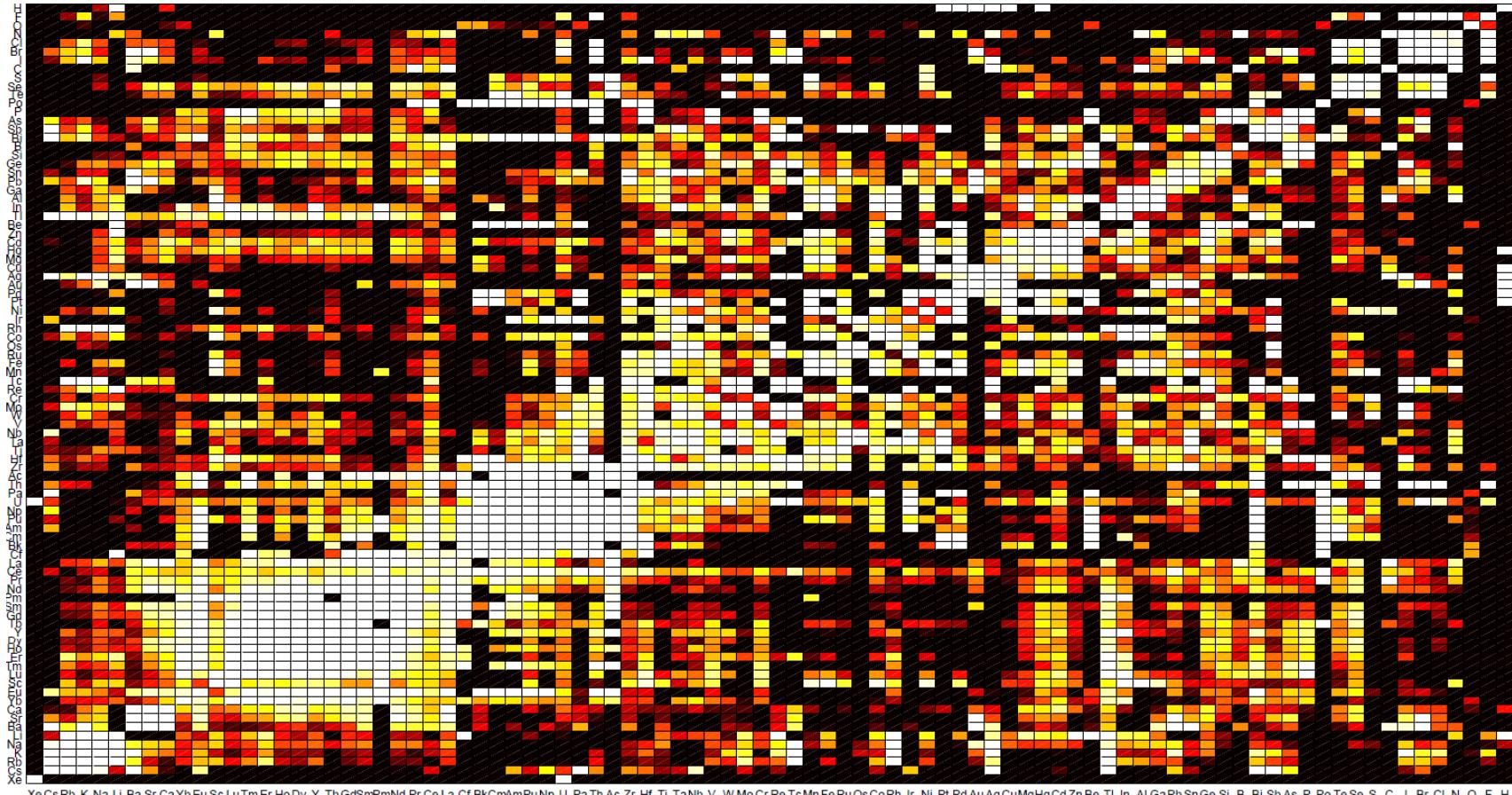
Goal: find a metal capable of removing 100ppbw Hg gas from a syngas stream at high T

Strategy: compute all formation enthalpies:



for all known binary Metal-Hg amalgams
(along with oxidation energies)

Data mining Substitution Probabilities



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