ICME 2016 Barcelona

Software Platforms for Electronic/Atomistic/Mesoscopic Modelling: Status and Perspectives

Erich Wimmer, David Reith, Roman Tarnovsky, Clive Freeman and Paul Saxe
Materials Design

15 April 2016
Outline

► Context and Purpose
► Integrated Computational Materials Engineering (ICME)
► Requirements of integrated software platforms
► Implementation
► Examples
  • Metallurgy
  • Li-ion batteries
► Perspectives
better, cheaper, faster, cleaner and sustainable
Software Platforms

Requirements

- State-of-the-art scientific capabilities
- Comprehensive functionality
- Validated
- Easy of use
- Well documented
- Robust
- Computationally efficient
- Fully supported over decades on changing hardware and operating systems
- Extensible
- Interoperability with other software components and platforms
- Other
Interoperability

Between …

- different computational codes
  - Ab initio solid state (VASP)
  - Ab initio molecular (Gaussian, Turbomole)
  - Semi-empirical molecular (MOPAC)
  - Forcefield (interatomic potentials) molecular dynamics (LAMMPS)
  - Forcefield Monte Carlo (GIBBS)

- different experimental databases

- experimental databases and computational codes

- electronic/atomistic/mesoscopic (e/a/m) codes

- e/a/m and continuum codes
## Database Search ICSD, Pearson, Pauling (639,632 entries)

<table>
<thead>
<tr>
<th>ID</th>
<th>completeness</th>
<th>space group name H-M</th>
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**Search Criteria**
- Require that Number of Elements is equal to
- Require that Structural Completeness is Complete

**Displaying 500 of 4156 hits**

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**MedeA®- InfoMaticA**

Database Search ICSD, Pearson, Pauling (639,632 entries)

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Search Criteria
- Formula
- Has a ratio of 1 to 3
- Transition metals
- Atoms of 0

Number of Elements is equal to 3

Displaying 500 of 756 hits

756 unique structures
Interoperability between Databases and e/a/m Codes

![Database and code interaction](image-url)

**LiNbO₃**

**Symmetry**
- Spacegroup: R-3
- Z: 6
- Volume: 317.912
- Calculated density:

**Cell**
- a: 5.147±0.
- b: 5.147±0.
- c: 13.857±0.
- α: 90.±0.
- β: 90.±0.
- γ: 120.±0.

**Reference**
Interoperability between e/a/m Codes
Interoperability between e/a/m Codes

Self-defining file format for structural data used as input for VASP, LAMMPS, …
Interoperability between e/a/m Codes

Perform property calculations with VASP and LAMMPS
## Computed Results

Contains complete record of all input and output data including computational protocol, version number

| □ 55220 | ewimmer | contractcluster | 5 | LGPS_200 amorphous Li diffusion 400...600 NVT 2ns pccf_LGPS_f47q | finished 2016-01-12 08:59:27 | 2016-01-12 08:59:28 | 2016-01-12 17:03:42 |
| □ 55218 | ewimmer | contractcluster | 5 | LGPS nanopowder_02 Li diffusion 400...600 NVT 2ns pccf_LGPS_f47q | finished 2016-01-12 08:59:02 | 2016-01-12 08:59:02 | 2016-01-13 11:09:01 |
| □ 55212 | ewimmer | contractcluster | 5 | LGPS 200 crystal Li diffusion 400...600 NVT 2ns pccf_LGPS_f47q | finished 2016-01-12 08:58:14 | 2016-01-12 08:58:14 | 2016-01-12 16:36:08 |

Diffusion coefficient information, derived from regression analysis above

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Results including value, units, and numerical uncertainty
Computed Materials Properties

Structural properties
- Molecular structures
- Crystal structures
- Surface structures
- Structure around defects
- Adsorption geometries
- Structures of interfaces
- Liquids and amorphous systems

Thermo-Mechanical properties
- Density
- Elastic moduli
- Thermal expansion coefficients
- Fracture

Thermodynamic properties
- $\Delta U$, $\Delta H$, $\Delta S$, $\Delta G$, heat capacity
- Binding energies
- Solubility
- Melting temperature
- Vapor pressure
- Miscibility
- Phase diagrams
- Surface tension

Chemical properties
- Chemical reaction mechanisms and rates
- Reactivity on surfaces
- Solid-solid reactions
- Photochemical reactions

Transport properties
- Mass diffusion coefficient
- Permeability
- Thermal conductivity
- Viscosity
- Electrical conductivity

Electronic, optical, and magnetic properties
- Electron density distribution - electrical moments
- Polarizabilities, hyperpolarizabilities
- Optical spectra
- Dielectric properties
- Piezoelectric properties
- Electrostatic potential
- Spin density distribution, magnetic moments
- Energy band structure - metal, semiconductor, insulator
- Band gaps, band offsets at hetero-junctions
- Ionization energies and electron affinities
- Work function

Low-strain Cathode Materials

Computational Design and Experimental Verification of Zero- and Low-strain Cathode Materials for Solid-State Li-ion batteries

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¹Toyota Motor Europe, Advanced Technology 1, Hoge Wei 33, Zaventem, Belgium
²Materials Design S.A.R.L., Montrouge, France

Background

- One of the major degradation factors in Li-ion batteries is the volume change occurring upon charge and discharge in electrode materials [1].
- This volume change can be easily accommodated in conventional composite electrodes, but in the case of a rigid, solid-state battery the stress generated at the grain interfaces will lead to the destruction of the device.
- For the anode, Li₄Ti₅O₁₂ is a strong candidate for solid-state Li-ion batteries, giving its zero-strain properties [2].
- For the cathode, on the other hand, no viable zero-strain candidate has been described in the literature yet.
- Finding a zero-strain cathode material with high-voltage by trial-and-error is clearly not a viable strategy → ab-initio Material Design
- We employed DFT calculations to link the volume change, the composition and the lithium content of various precursor materials with the spinel structure.
- Using Vegard’s law [3] we mixed the precursors to obtain materials with the desired strain properties

International Battery Association (IBA) Conference, Brisbane, Australia, 3-7 March 2014

Low-strain Cathode Materials

\[
x \text{LiM}^1_2 \text{O}_4 + y \text{LiM}^2_2 \text{O}_4 + z \text{LiM}^3_2 \text{O}_4 = \text{Li(M}^1_x \text{M}^2_y \text{M}^3_z \text{)} \text{O}_4
\]

M = Mg, V, Cr, Mn, Fe, Co, Ni, Cu, Al

Expansion (\(\Delta V\))

Contraction (-\(\Delta V\))
(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
4 December 2014 (04.12.2014)

(51) International Patent Classification:
C01G 45/12 (2006.01)
H01M 10/052 (2010.01)
H01M 4/505 (2010.01)
H01M 10/0562 (2010.01)

(21) International Application Number:
PCT/EP2013/060881

(22) International Filing Date:
27 May 2013 (27.05.2013)

(25) Filing Language: English

(26) Publication Language: English

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(74) Agents: HART-DAVIS, Jason et al.; Cabinet Beau de Lomenie, 158 rue de l'Université, F-75340 Paris Cedex 07 (FR).


Published: — with international search report (Art. 21(3))
Elastic Coefficients of Aluminum Nitride

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<th>Expt¹</th>
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<td>$C_{12}$</td>
<td>125</td>
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<tr>
<td>$C_{13}$</td>
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<td>$C_{44}$</td>
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<td>$C_{66}$</td>
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<tr>
<td>$B$</td>
<td>202</td>
<td>212</td>
<td>195</td>
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Thermal expansion

Interface matching between Si$_3$N$_4$ and Al is affected by temperature due to rather different thermal expansion coefficients of the constituent materials.

Thermal expansion is computed from first principles with MedeA.
The diffusion coefficient of H in Ni computed from first-principles has similar accuracy as experimental data at ambient and medium temperatures.

Isotope effects are well explained and quantitatively described.

Strength of Ni Grain Boundaries

Quantitative ranking of impurities and alloying elements

Grain boundaries with impurities

Cleavage Energy (J/m²)

strengthening

weakening

Impurity Atom

Interfaces: Al/Si$_3$N$_4$

Si-terminated

Al
Si
N

1.8 J/m$^2$

Work of separation

N-terminated

Al
N
Si

7.4 J/m$^2$
Bridging Length- and Time-Scales

- Larger systems
- More configurations
  - ab initio
  - cluster expansion
  - forcefields
  - Materials properties
  - engineering equations
  - continuum models
Determination of most stable ordered structures in Ni-Cr alloy system achieved by cluster expansion method as implemented in MedeA®-UNCLE.

- CrNi$_2$ with particular magnetic ordering is thermodynamically stable.
- There is a range of metastable structures such as S8.
Monte-Carlo simulation based on cluster expansion for fcc Ni-Cr system
Viscosity of Molten Ni

Stage 1.4: Viscosity using Green-Kubo. NVE integration for 2 ns with a timestep of 2 fs
The autocorrelation of the pressure tensor uses 2000 bins, starting with sampling every step, then every 2 steps, then every 4, etc.

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<th>After Steps</th>
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<td>T:</td>
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Perspectives

- Productivity of simulations
- Range and accuracy of simulations
- Compute power per €

Time
Perspectives

- Improve accuracy and estimation of error bars of theoretical approaches
- Stronger ties between modelers working on different length/time-scales and approaches
- Integration of experimental and computational data

Education, Training, and Communication
Acknowledgements

► Our customers

► Our partners

  • Georg Kresse and his team, Univ. Vienna - VASP
  • Steve Plimpton and his team, Sandia Natl. Lab. – LAMMPS
  • IFPEN and Univ. Paris - GIBBS
  • Jimmy Stewart – MOPAC
  • FIZ Karlsruhe – ICSD
  • Pierre Villars / ASM – Pearson, Pauling
  • NIST – NIST Crystal Data
  • Krzysztof Parlinski - Phonon
  • and all other partners

► All colleagues in our company