MatCASE

Forward Simulation & Inverse Degisn

Zi-Kui Liu

Department of Materials Science and Engineering

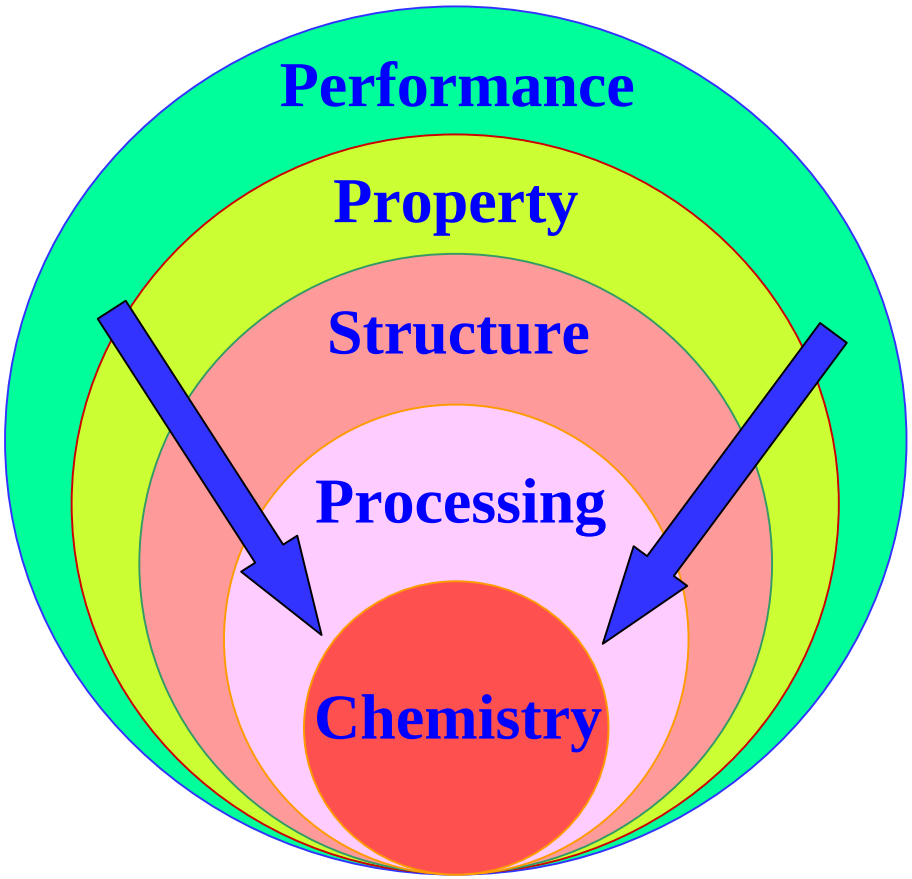
Pennsylvania State University

http://www.phases.psu.edu

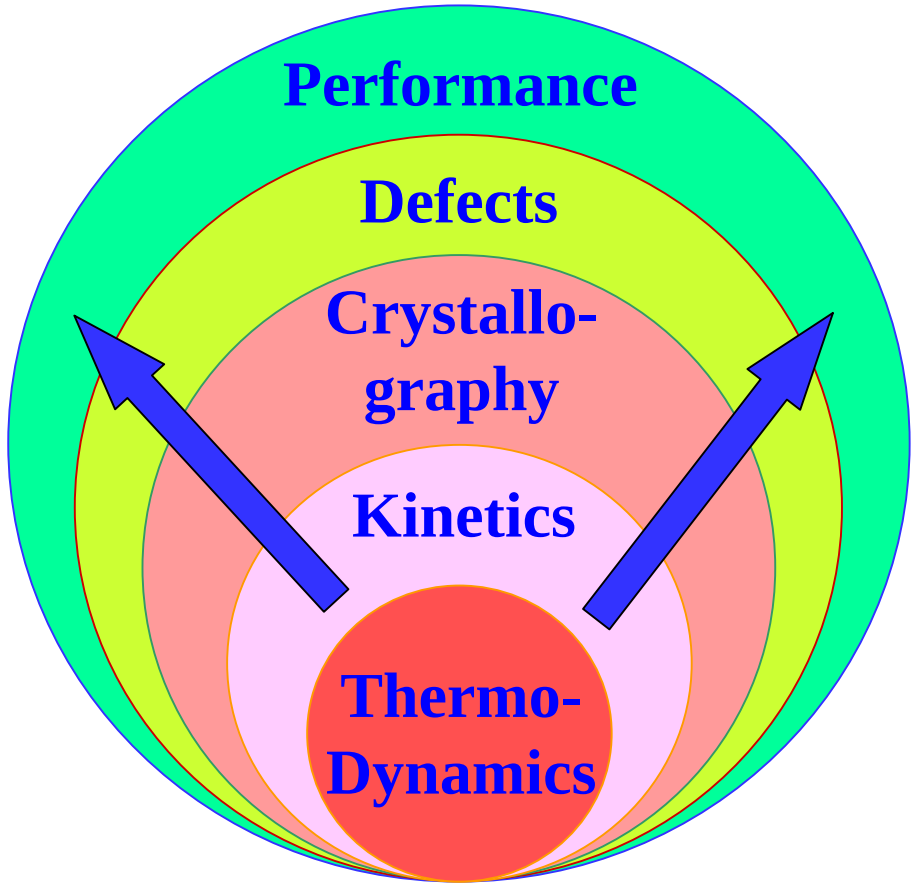
Outline

- Introduction of NSF ITR Project (MatCASE)
- Data for thermodynamic modeling
  - First-principles calculations
  - Experimental data
- High throughput of thermodynamic modeling
  - A new algorithm for automation of phase equilibrium calculations
  - Automation of thermodynamic modeling
- Summary

Materials Engineering and Science



Top-Down, Inverse Design



Bottom-Up, Forward Simulation

MATCASE Project

NSF ITR (Materials Computation and Simulation Environment)

PI: Zi-Kui Liu, Long-Qing Chen, Padma Raghavan, Qiang Du, (Penn State), Stephen Langer (NIST), Christopher Wolverton (Ford)

Postdoctoral Fellows

- **Edwin Garcia, , Keita Teranishi, Yi Wang, Peng Yu, Wenxiang Zhu**

Graduate Students

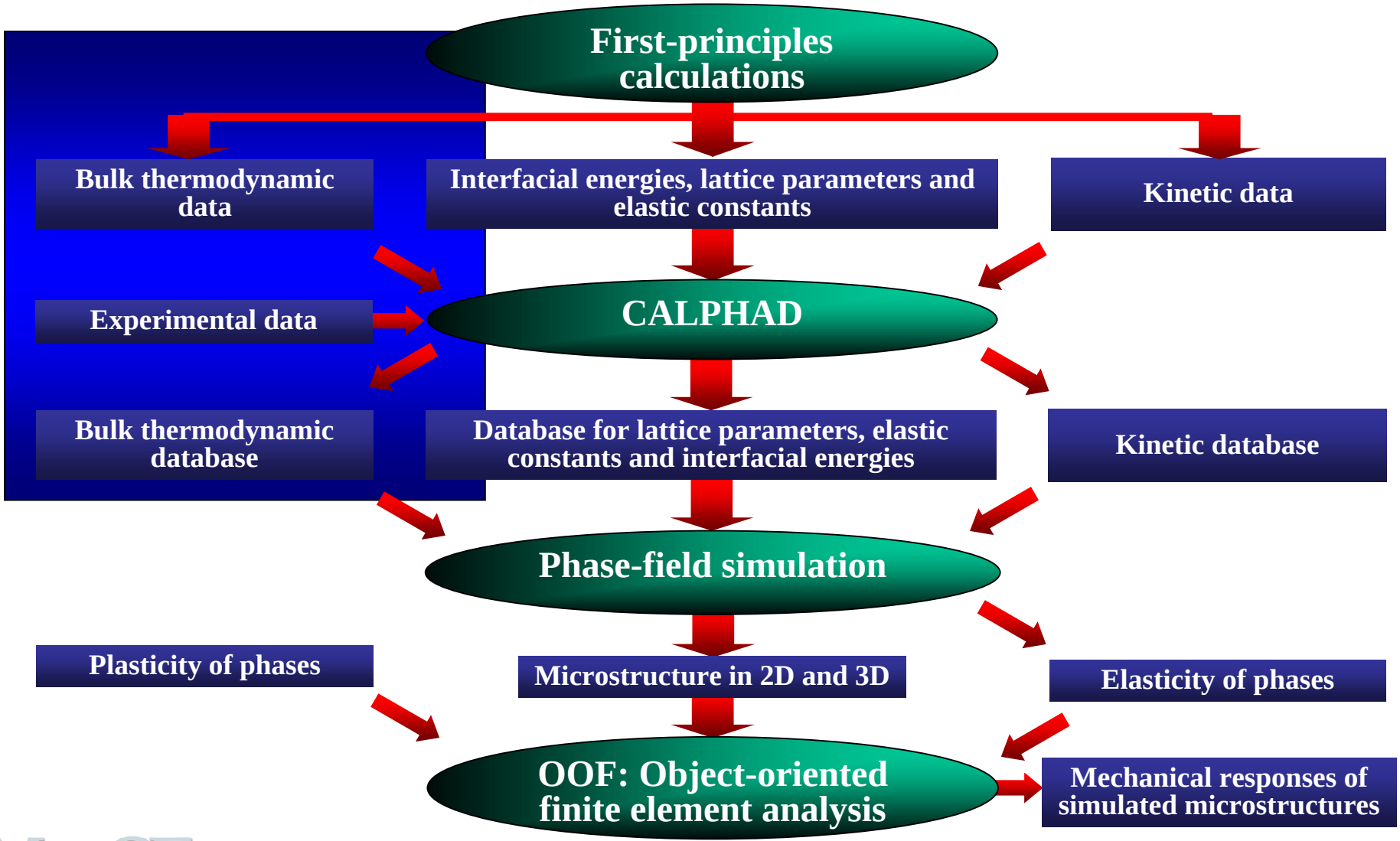
- **Maria Emelianenko, Weiming Feng, Qiujiang Li, Manjeera Mantina, Dongwon Shin, Anusha Srirama, Tao Wang, Hui Zhang, Jingxian Zhang**

Project Alumni

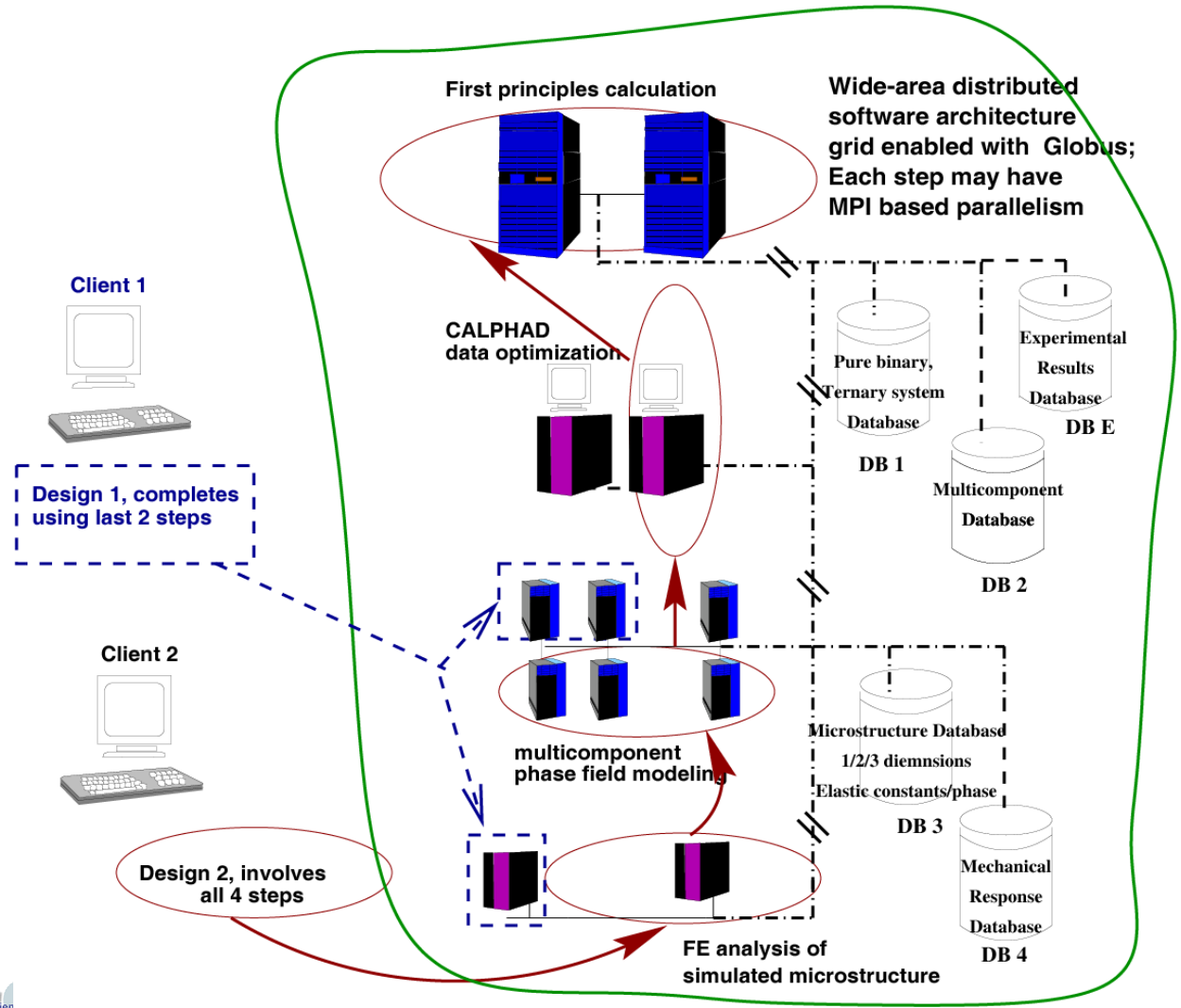
- **PhD awarded: Shenyang Hu, Chao Jiang, Keita Teranishi**
- **MS awarded: William Stevenson, Jianwei Wang**
- **Postdoc: Shenyang Hu, Chinnappan Ravi , Shihuai Zhou, Jingzhi Zhu**
- **Senior Personnel: Jorge Sofo**

Web site: http://www.matcase.psu.edu

MatCASE: Forward Simulation



MatCASE Software Architecture



Thermodynamic Modeling

Less accurate experimental measurements

Accurate experimental measurements

Thermochemical measurements: enthalpy, entropy, heat capacity, activity

Phase equilibria: liquidus, solidus, phase boundary,

Gibbs Energy of Individual Phases

Applications

$G = H - TS$

http://www.calphad.org

Pure elements→Binary→Ternary→Multicomponent

First-Principles Approach

Many-body Schrodinger's equation

$$\hat{H}\Psi(\vec{R}_1,\vec{R}_2,...\vec{R}_N,\vec{r}_1,\vec{r}_2,...\vec{r}_n) = E\Psi(\vec{R}_1,\vec{R}_2,...\vec{R}_N,\vec{r}_1,\vec{r}_2,...\vec{r}_n)$$

Born-Oppenheimer approximation

Many-electron Schrodinger's equation

$$\hat{H}\psi(\vec{r}_1,\vec{r}_2,...\vec{r}_n) = E\psi(\vec{r}_1,\vec{r}_2,...\vec{r}_n)$$

Density Functional Theory

$$E = E[\rho(\vec{r})]$$

Set of one-electron Schrodinger's equation

$$\left[-\frac{\hbar^2}{2m_e}\nabla_i^2 - \frac{e^2}{4\pi\varepsilon_0}\sum_{I=1}^{N}\frac{Z_I}{|\vec{r}-\vec{R}_I|} + \frac{e^2}{4\pi\varepsilon_0}\int\frac{\rho(\vec{r}')}{|\vec{r}-\vec{r}'|}d^3\vec{r}' + V_{XC}[\rho(\vec{r})]\right]\psi_i(\vec{r}) = \varepsilon_i\psi_i(\vec{r})$$

Pure Elements

- **78 pure elements in bcc, fcc and hcp structures**
  - **Y. Wang, S. Curtarolo, C. Jiang, R. Arroyave, T. Wang, G. Ceder, L.-Q. Chen and Z.-K. Liu, "Ab initio lattice stability in comparison with CALPHAD lattice stability," *CALPHAD, Vol.28, 2004, 79-90.***

Total Energy in bcc structure

Li 3.442 -1.8976 bcc	**Be** 2.505 -3.6046 hcp											**B** 2.313 -4.9213 rhomb	**C** 2.381 -4.8349 diamond	**N** - - -	**O** - - -	**F** - - -
Na 4.197 -1.3165 bcc	**Mg** 3.571 -1.4528 hcp	bcc lattice constant in Å total energy in eV/atom room temperature structure										**Al** 3.244 -3.6012 fcc	**Si** 3.112 -4.8468 diamond	**P** 3.076 -4.6498 complex	**S** 3.168 -3.0245 complex	**Cl**
K 5.271 -1.0384 bcc	**Ca** 4.362 -1.8997 fcc	**Sc** 3.666 -6.1185 hcp	**Ti** 3.241 -7.7002 hcp	**V** 2.992 -8.9632 bcc	**Cr** 2.847 -9.4655 bcc	**Mn** 2.792 -8.8117 complex	**Fe** 2.822 -8.2748 bcc	**Co** 2.809 -6.8834 hcp	**Ni** 2.799 -5.2954 fcc	**Cu** 2.886 -3.6082 fcc	**Zn** 3.137 -1.0269 hcp	**Ga** 3.378 -2.8504 complex	**Ge** 3.389 -4.1302 diamond	**As** 3.368 -4.2373 rhomb	**Se** 3.443 -2.9450 hex.	**Br** 3.758 -1.0369 -
Rb 5.661 -0.9292 bcc	**Sr** 4.736 -1.6197 fcc	**Y** 4.029 -6.2577 hcp	**Zr** 3.574 -8.3598 hcp	**Nb** 3.322 -10.0466 bcc	**Mo** 3.178 -10.7799 bcc	**Tc** 3.094 -9.9384 hcp	**Ru** 3.068 -8.4677 hcp	**Rh** 3.082 -6.8033 fcc	**Pd** 3.145 -5.1001 fcc	**Ag** 3.306 -2.7032 fcc	**Cd** 3.623 -0.7060 hcp	**In** 3.814 -2.5503 tetr	**Sn** 3.840 -3.7409 diamond	**Sb** 3.789 -3.9182 rhomb	**Te** 3.852 -2.8581 hex	**I** 4.149 -1.0833 complex
Cs 6.122 -0.8573 bcc	**Ba** 5.006 -1.9233 bcc		**Hf** 3.538 -9.6562 hcp	**Ta** 3.320 -11.7358 bcc	**W** 3.190 -12.7781 bcc	**Re** 3.125 -11.9107 hcp	**Os** 3.099 -10.2440 hcp	**Ir** 3.120 -8.1765 fcc	**Pt** 3.175 -5.9637 fcc	**Au** 3.319 -3.1829 fcc	**Hg** 3.883 -0.1957 -	**Tl** 3.970 -2.2301 hcp	**Pb** 4.009 -3.5042 fcc	**Bi** 4.005 -3.7182 rhomb	**Po** Sc	**At** - - -
		La 4.219 -4.7823 hex	**Ce** 3.772 -5.7318 fcc	**Pr** 4.161 -4.6262 hex	**Nd** 4.122 -4.5791 hex	**Pm** 4.096 -4.5436 -	**Sm** 4.066 -4.4984 complex	**Eu** 4.429 -1.8587 bcc	**Gd** 4.027 -4.4549 hcp	**Tb** 4.006 -4.4213 hcp	**Dy** 3.987 -4.3882 hcp	**Ho** 3.969 -4.3574 hcp	**Er** 3.947 -4.3384 hcp	**Tm** 3.933 -4.3158 hcp	**Yb** fcc	**Lu** 3.896 -4.2813 hcp
Fr -	**Ra** -	**Ac** 4.494 -3.9339 fcc	**Th** 4.018 -7.2039 fcc	**Pa** 3.677 -9.2207 tetr	**U** 3.437 -10.8731 complex	**Np** 3.294 -12.3007 complex	**Pu** 3.207 -13.4703 complex	**Am** hex	**Cm** -	**Bk** -	**Cf** -	**Es** -	**Fm** -	**Md** -	**No** -	**Lr** -

Y. Wang et al., CALPHAD, Vol.28, 2004, 79-90.

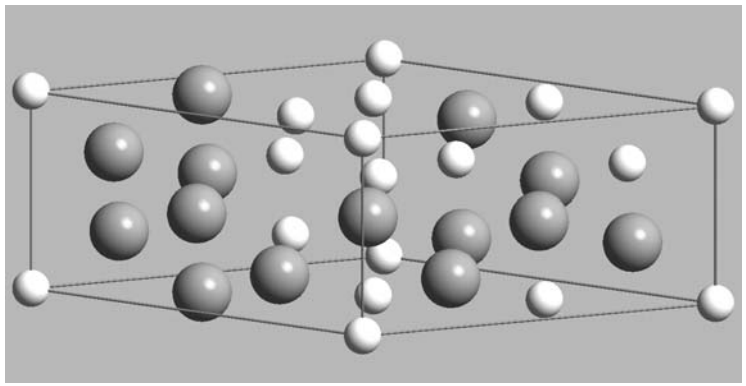
Binary: Energy of Formation

$$\Delta H(A_{1-x}B_x) = E(A_{1-x}B_x) - (1-x)E(A) - xE(B)$$

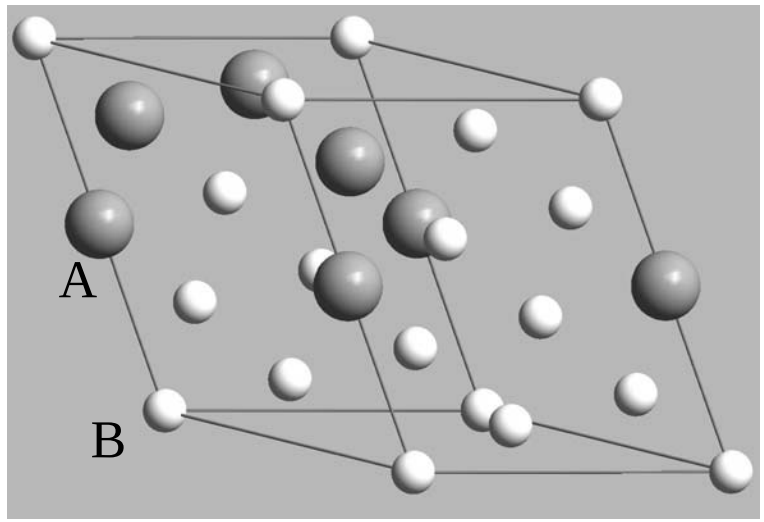
- **Stefano Curtarolo, Dane Morgan, and Gerd Ceder, "Accuracy of *ab-initio* methods in predicting the crystal structures of metals: review of 80 binary alloys", CALPHAD (2005), in press**
  - **15000+ calculations with statistical analysis.**
  - **Data mining to predict new structures.**
- **Phases Research Lab at Penn State**
  - **Y. Zhong, C. Wolverton, Y. A. Chang and Z. K. Liu, Acta Mater., 52, (2004) 2739-2754.**
  - **R. Arroyave, D. Shin and Z.-K. Liu, Acta Mater., 53, (2005) 1809-1819.**
  - **K. Ozturk, Y. Zhong, L. Q. Chen, C. Wolverton, J. O. Sofo and Z. K. Liu, Metall. Mater. Trans. A, 36A, (2005) 5-13.**
  - **Y. Wang, C. Woodward, S. H. Zhou, Z. K. Liu and L. Q. Chen, Scr. Mater., 52, (2005) 17-20.**

Solution Phases

- **Special Quasirandom Structures (SQS)**
  - **Small supercells that mimic the local pair and multibody correlations of random alloys**
  - **Full advantage of first-principle calculations**
  - **Applicable to more complex structures**
- **Created SQS for BCC, HCP, B2, Laves phases, halite, and perovskite**



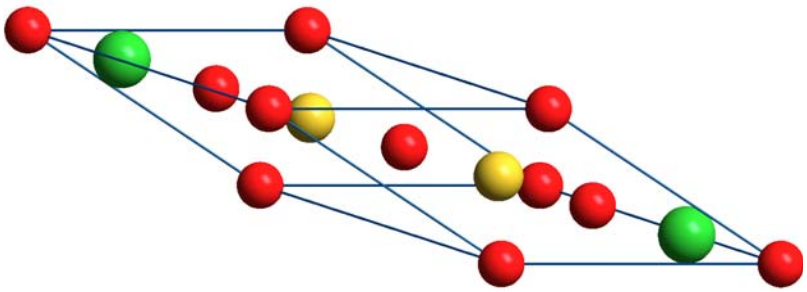
SQS-16 for bcc $A_{0.5}B_{0.5}$



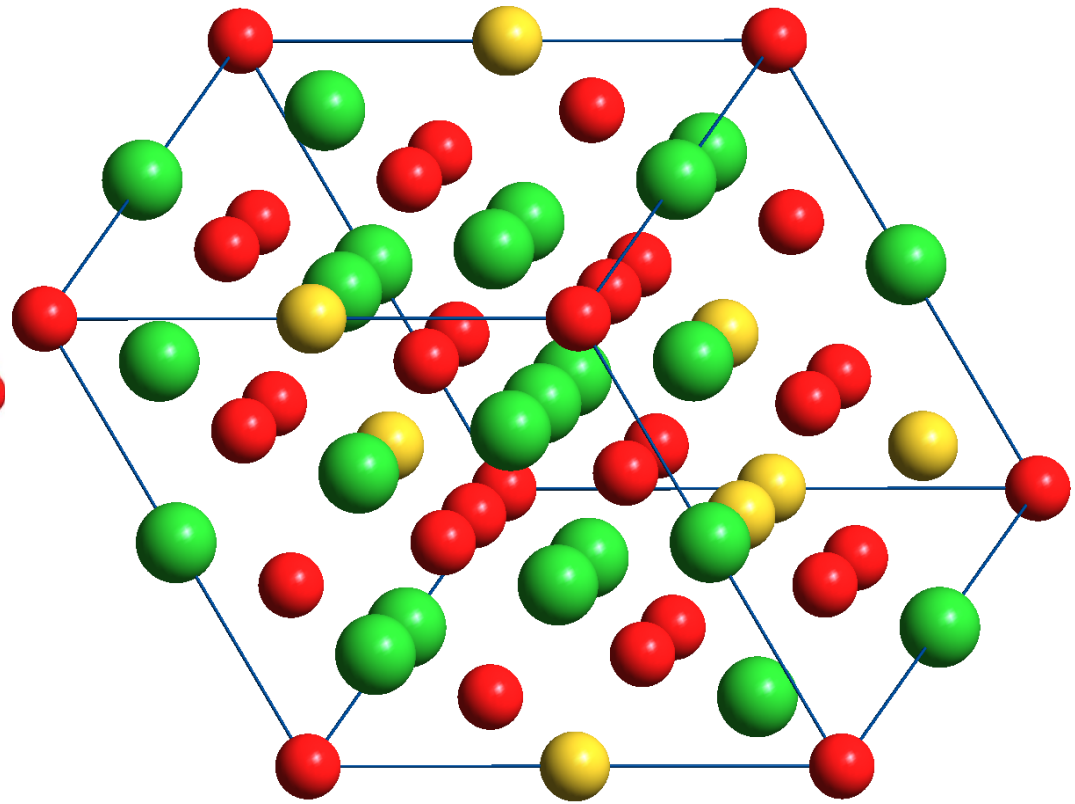
SQS-16 for bcc $A_{0.25}B_{0.75}$

C. Jiang, C. Wolverton, J. Sofo, L. Q. Chen and Z. K. Liu, *Phys. Rev. B,* **69**, (2004) 214202.

SQS for B2



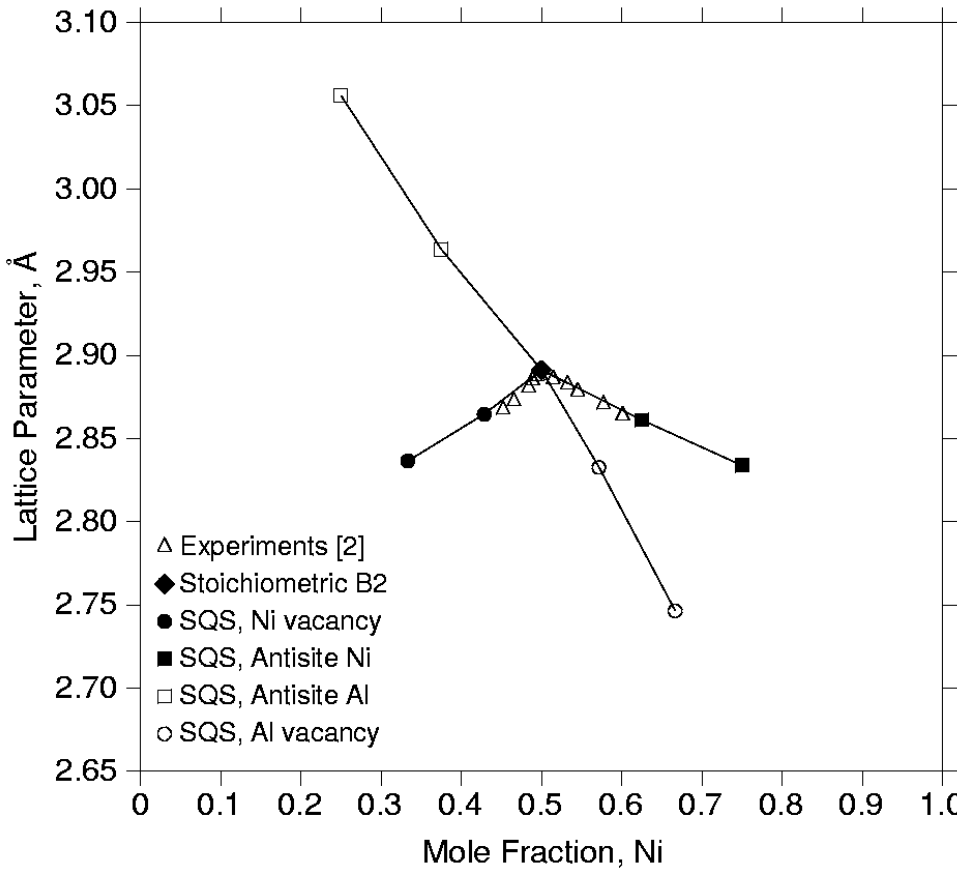
SQS-4 for $(A_{0.5}\ B_{0.5})C$

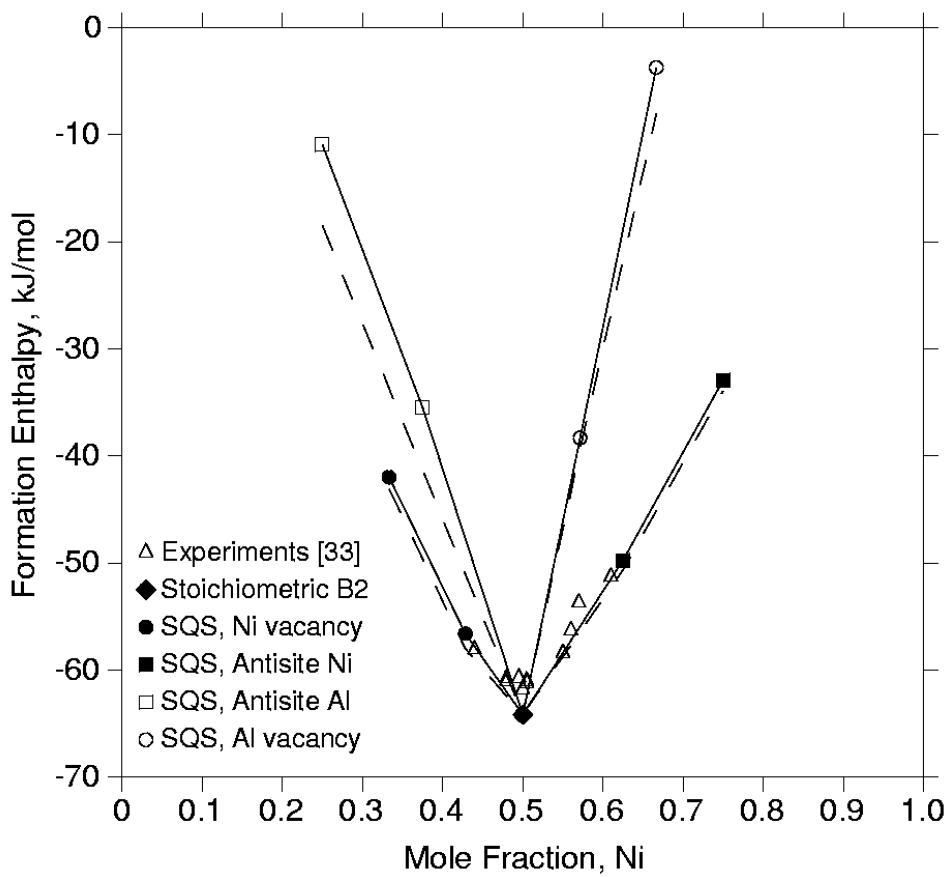


SQS-16 for $(A_{0.75}\ B_{0.25})C$

Chao Jiang, Long-Qing Chen and Zi-Kui Liu: Acta Mater. in press

B2 NiAl: (Al,Ni,Va)(Al,Ni,Va)



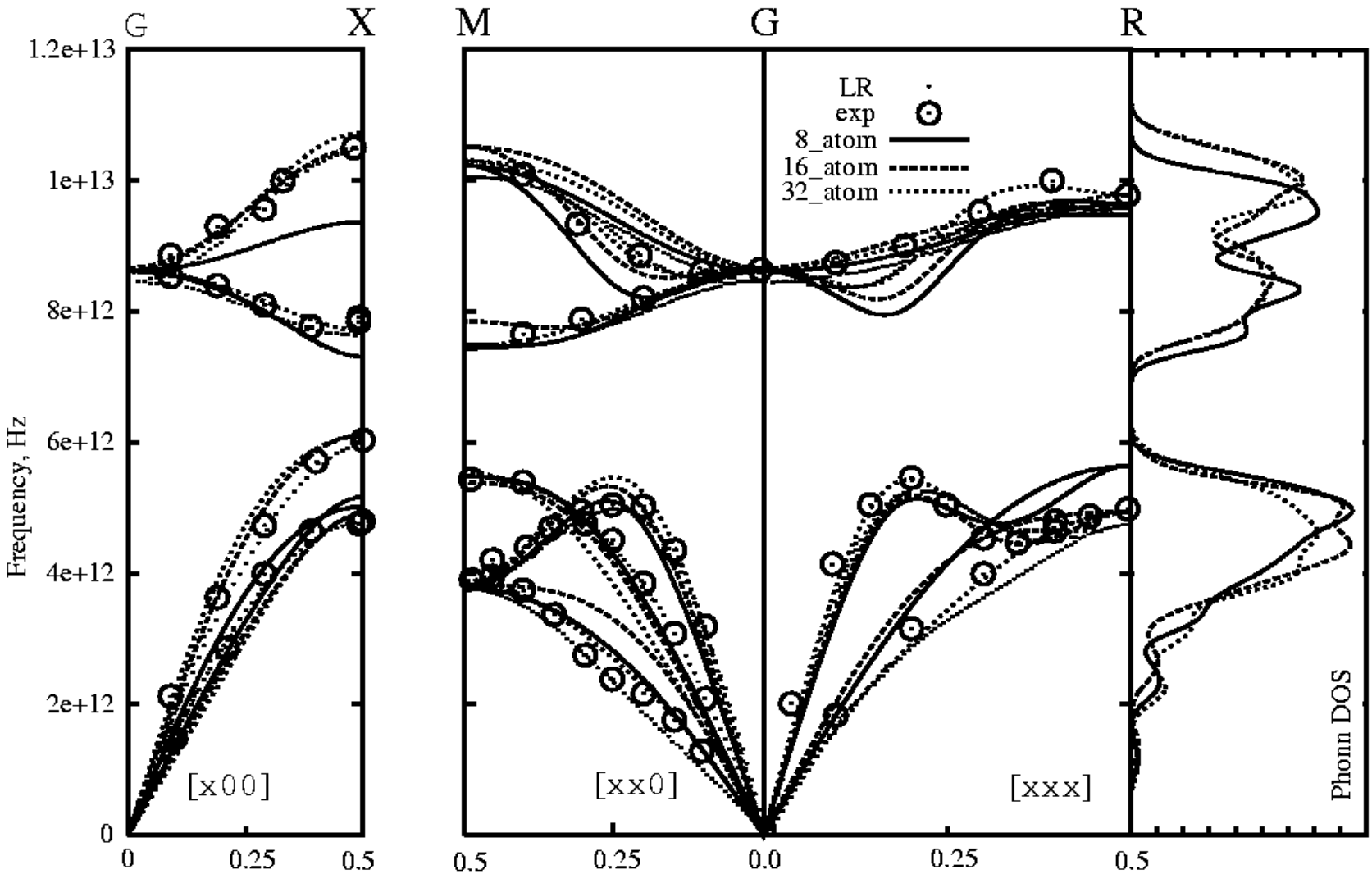


Vibrational Entropy

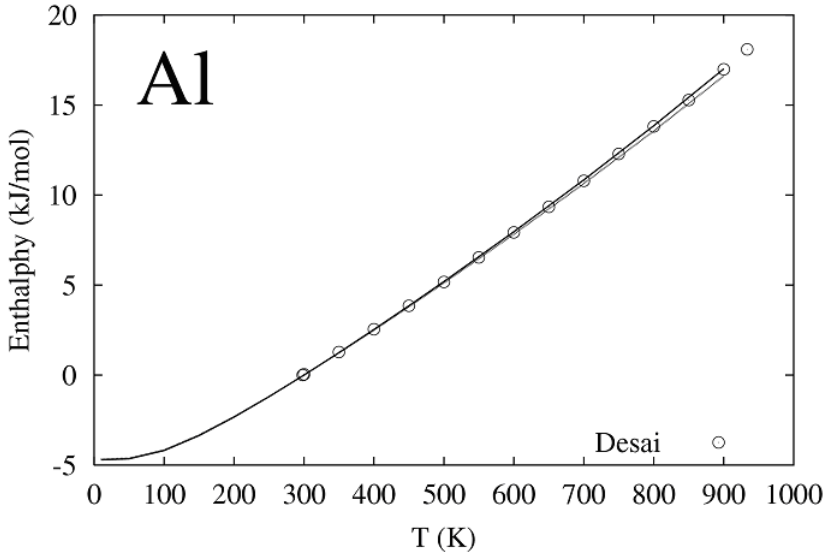
$$F(V,T) = E_c(V) + F_{ph}(V,T) + F_{el}(V,T)$$

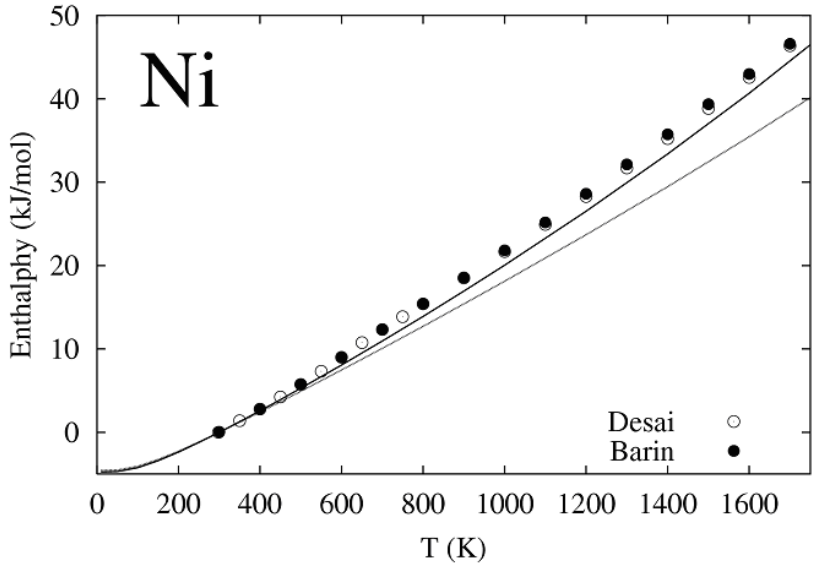
- **Linear response theory**
  - **Evaluate the force constants by calculating the second derivatives of the energy with respect to atomic displacements.**
  - **Y. Wang, Z. K. Liu and L. Q. Chen, "Thermodynamic properties of Al, Ni, NiAl, and Ni3Al from first-principles calculations," Acta Mater., Vol.52, 2004, 2665-2671.**
- **Frozen phonon method**
  - **Perturb the positions of the atoms from their equilibrium positions and calculating the resulting forces.**
  - **Use ATAT**
  - **R. Arroyave, D. Shin and Z. K. Liu, "Ab initio thermodynamic properties of stoichiometric phases in the Ni-Al system", Acta Mater., Vol.53, 2005, pp. 1809-1819.**

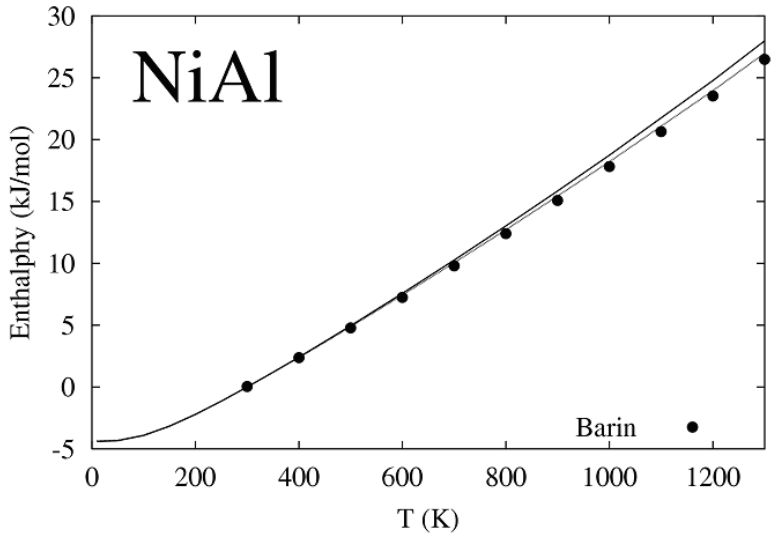
Phonon: NiAl

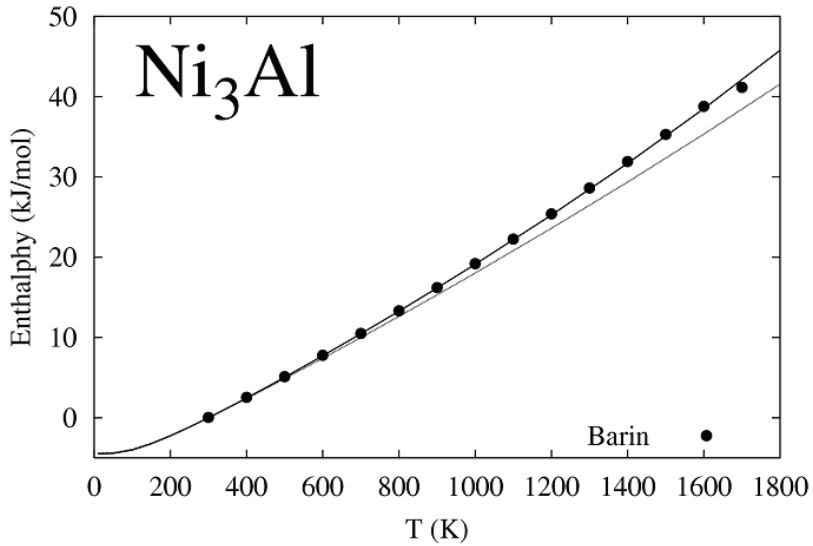

G
X
M
G
R
1.2e+13
1e+13
8e+12
6e+12
4e+12
2e+12
0
Frequency, Hz
LR
exp
8_atom
16_atom
32_atom
[x00]
[xx0]
[xxx]
Phonn DOS
0
0.25
0.5
0.5
0.25
0.0
0.25
0.5

Enthalpy


Al
Enthalpy (kJ/mol)
T (K)
Desai
20
15
10
5
0
-5
0
100
200
300
400
500
600
700
800
900
1000


Ni
Enthalpy (kJ/mol)
T (K)
Desai
Barin
50
40
30
20
10
0
0
200
400
600
800
1000
1200
1400
1600


NiAl
Enthalpy (kJ/mol)
T (K)
Barin
30
25
20
15
10
5
0
-5
0
200
400
600
800
1000
1200


Ni3Al
Enthalpy (kJ/mol)
T (K)
Barin
50
40
30
20
10
0
0
200
400
600
800
1000
1200
1400
1600
1800

Experimental Data

PaulingFile - Microsoft Internet Explorer

File Edit View Favorites Tools Help

Back

Address http://crystdb.nims.go.jp/

Go Links

NIMS HomePage | MITS HomePage | NIMS Database | News | Staff | Link

PaulingFile

Basic Database for Crystal Structures

since Apr 1 2003
0084601

MiTS

Login
Registration
Password Inquire
Modification
Secessione

- Outline
- What's New
- Database Editors
- Requirement System
- Notes on use
- Inquire
- Privacy Policy
- TopPage

Outline

The PAULING FILE project is a collaboration between Japan Science and Technology Corporation (JST) and Material Phases Data System (MPDS). The project started 1995. National Institute for Materials Science (NIMS) obtains the right to provide the online service of this system. The Pauling File aims at a comprehensive materials database which covers all non-organic solid state materials and consists of structure, diffraction, constitution, and physical property data. It is named by the name of the famous chemist Linus C. Pauling, who gave his permission to use his name in 1993.

The source of Pauling File data are around 150,000 original publications taken from more than 1,000 scientific journals since 1900. The data are processed by an international, highly-experienced group of scientists with going through a sophisticated data evaluation, standardization and derivation procedure.

The as-published data are accompanied by value-added information, such as calculated powder patterns (LAZY PULVERIX) and fully standardized structure data (STRUCTURE TIDY).

Online Pauling File

The present Pauling File includes about 80,000 structure entries, 34,000 diffraction entries, about 52,000 property data counts, about 6,000 constitution entries and 6,000 images of phase diagram.

This web-based system disseminates these four groups of data with the dynamic link between them, also provides a design platform as a tool for data mining and materials design.

	Multinary	Binary
Structure entries	80,000	28,000
Diffraction entries	34,000	28,000
Property data counts	52,000	42,000
Constitution entries	6,000	6,000
Images of phase diagram	6,000	6,000

Note: "Multinary" is inclusive of "Binary".

Done

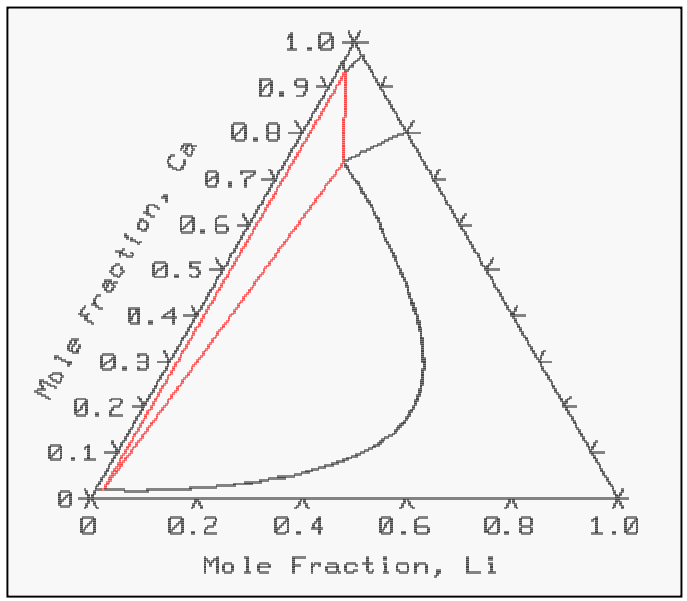
start | Endnote | 4 Microsoft PowerP... | EndNote 6 | PaulingFile -

New Algorithm

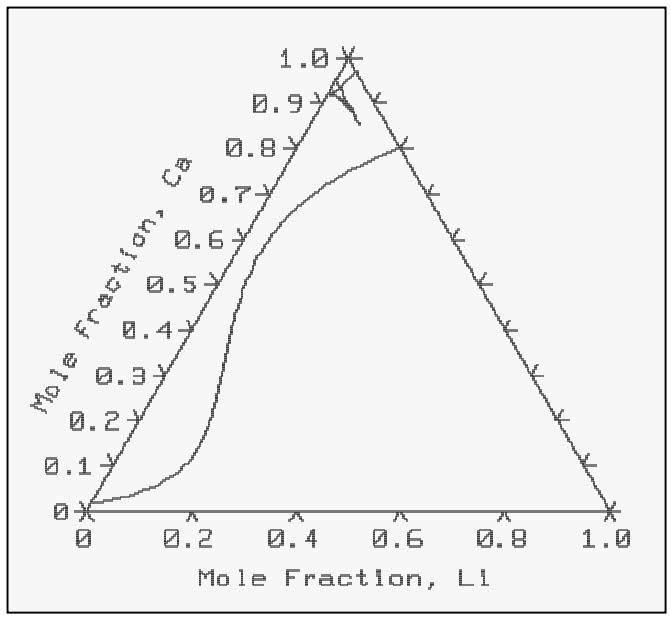
- **Iterative software drawbacks**
  - ✘ (user-dependent) **Use of prior knowledge of the system to generate a suitable starting point**
  - ✘ (unstable) **Possible divergence or convergence to metastable equilibria**

Ca-Li-Na system at T=900K

Correct diagram
Miscibility gap is specified



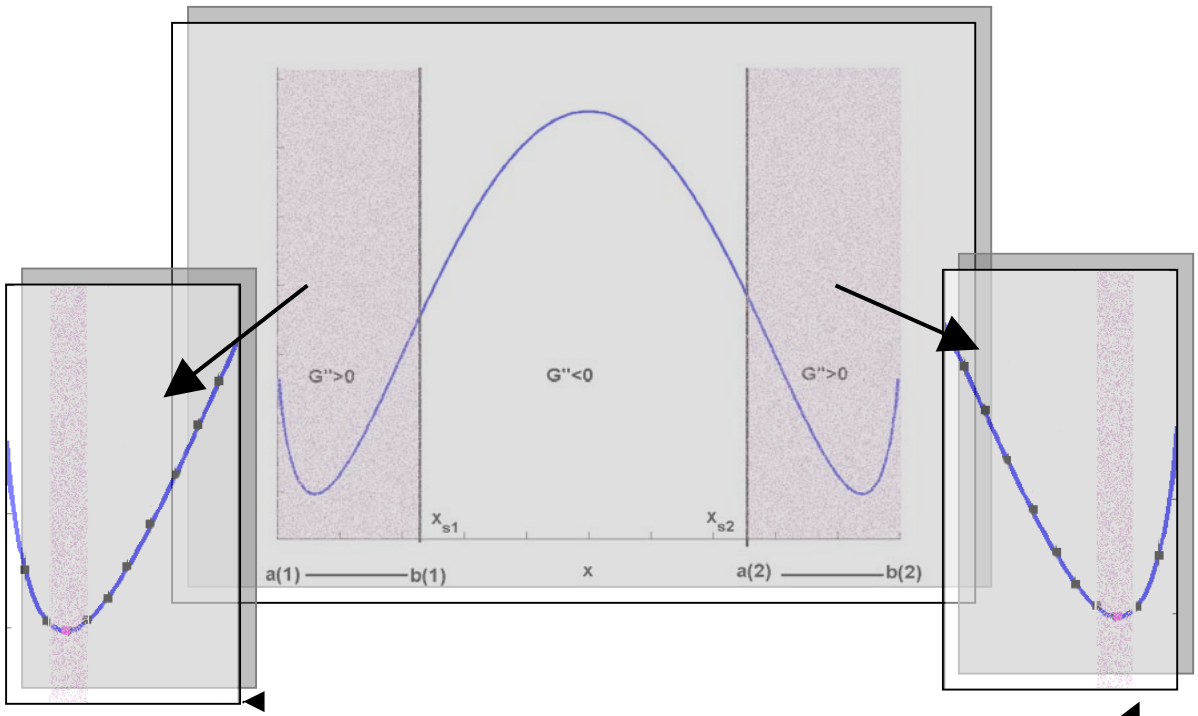
Miscibility gap is not specified



New algorithm: binary case

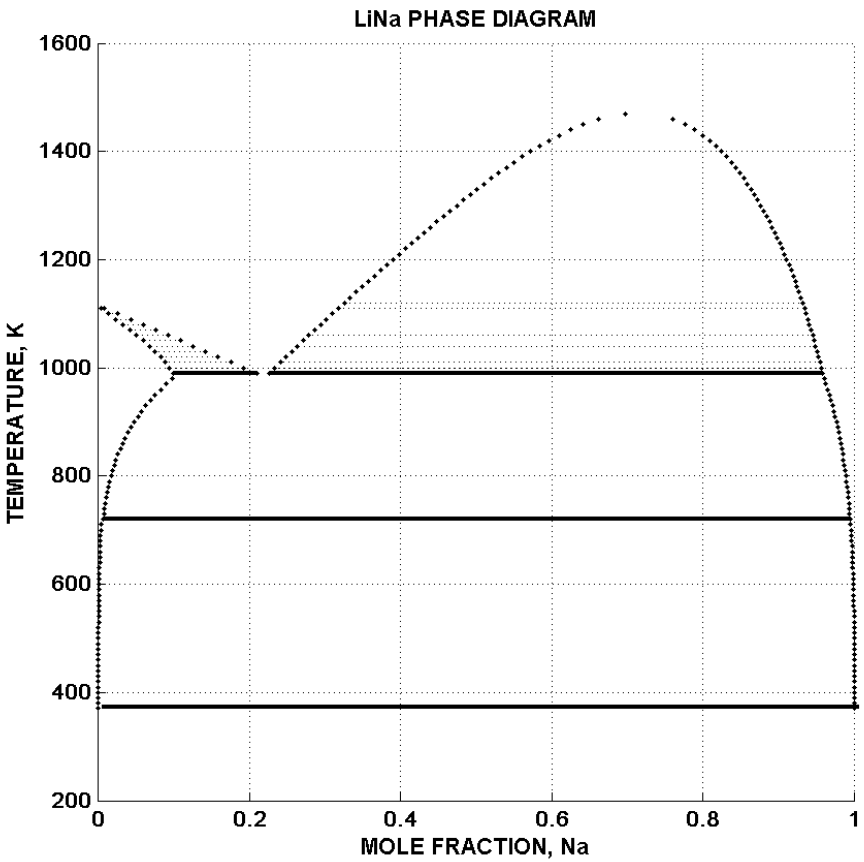
Schematic view of the adaptive refinement

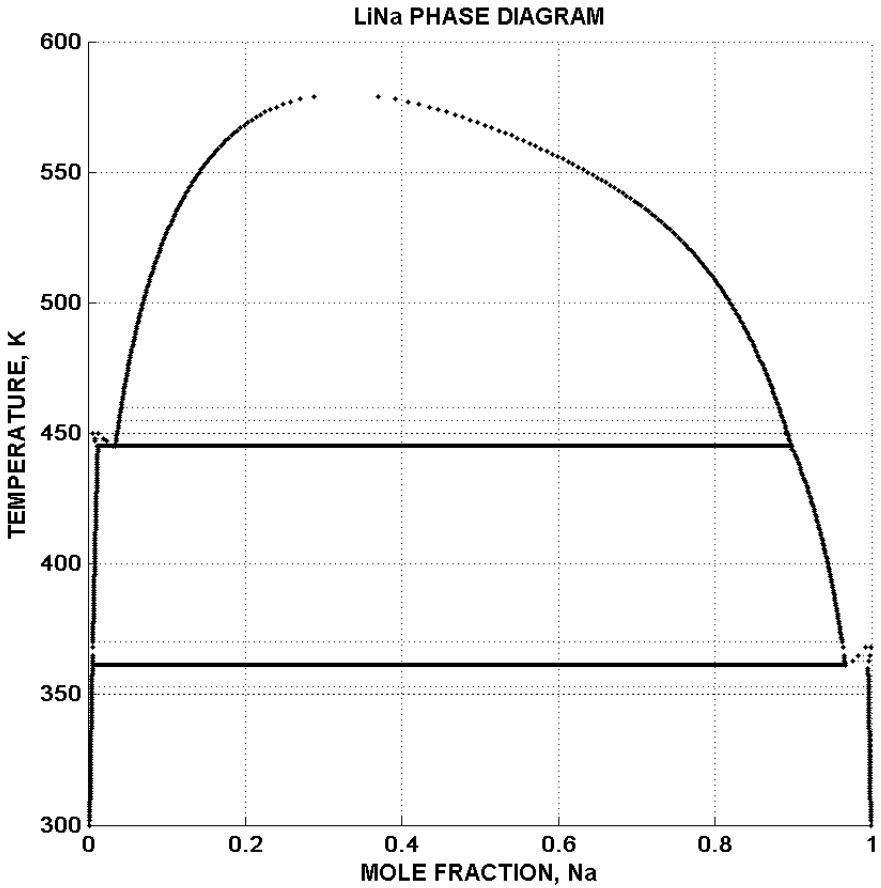
Continue until some stopping criterion is met (tolerance or maxIter is reached)



Sample N points after each refinement and locate lowest derivative value

Ca-Na and Li-Na Systems

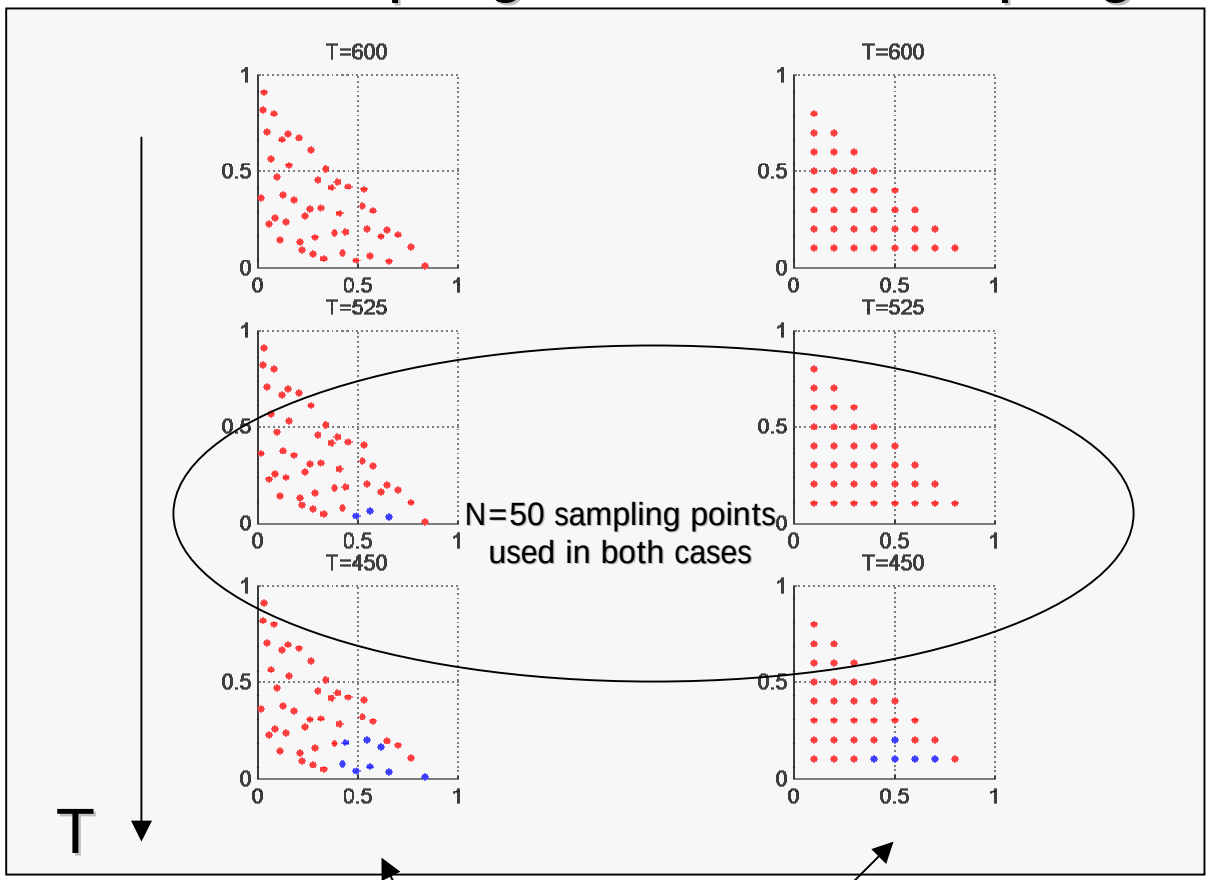




New algorithm: ternary case

Sobol sampling vs. uniform sampling

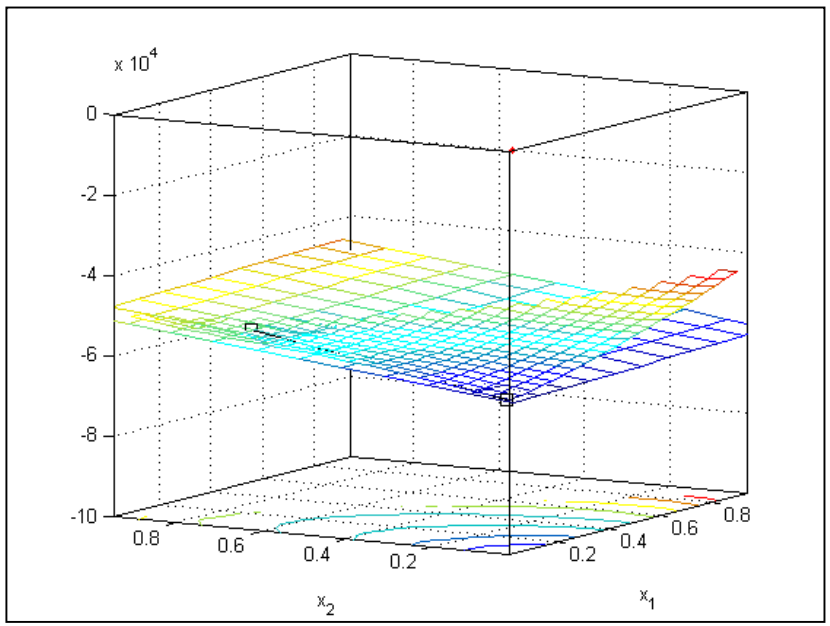
Miscibility gap detection in the CaLiNa system

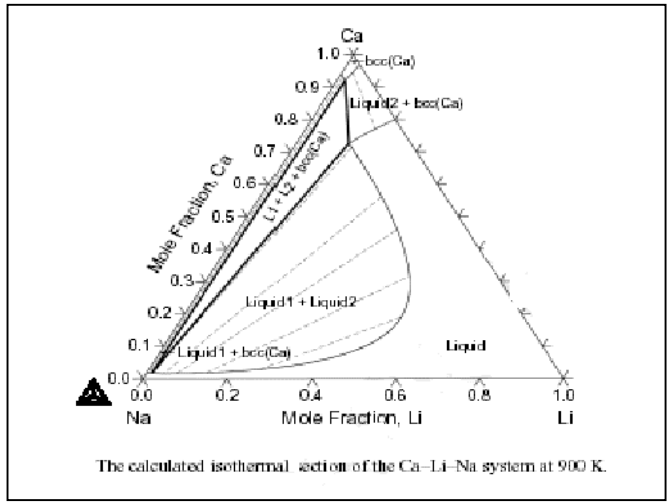


Numerical examples

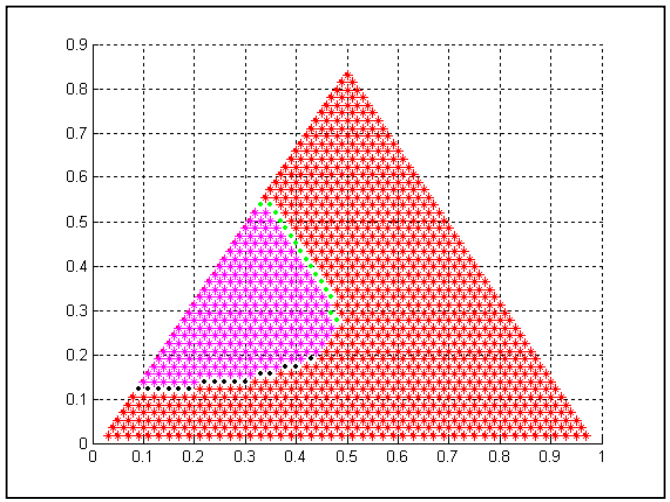
Ternary Ca-Li-Na system at T=900K

Gibbs energy profile with the miscibility gap and a corresponding common tangent plane

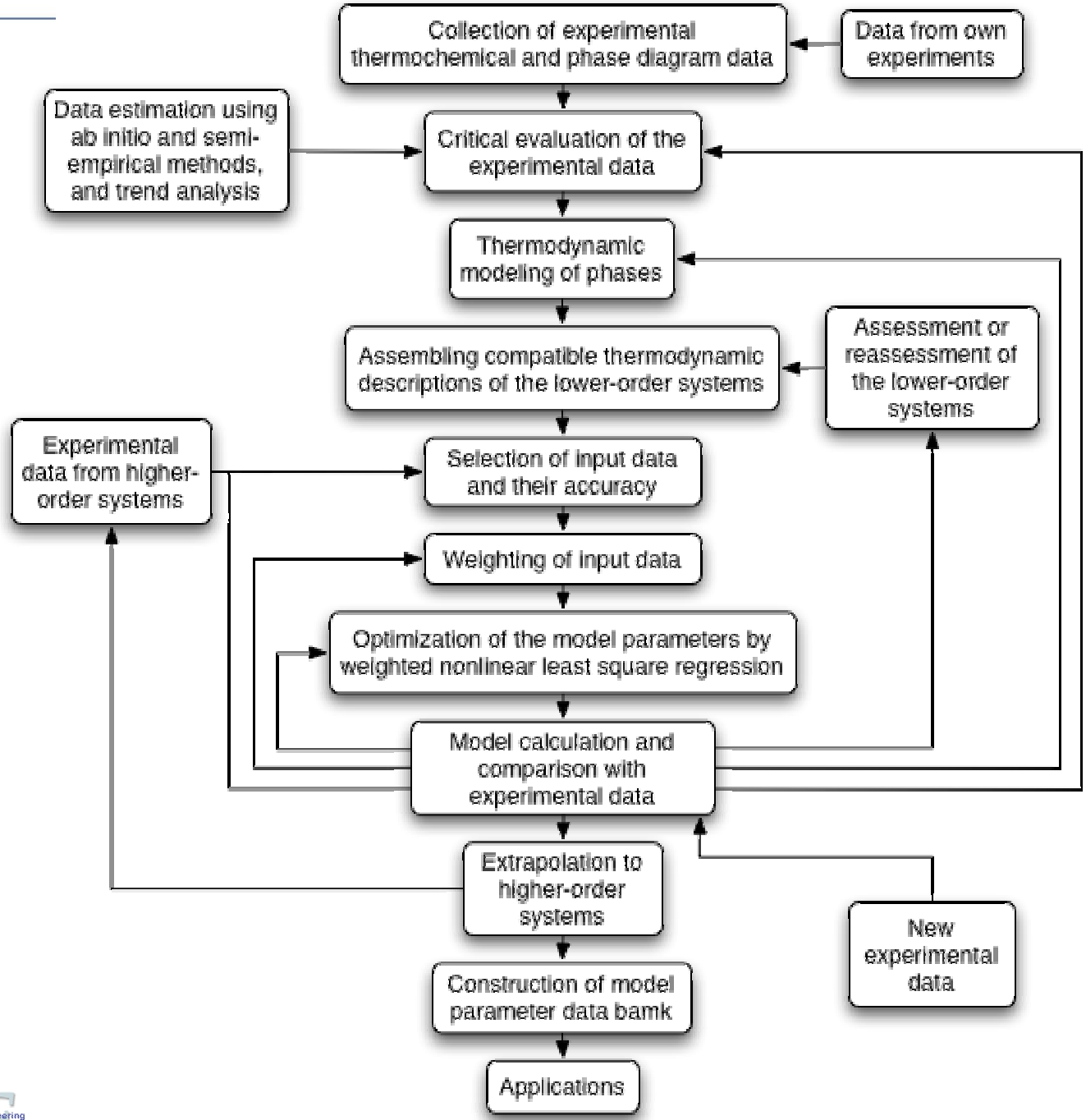




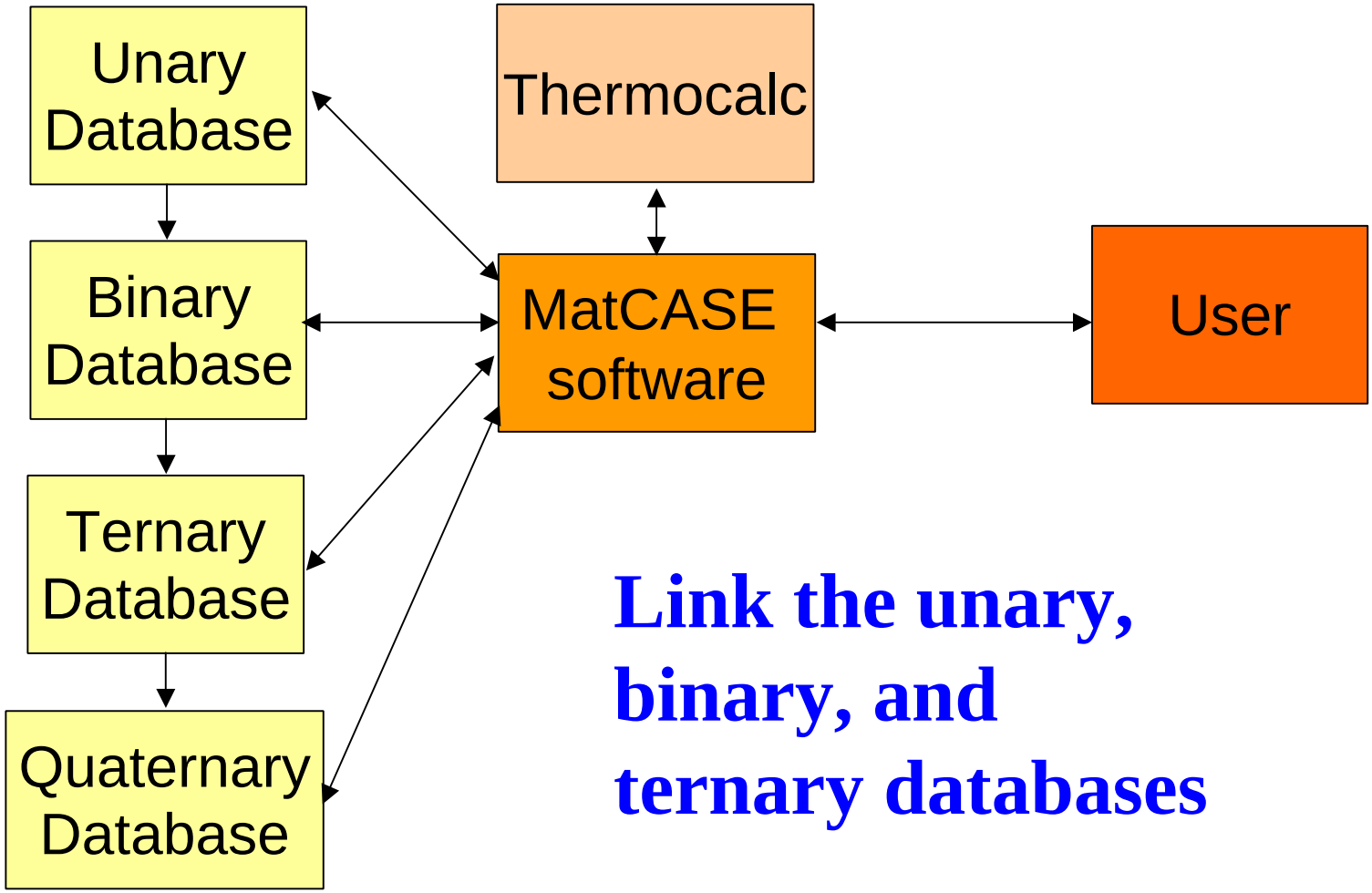
The calculated isothermal section of the Ca–Li–Na system at 900 K.



M.G. Emelianenko, Z.K. Liu, Q. Du, Computational Materials Science, 2005, in press

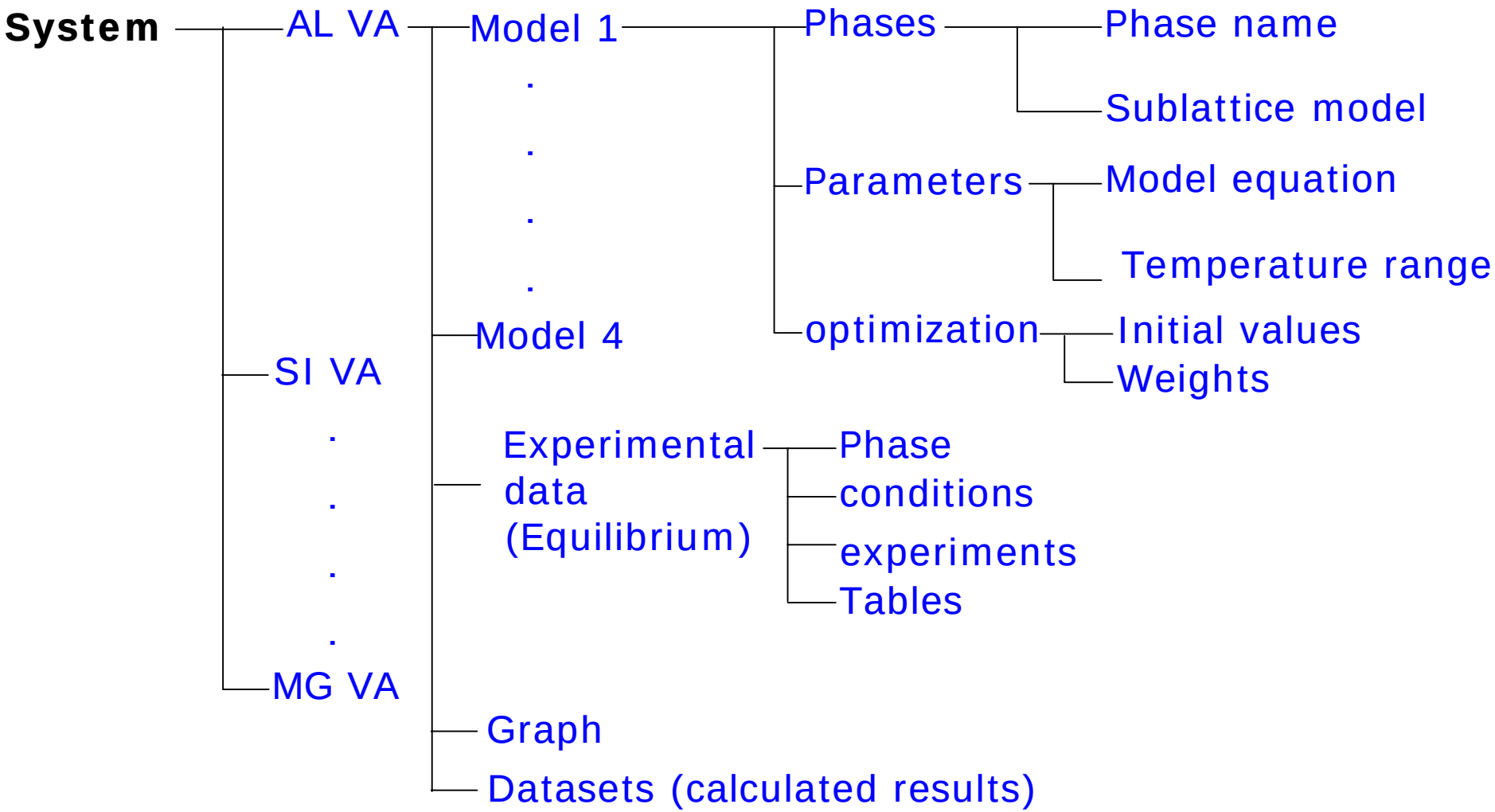

Collection of experimental thermochemical and phase diagram data
Data from own experiments
Data estimation using ab initio and semi-empirical methods, and trend analysis
Critical evaluation of the experimental data
Thermodynamic modeling of phases
Assembling compatible thermodynamic descriptions of the lower-order systems
Assessment or reassessment of the lower-order systems
Experimental data from higher-order systems
Selection of input data and their accuracy
Weighting of input data
Optimization of the model parameters by weighted nonlinear least square regression
Model calculation and comparison with experimental data
Extrapolation to higher-order systems
New experimental data
Construction of model parameter data bamk
Applications

Modeling Automation

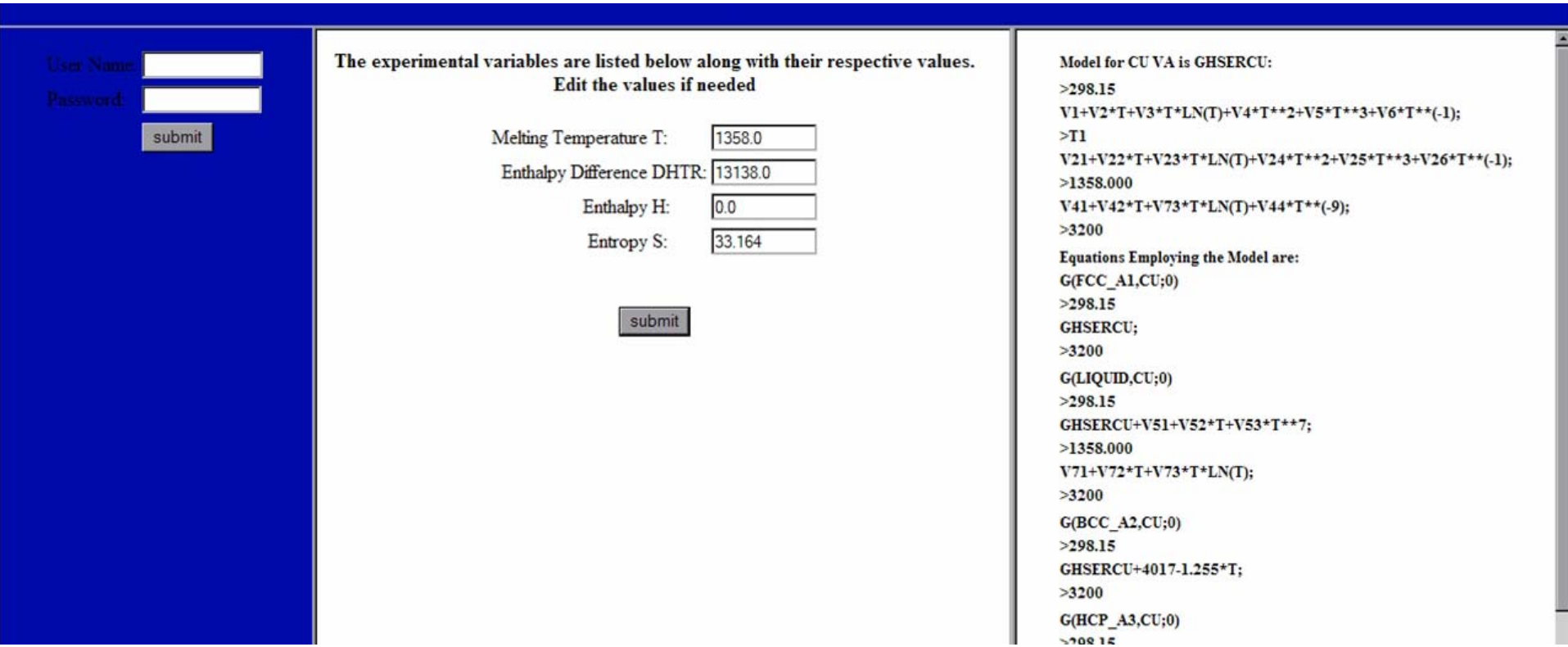


Link the unary, binary, and ternary databases

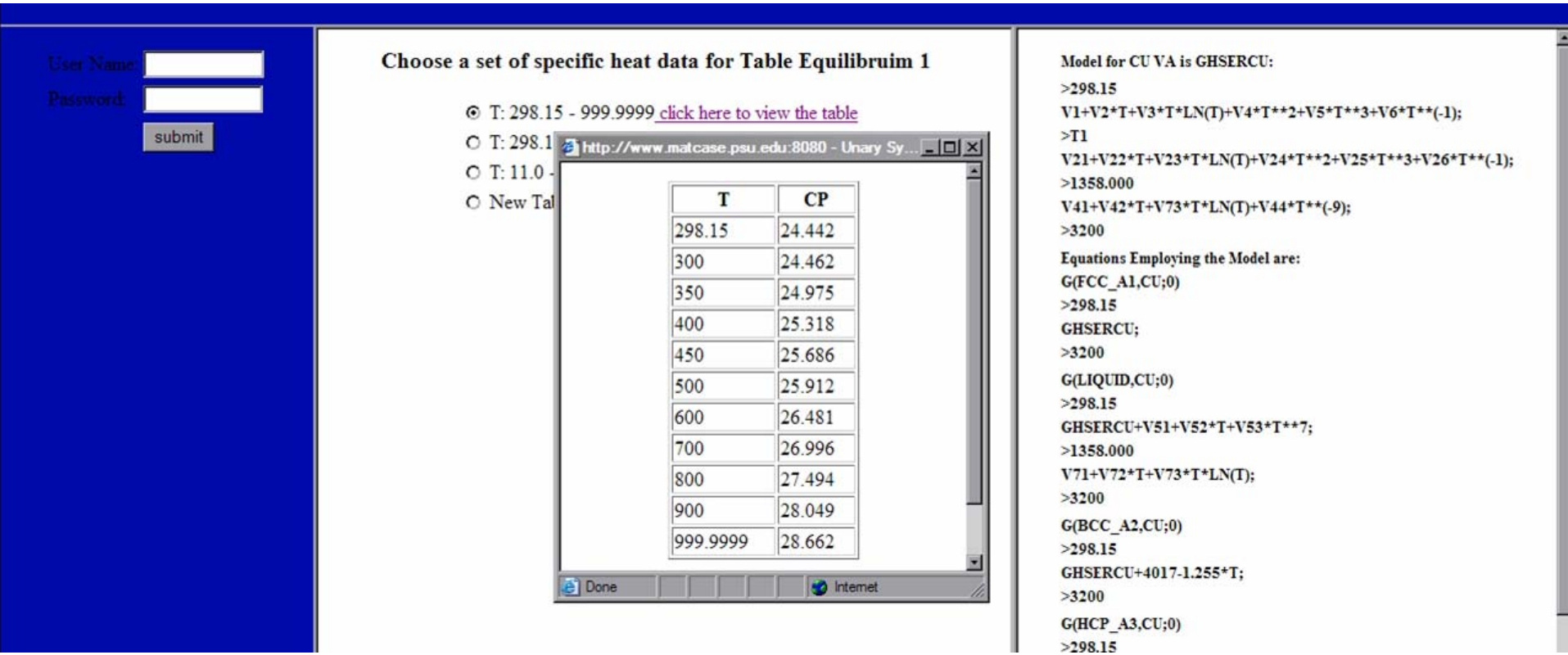
XML: Unary System



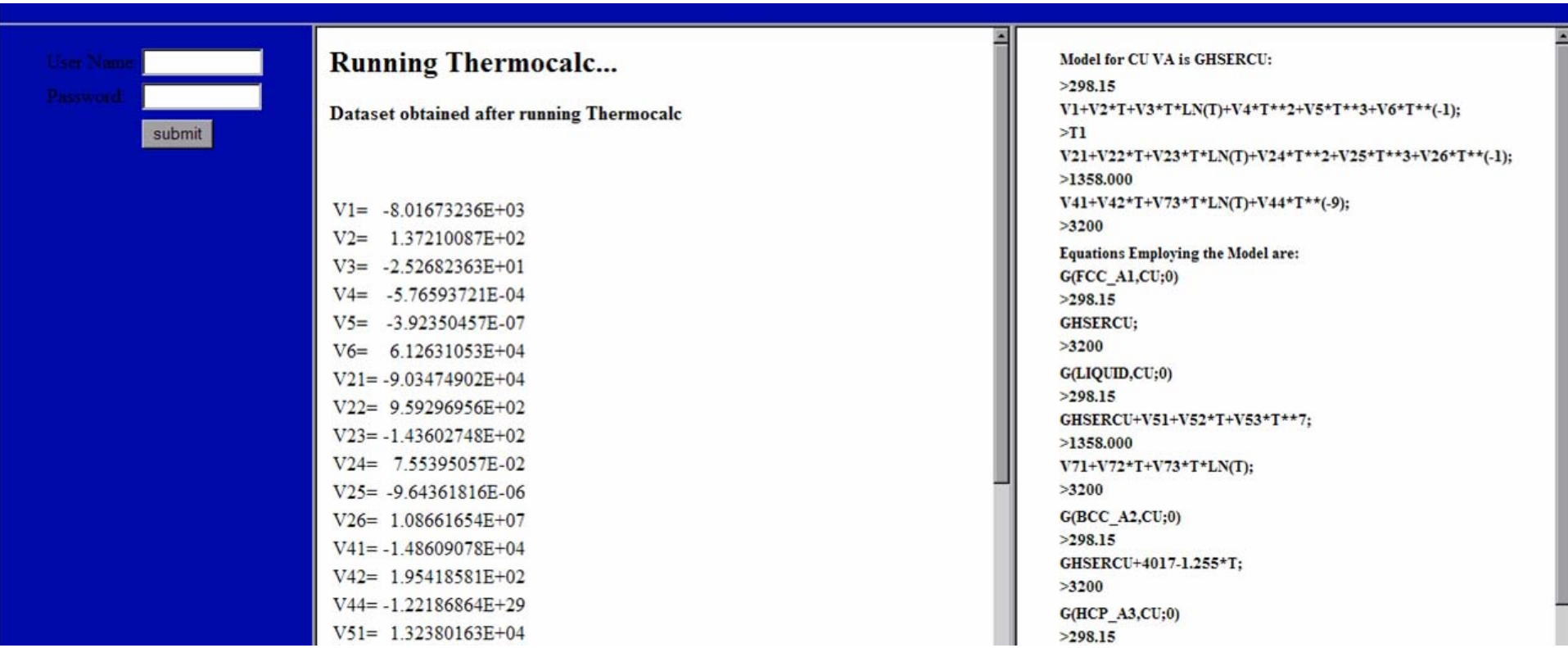
Screen Shot of Unary System



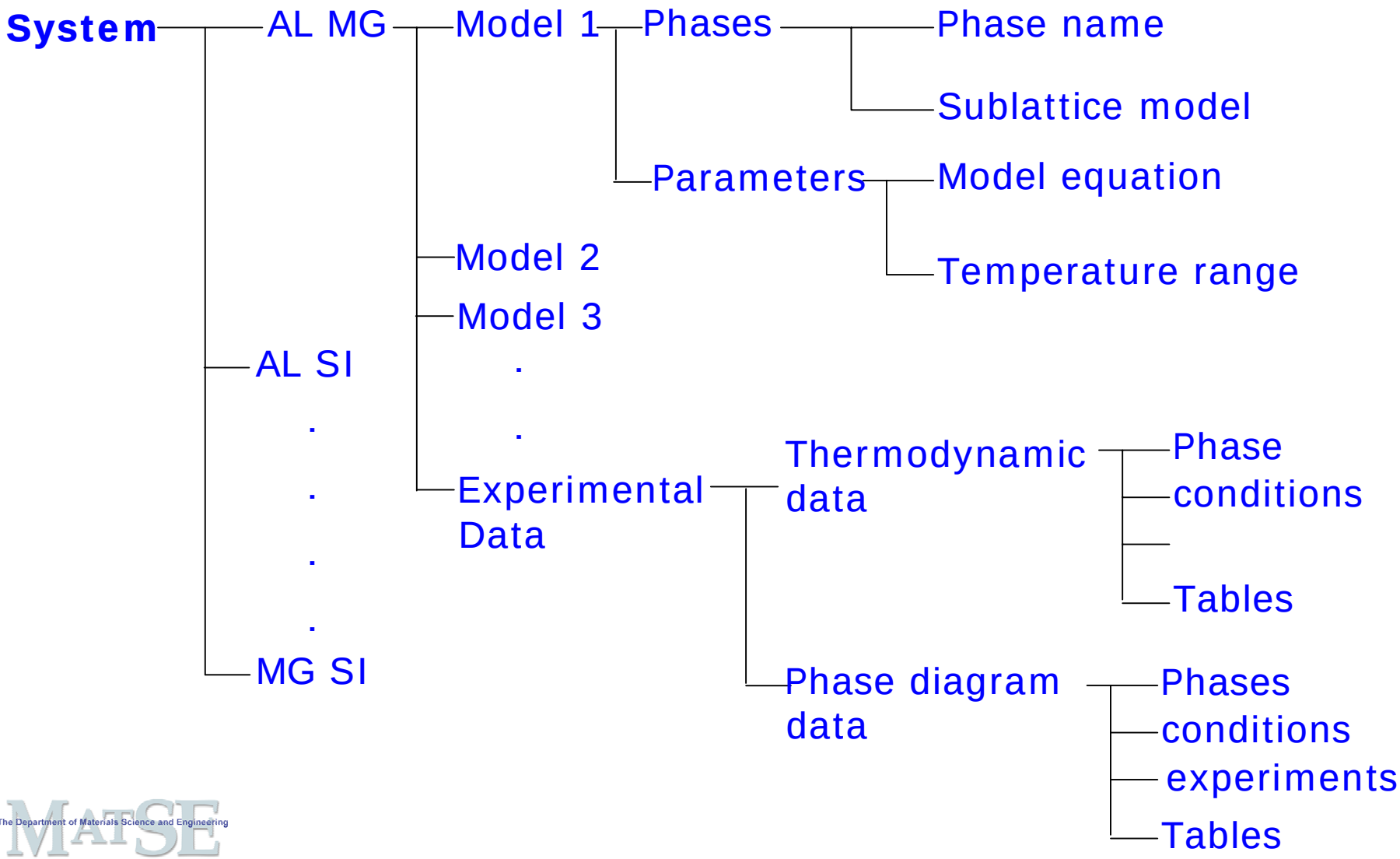
Screen Shot of Unary System



Screen Shot of Unary System

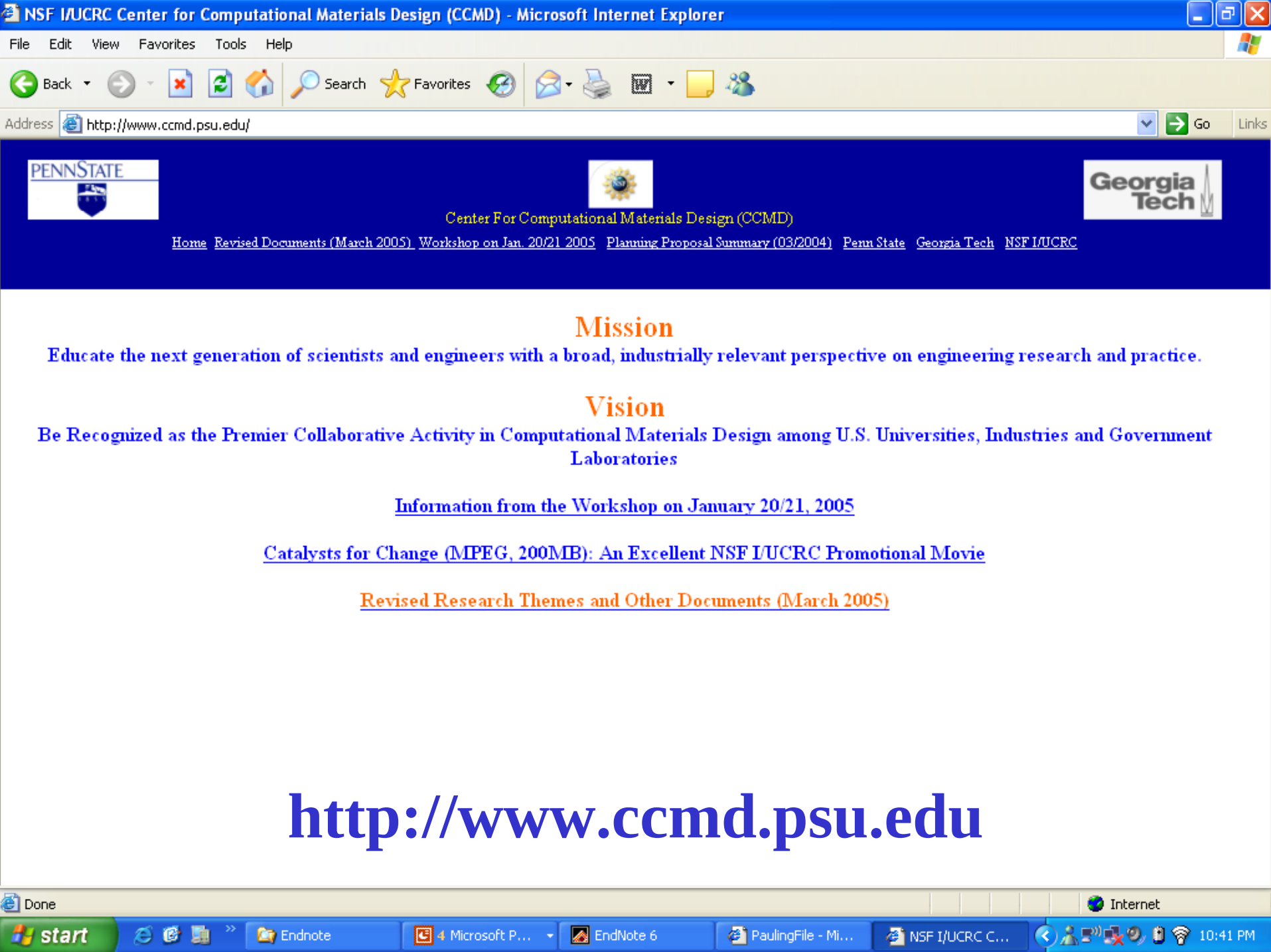


XML: Binary Systems



Summary

- First-principles calculations efficiently provide enthalpy of formation of stable stoichiometric compounds.
- Entropy of formation and entropy of mixing can be calculated with various approximations.
- New algorithm for robust phase equilibrium calculations is developed.
- The automation of thermodynamic modeling is being constructed.
- The automation will be extended to modeling of mobility and other properties.


NSF I/UCRC Center for Computational Materials Design (CCMD) - Microsoft Internet Explorer
File Edit View Favorites Tools Help
Back Search Favorites
Address http://www.ccmd.psu.edu/
Go Links
PENNSTATE
Georgia Tech
Center For Computational Materials Design (CCMD)
Home Revised Documents (March 2005) Workshop on Jan. 20/21 2005 Planning Proposal Summary (03/2004) Penn State Georgia Tech NSF I/UCRC
Mission
Educate the next generation of scientists and engineers with a broad, industrially relevant perspective on engineering research and practice.
Vision
Be Recognized as the Premier Collaborative Activity in Computational Materials Design among U.S. Universities, Industries and Government Laboratories
Information from the Workshop on January 20/21, 2005
Catalysts for Change (MPEG, 200MB): An Excellent NSF I/UCRC Promotional Movie
Revised Research Themes and Other Documents (March 2005)
http://www.ccmd.psu.edu
Done
Internet
start
Endnote
4 Microsoft P...
EndNote 6
PaulingFile - Mi...
NSF I/UCRC C...
10:41 PM