Large Scale Screening of Low cost Ferritic Steel Designs for Advanced Ultra-SuperCritical Boiler Using First Principles Methods

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Outline

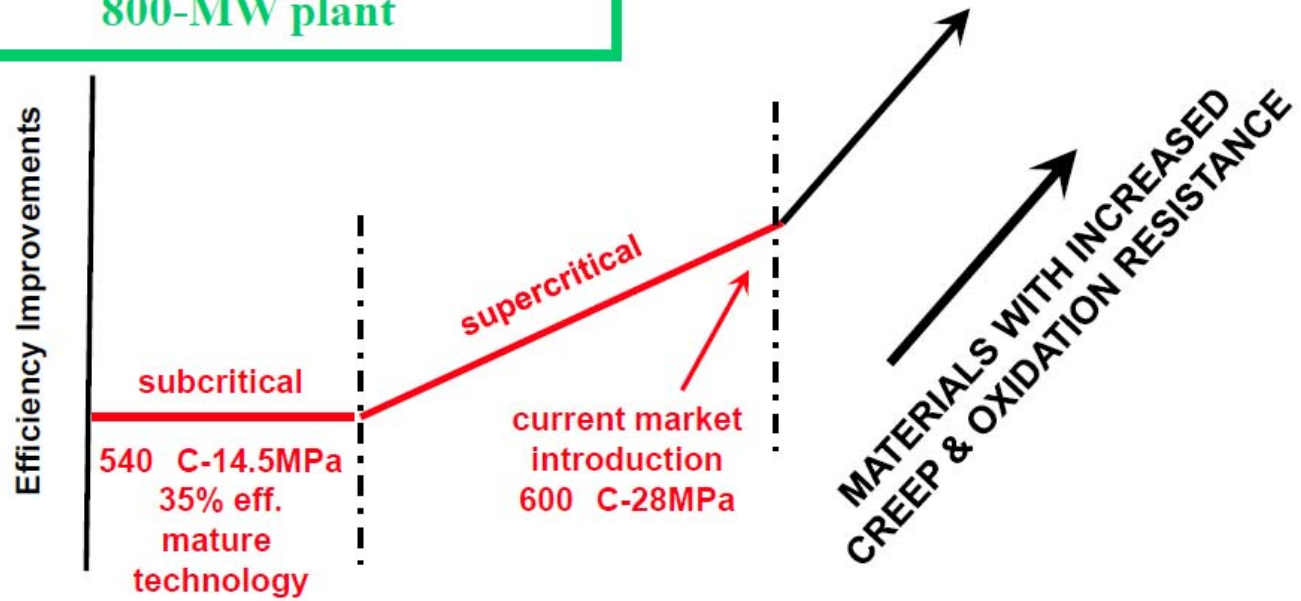
- Background
- Goal
- Approach
- Project status

Background

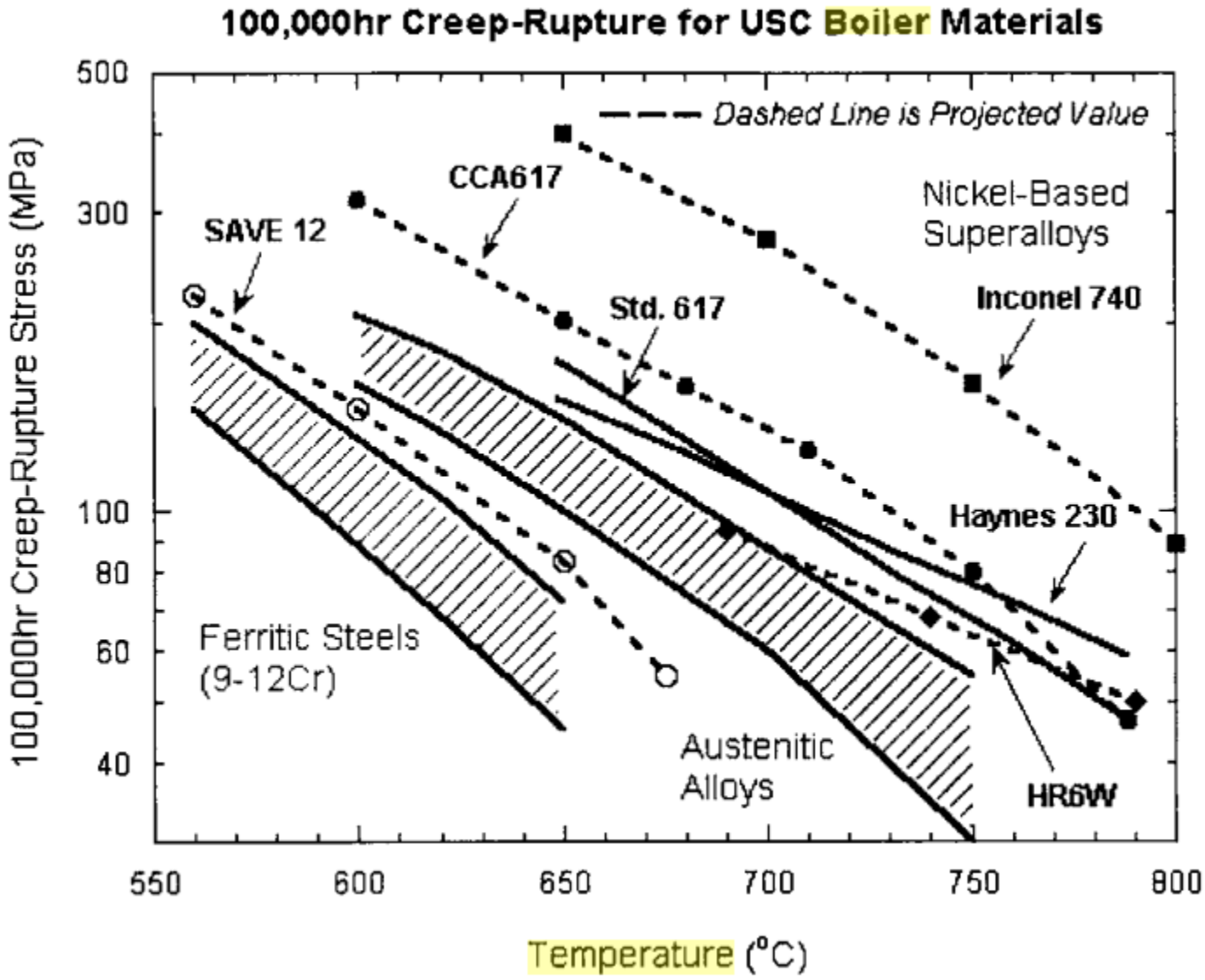
Increasing Efficiency

Each 1% increase in efficiency eliminates ~1,000,000 tons of CO_2 emissions over the lifetime of an 800-MW plant

US-DOE Advanced Power Systems:
46%-48% efficiency from coal generation
Steam condition: 760 C - 35MPa
~ 5ksi



Background


100,000hr Creep-Rupture for USC Boiler Materials
Dashed Line is Projected Value
CCA617
SAVE 12
Std. 617
Nickel-Based Superalloys
Inconel 740
Haynes 230
HR6W
Ferritic Steels (9-12Cr)
Austenitic Alloys
100,000hr Creep-Rupture Stress (MPa)
500
300
100
80
60
40
550
600
650
700
750
800
Temperature (°C)

Background

- Material requirements for Advanced Ultra-SuperCritical (AUSC) boiler:
  - ***Low cost*** metal
  - Compatible thermal properties
  - Sufficient high temperature performance
    - Sufficient mechanical strength
    - Creeping resistance
    - Corrosion resistance
  - *Low temperature ductility*

Motivation

- Ferritic steel
  - BCC matrix with low Ni content (low cost)
  - Low temperature brittleness
  - Weldability
- New design of ferritic steel
  - Composition modulation
  - Microstructure engineering

Complexity of the Alloy Strategy

- Many structural factors:
  - **Matrix**
  - Precipitation
  - Grain boundary
  - Interphase
- Correlated problem
  - Doping may solve a problem but bring in more problems
  - Difficult to assess the effect of alloying

Ferritic Matrix

- BCC structure
- Composition of the BCC matrix in 9-12% Cr steels

Name	Fe	Cr	Ni	Mo	Si	Al
P91	0.9898	5.87E-4		8.42E-9	6.64E-3	2.77E-3
E911	0.9969	5.29E-4		1.13E-8	2.03E-3	5.75E-12
P92	0.9944	5.55E-4	6.10E-4	2.30E-8	3.83E-3	6.14E-4
AXM	0.9964	5.45E-4	5.96E-4	1.97E-7	1.31E-3	1.15E-3
HCM12	0.9977	5.36E-4	1.03E-5	1.19E-8	1.72E-3	
P122	0.9986	5.15E-4	6.22E-4	1.08E-11	2.99E-4	4.18E-11

Objectives

- Screening ferritic steel design based on properties of the solid solution matrix
  - Elastic properties
  - Low temperature ductility (Rice-Thompson parameter)

- Develop efficient parallel software for large scale screening calculations
  - first principles quality for solid solution system
  - Automated solid solution structure sampling
  - Automated properties calculations

Approach: Structure Modeling

- Dilute multi-component solid solution
  - requires huge lattice cell
  - components of very low concentration considered as point defect
- Special quasi-random structures

Structure → Properties

- Classification of discrete structure models
  - Atomistic point model
    - Gas: position of atom all over the space
    - Crystal: atom positions confined to the regular lattice sites
    - **Ordered solid solution**: random occupation on regular lattice sites
    - Disordered : atom positions confined to the irregular lattice sites
  - Multipole grain model

Gas: Mayer cluster expansion

Statistical Mechanics / *Classical N same particles gas in a box problem*:

Canonical partition function (boundary condition)

$$Z(\beta) \equiv \frac{1}{N!\hbar^{3N}} \int_{\mathcal{V}} d\mathcal{V} \mathrm{e}^{-\beta H(\mathbf{p},\mathbf{q})} \qquad H(\mathbf{p},\mathbf{q}) \equiv \frac{1}{2\mu} \sum_{j=1}^{3N} p_j^2 + U(\mathbf{q})$$

Therefore

$$Z(\beta) \equiv \frac{1}{N!} \left(\frac{2\pi\mu\alpha_2^2}{\hbar^2\beta} \right)^{\frac{3N}{2}} Z_{pos}(\beta) \qquad Z_{pos}(\beta) \equiv \frac{1}{\alpha_2^{3N}} \int_V \cdots \int_V dq_1 \cdots dq_{3N} \, \mathrm{e}^{-\beta U(\mathbf{q})}$$

Mayer cluster expansion to calculation configurational integral

Assuming only pair-wise interaction

$$\mathrm{e}^{-\beta U(\mathbf{q})} = \prod_{i=1}^{N-1} \prod_{j=i+1}^{N} \mathrm{e}^{-\beta U_{i,j}(r_{i,j})}$$

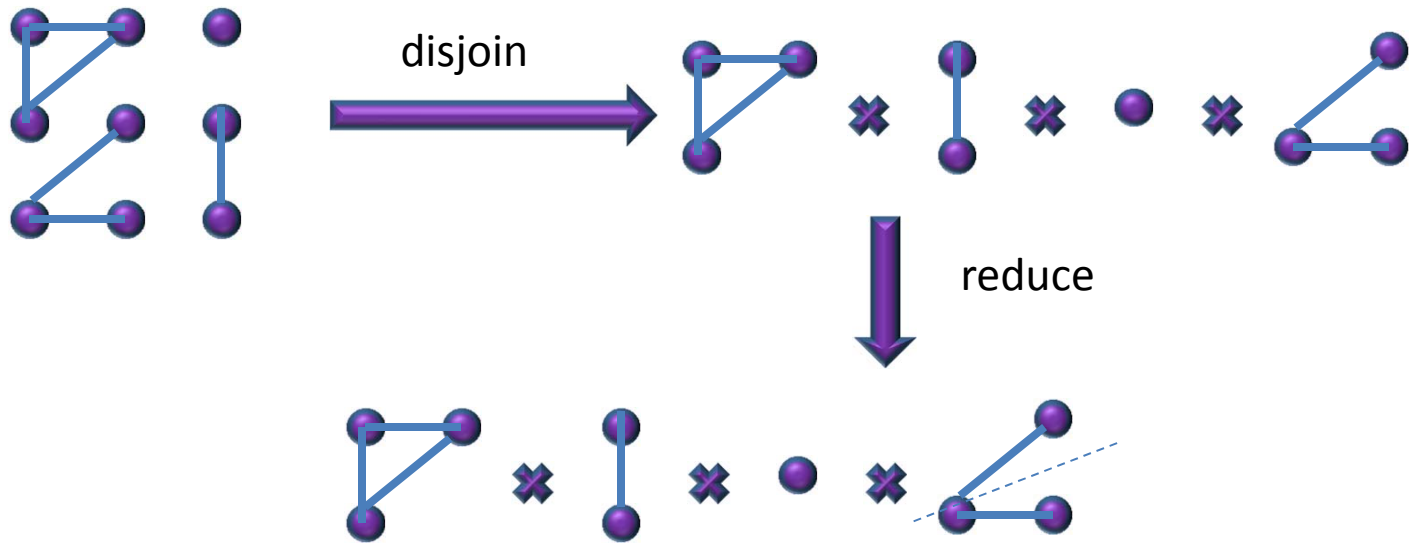
Introducing Mayer function $\mathcal{M}_{i,j} \equiv \mathrm{e}^{-\beta U_{i,j}(r_{i,j})} - 1$ (small)

$$\mathrm{e}^{-\beta U(\mathbf{q})} = \prod_{i=1}^{N-1} \prod_{j=i+1}^{N} (1 + \mathcal{M}_{i,j}) = 1 + \sum_{i=1, i<j}^{N} \mathcal{M}_{i,j} + \sum_{i=1, i<j, k=1, k<l}^{N} \mathcal{M}_{i,j}\mathcal{M}_{k,l} + \cdots$$

Irreducible Integrals

$$\int dV M_{ij} M_{kl} \ldots$$

Graphical representation of Mayer cluster expansion terms



Singly jointed graph can be reduced into product of irreducible integrals

Lattice: Solid Solution

- **Free energy calculations**
  - *Supercell Approaches*
    - *Ensemble Average of Supercells:* $F(\vec{\sigma},T) \approx \sum_{i \in \vec{\sigma}} w_i(T) F_i(T)$

      supercells are local snapshots in the infinite solid solution lattice $\vec{\sigma}$

  - *Cluster Expansion Methods*
    - *Weighted average of clusters:* $F(\vec{\sigma},T) \cong \sum_{\alpha,s} K_\alpha^s(T) \Phi_\alpha^s$

      α,s are cluster indices and cluster order indices
      clusters are local structures in the infinite solid solution lattice $\vec{\sigma}$
    - *Mathematically rigorous*

Lattice: Solid Solution

❑ Properties calculations

- ➢ *Supercell Approaches*
  - ➢ *Ensemble Average of Supercells:* $F(\vec{\sigma},T) \approx \sum_{i\in\vec{\sigma}} w_i(T)F_i(T)$

    supercells are local snapshots in the infinite solid solution lattice $\vec{\sigma}$

- ➢ *Cluster Expansion Methods*
  - ➢ *Weighted average of clusters:* $F(\vec{\sigma},T) \cong \sum_{\alpha,s} K_\alpha^s(T)\Phi_\alpha^s$

    α,s are cluster indices and cluster order indices
    clusters are local structures in the infinite solid solution lattice $\vec{\sigma}$
  - ➢ *Mathematically rigorous*

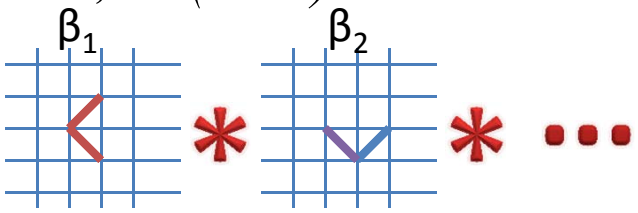
G(P,T) Module: UnitCell Expansion

Cluster Expansion Method for multi-component multi-sublattice systems:

P.D. Tepesch, et al PRL 74, 12 (1995)

$$F(\vec{\sigma},T) \cong \sum_{\alpha,s} K_{\alpha}^{s}(T)\Phi_{\alpha}^{s},$$

$$\Phi_{\alpha}^{s} = \prod_{i} \Phi_{\beta_i}^{s_i}; \alpha = \bigcup_{i} \beta_i$$

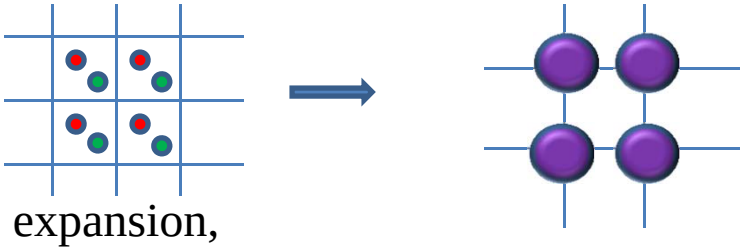


Challenge: number of cluster expansion terms n

$$n \sim (N-1)^{|\alpha| Nsublattice}$$

UnitCell Expansion Method for multi-component multi-sublattice systems:

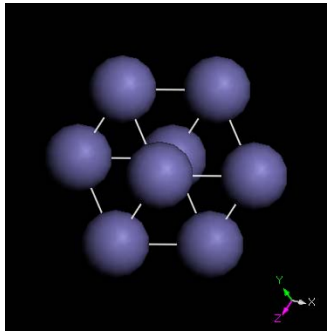
$$F(\vec{\sigma},T) \cong \sum_{\gamma,s} K_{\gamma}(T)\Phi_{\gamma}^{s},$$



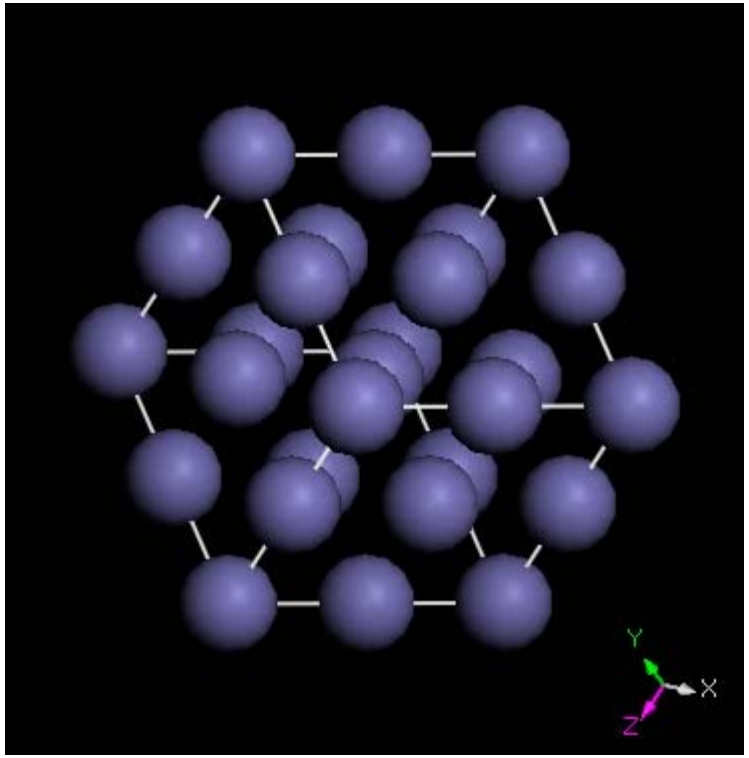
Rationale: Coarse grained cluster (CGC) expansion,

- Unitcells are treated as pseudo atom types
- Simplify lattice
- Expected must faster cluster interaction decaying over distance (*up to pair*)
- Much larger number of components (pseudo atoms) (*unitcell types*)

CGC



2x2x2



3x3x3

Grouping CGC

Reducing CGC set size:

- *CGC with the same composition can be grouped*
  - *Symmetrically equivalent CGC to be assigned the same CGC id*
  - *Cluster =* $< C_i , n_{Ci} , o_{Ci,n} , r_i >$

    C_i : *concentration vector at size cluster site i,*

    n_{Ci} : *order id of CGC with the same* C_i

    $o_{Ci,n}$: *orientation of CGC with order* n_{Ci} *and* C_i *at site i*

    r_i: *relative position of site i in the cluster*

  - *With proper choice of basis, it is expected that only a much reduced set of* n_{Ci} *and* $o_{Ci,n}$ *will have significant ECI*

Discrete Chebyshev Basis

- **Orthogonal discrete Chebyshev basis for Multi-components system**
  - $\boldsymbol{\sigma}=\{\sigma_1, \sigma_2, \ldots, \sigma_N\}$: N (number of lattice sites), σ_i (site occupation)
    $\sigma_i = \{-1, -(m-1)/m, \ldots, 1\}$: M (number of components) M=2m *or* 2m+1
  - *Discrete chebyshev polynomial* Θ_n *:*

$$\Theta_n(\sigma) = \sum_k c_{n,k}\sigma^k, \qquad \left\langle \Theta_n(\sigma), \Theta_m(\sigma) \right\rangle_{all\sigma} = \delta_{nm}$$

  - *Cluster function* Φ*:* $\Phi_\alpha^s = \prod_{\substack{s=\{n_1,\ldots\} \\ \alpha=\{p_1,\ldots\}}} \Theta_{n_i}(\sigma_{p_i})$

  - *Orthogonal cluster functions:*

$$\left\langle \Phi_\alpha^s, \Phi_\alpha^t \right\rangle_{all\vec{\sigma}} = W\delta_{st}$$

Algorithm

First principles calculations of all possible unitcells to locate group of unitcells with lower energies

- To reduce the number of unitcell types to be included in further calculations.

First Principle calculations on small supercells built from selected unitcells

- To generate datasets for evaluating effective cluster interaction parameters (ECI)

Solve the over determined equations to find out the effective cluster interactions (ECI)

- To obtain ECI from the supercell calculations.

Do Monte Carlo simulations to calculate free energy of much large systems based on ECI

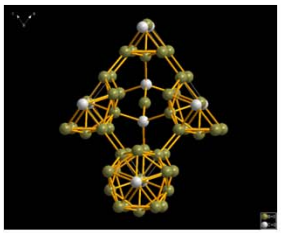
The large supercell's energy is calculated by ECI instead of first principle calculation

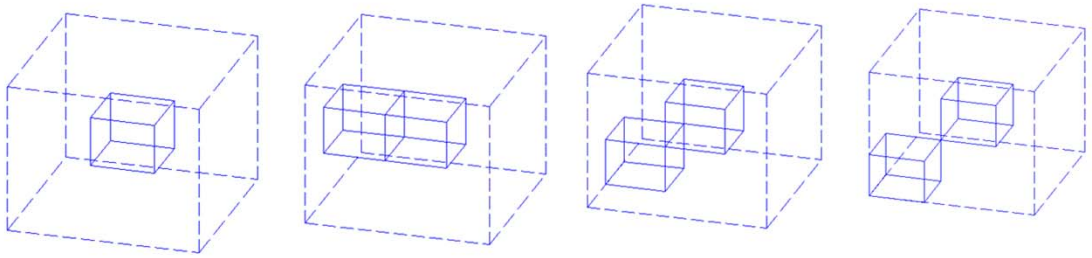
- To estimated configurational free energy.

Complex solid solution: B_4C

Clusters:

(1) self
(2) face-share
(3) edge-share
(4) corner-share





Unitcell Selection:

(1) for periodic structures consisted of one type of unitcell
compute the total energy and pressure

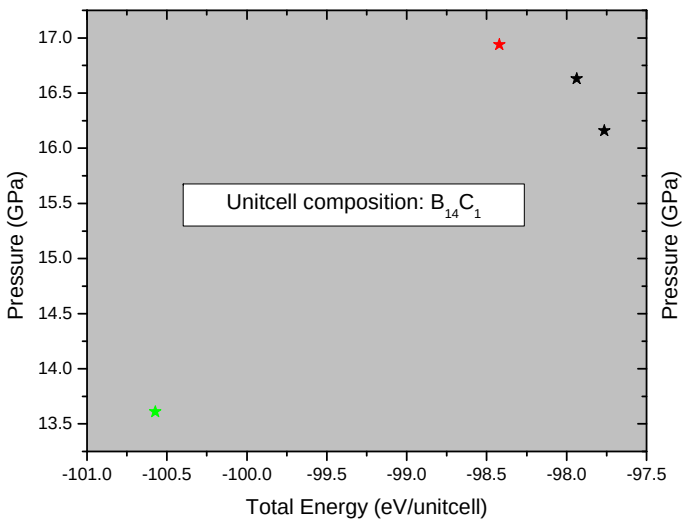
(2) group analysis of the total energies and pressures of the unitcells with the same concentration

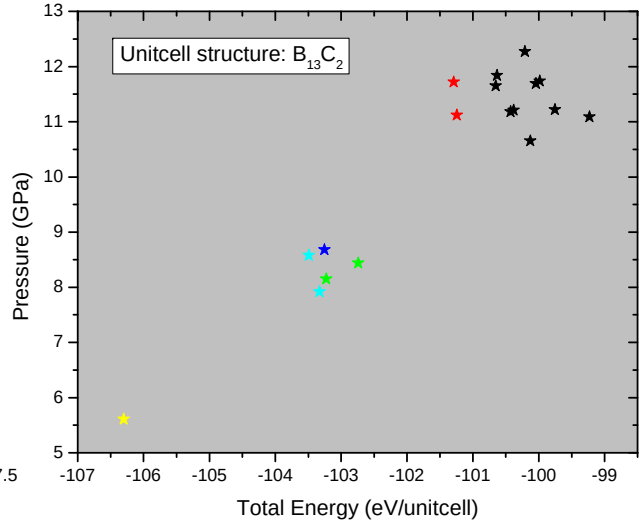
(3) select the lowest group in the total energies-pressure plot to be included in the set of unitcells (prefers unitcells with minimal intercell interactions)

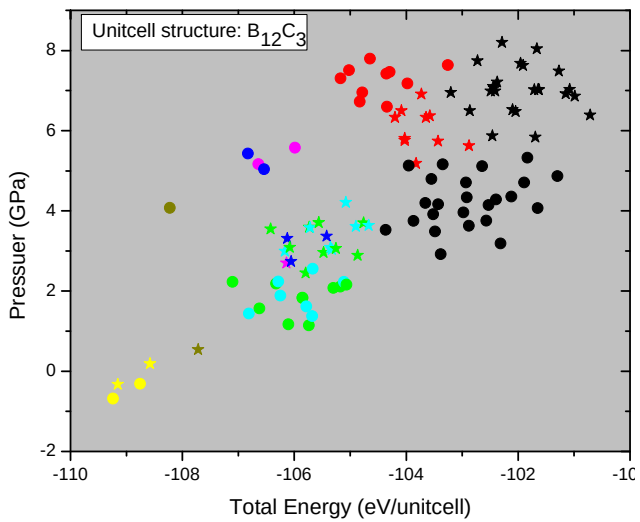
(4) it is possible to add more unitcells to the set using the criteria of cross-validation

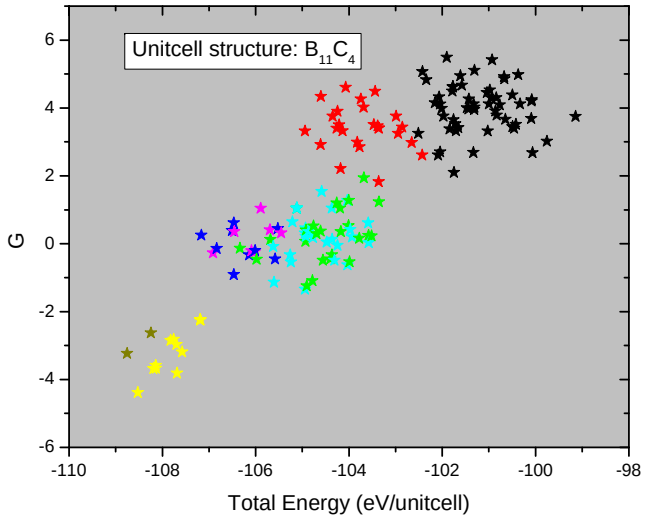
Unitcell Selection

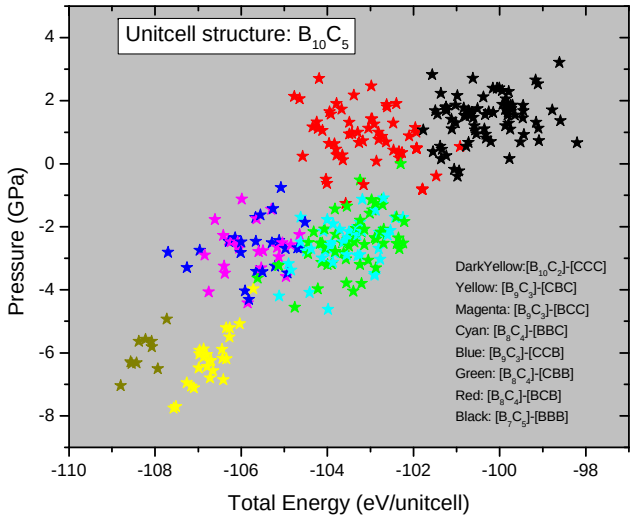
Concentration dependent energies of lattice with 1 unitcell type



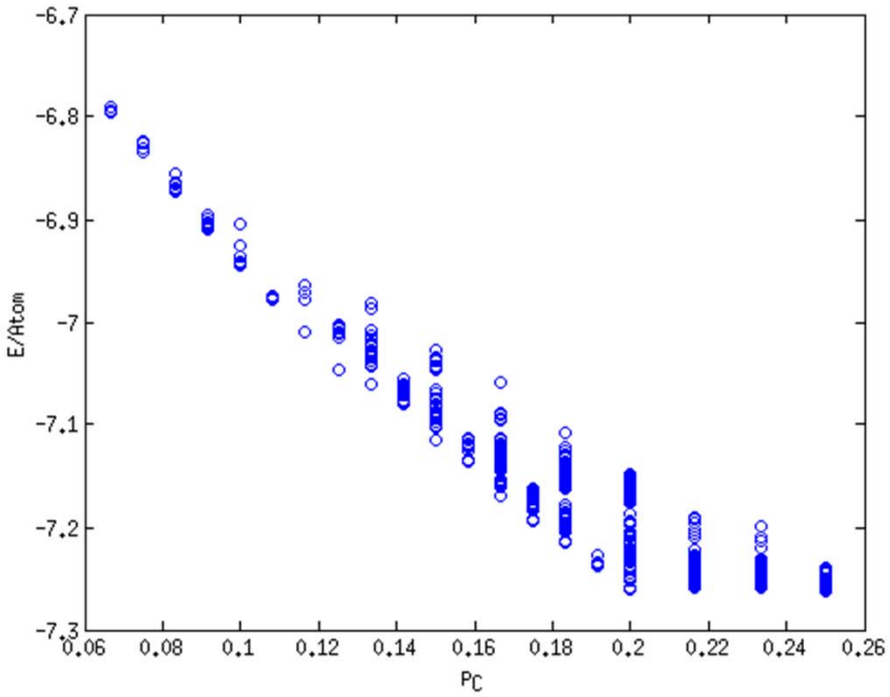


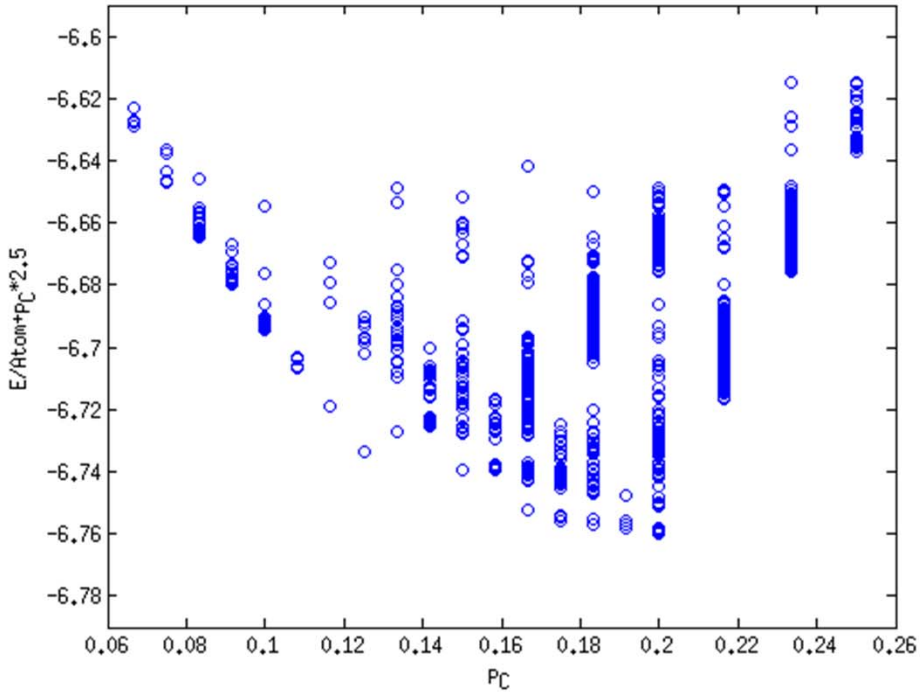




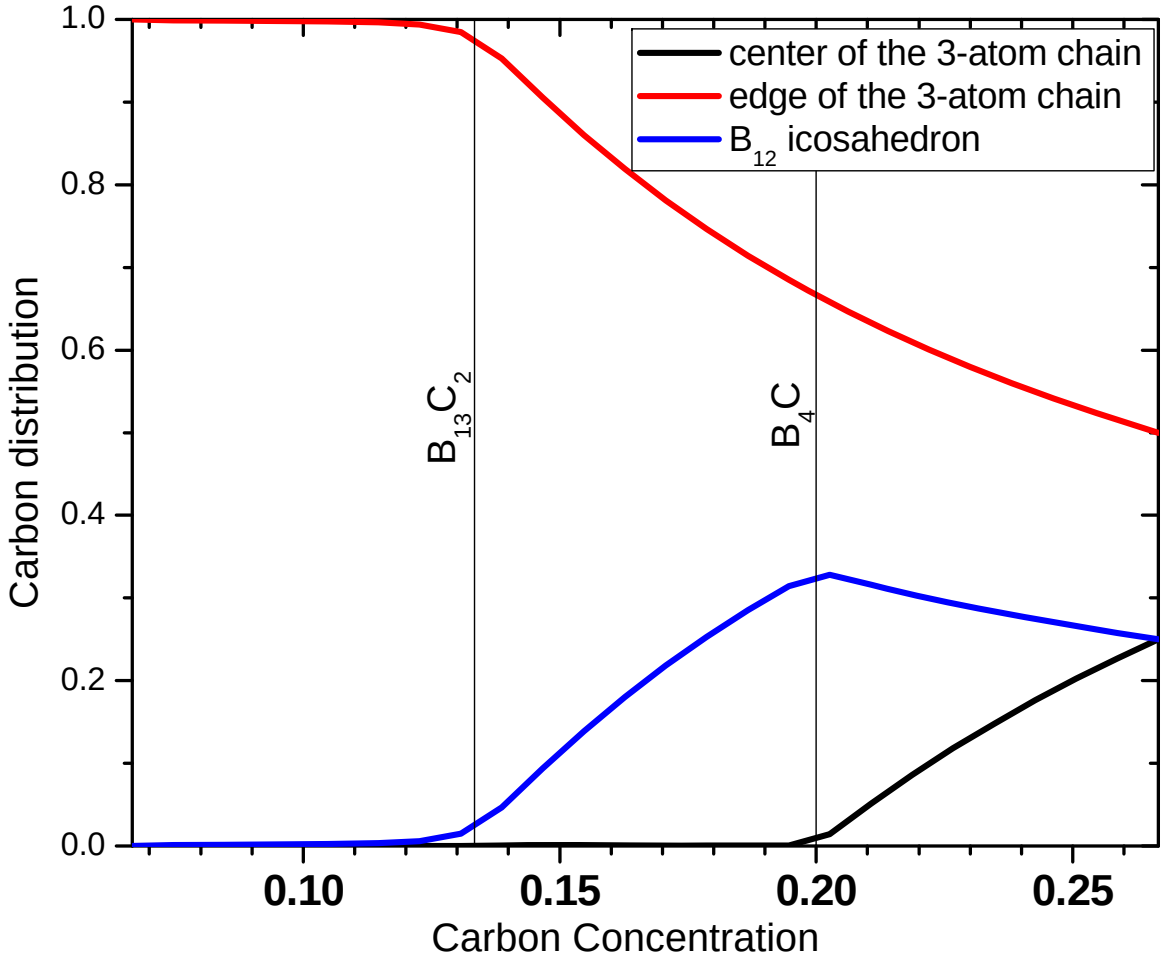


Convex Plot of Supercell Sampling





Site Occupation from Monte Carlo Simu.



Carbon distribution in Boron carbide $B_{1-x}C_x$. Red line indicates carbon percentage at two edge sites of the 3-atom chain. Blue line shows the carbon percentage in the icosahedrons. Black line depicts carbon percentage at the center of the 3-atom chain.

Special Quasirandom Structure

- **Poor man's approximation to cluster expansion method**

$$F(\vec{\sigma}, T) \cong \sum_{\alpha, s} K_{\alpha}^{s}(T)\Phi_{\alpha}^{s},$$

$$\left\langle F \right\rangle_{SQS} \cong \sum_{\alpha, s} K_{\alpha}^{s}(T)\left\langle \Phi_{\alpha}^{s} \right\rangle_{SQS},$$

$$\left\langle \Phi_{\alpha}^{s} \right\rangle_{SQS} \approx \left\langle \Phi_{\alpha}^{s} \right\rangle$$

Generating SQS Set

- **At high temperature limit**
  - site occupation is complete random
  - correlation function is known

$$\rho_\alpha(\sigma_{HT}) = \prod_{i\in\alpha}\left\langle \gamma_{\alpha_i, M_i}(\sigma_i)\right\rangle_\alpha$$

  - match SQS correlation with the known correlation function
    - ✓Exhaustive search
    - ✓Genetic algorithm
    - ✓Other global optimization approach may also be used

Project Timeline/Milestones

Project tasks	Year 1				Year 2			
A.1 G(P,T) module for automated structure modeler								
A.2 G(P,T) module for implement the SQS method								
B.1 Application to known 9-12Cr ferritic steels								
B.2 Screening Studies of BCC solid solution								

Tasks

Methodology Development

- **A.1 G(P,T) module for automated structure modeler**

  The module will have the capability of:

  - generate solid solution model based on supercell specified in lattice vectors, i.e. the miller indices, and composition specified in partial occupations;
  - batch process without human intervene;
  - generate the special quasirandom supercell structures set for the SQS method.

Tasks

Methodology Development

- **A.2 G(P,T) module implementing the special quasirandom structures method**

  The module is responsible to invoke structure modeler implemented in A.1 to produce the set the special structures and setup VASP calculations to obtain properties of the set.

Tasks

B. Application to 9-12Cr Ferritic Steels

- **B.1 Properties of known 9-12Cr ferritic steel**
  - Positive control using well characterized 9-12Cr ferritic steels
    - P91, E911, P92, AXM, HCM12, P122, T122, NF12, FN5, TB12, VM12 and X20
    - Formation energy and Elastic constants will be assessed initially.
    - Assess ductility using the ratio of bulk modulus and shear modulus.
    - Stacking fault energy and surface cleavage energy will be carried out to estimate the Rice-Thomson parameter which is widely used as ductility criterion.
    - Trend analysis of the Rice-Thomson parameter.

Tasks

B. Application to 9-12Cr Ferritic Steels

- **B.2 Screening studies of BCC solid solution**
  - Large screening in a progressive manner with 4 components BCC solid solutions examined first.
  - The surfaces of the properties in the composite space are examined to refine the likely zone of sampling for additional components based our results of known ferritic steels.
  - Global optimization methods such as simulated annealing and genetic algorithm to locate the optimal ferritic design.

Budget

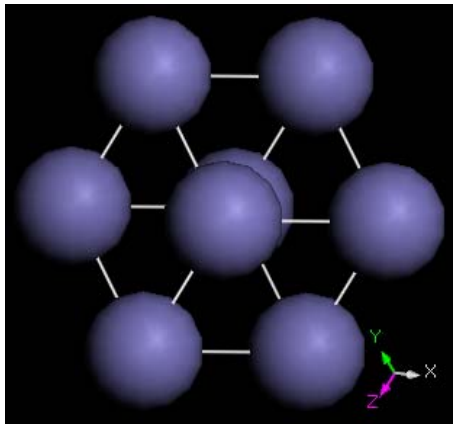
- PI: 1 summer month ($5851 + benefit)
- Postdoc: 12 months ($42000 + benefit)
- Travel: $5000 for two trips
- Supplies: $1865
- Computer: $1000

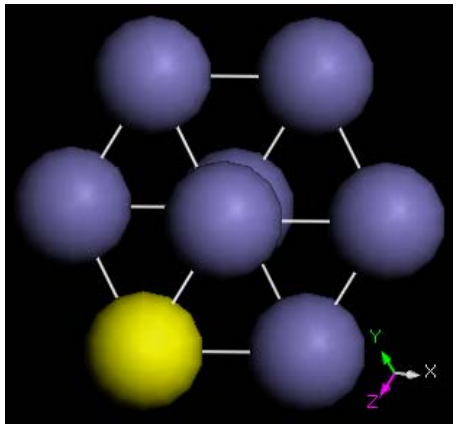
Status

- Coarse grained cluster expansion of dilute BCC iron
- Preliminary results on solid solution z-phase in steel

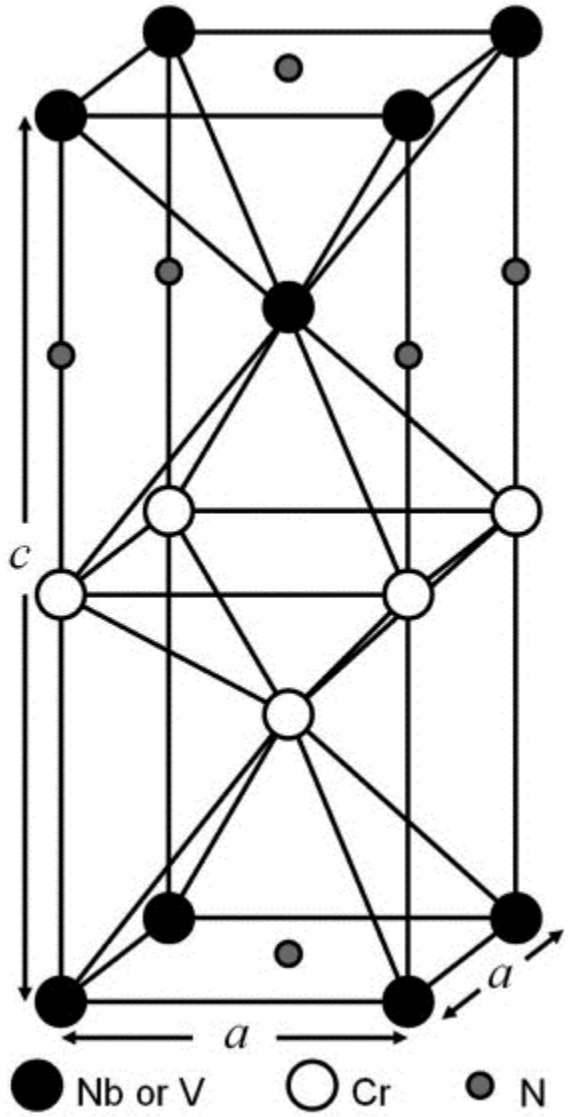
CGCE: BCC Iron

- **Choice of Coarse Grained Cell**
  - ✓ 2x2x2 supercell is used. (dilute BCC iron)
  - ✓ 8 atoms per CGC
  - ✓ 4 types of unit cell: Fe_8, $CrFe_7$, $AlFe_7$, $SiFe_7$



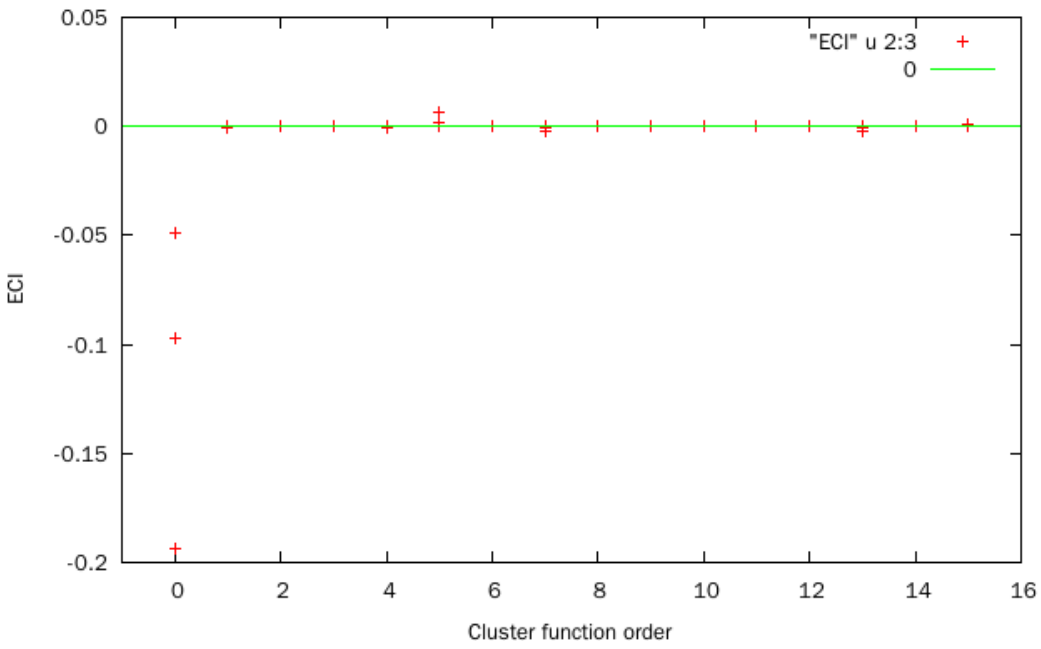


Z-phase in steel

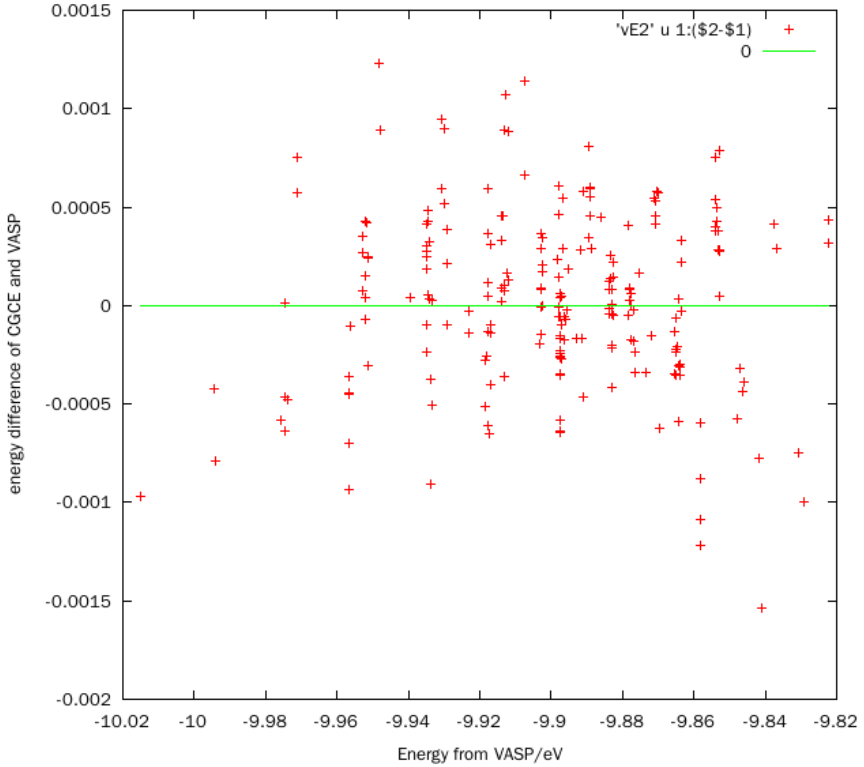


- Based centered tetragonal lattice
- Primitive cell contains two disordered sublattice sites that are occupied by Nb or V
- 4 type of cells used in the UEM calculation.
- Clusters limited no more than pairs.

ECI and Cross-Validation



ECI vs. Cluster function order parameter *s*
Higher order negligible



Cross-Validation: (energy difference between energies obtained from direct VASP calculations and cluster expansion) . ECIs are from obtained from different supercell set.

Project Team

- PI: Lizhi Ouyang
- Postdoc:
  - Ranganathan Parthasarathy (BioEngineering, KU)
- Graduate Students:
  - Jonathan Reynolds
  - Ayodeji Borode

Thank you!