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

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Outline

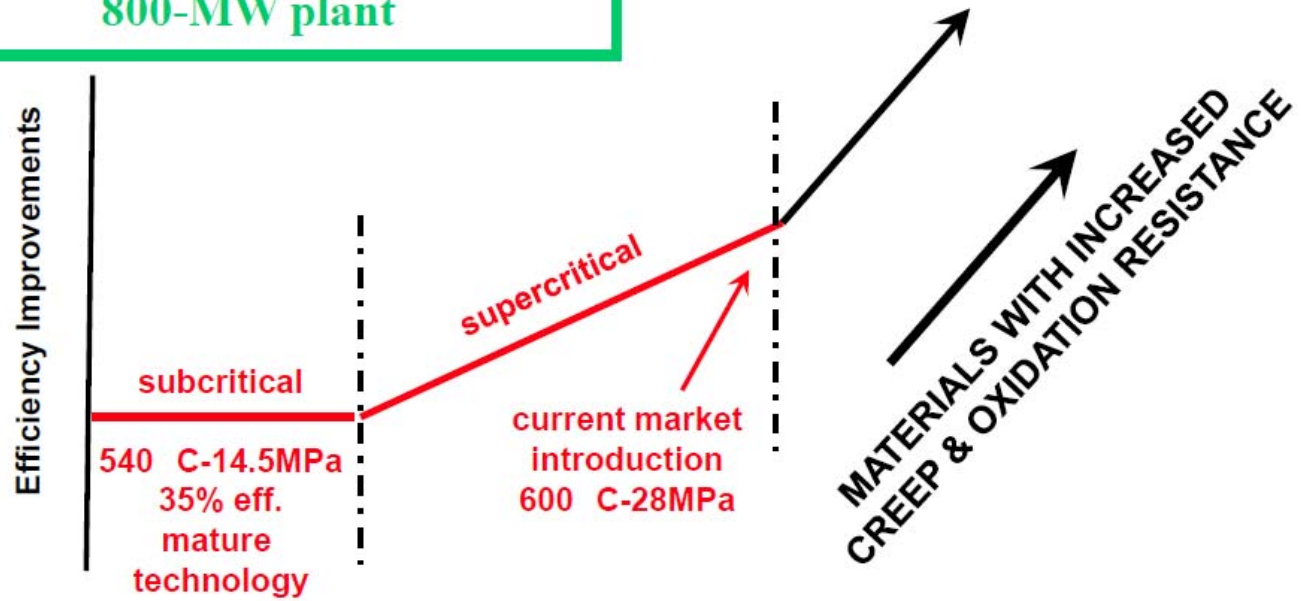
- Background
- Goal
- Approach
- Project status

Background

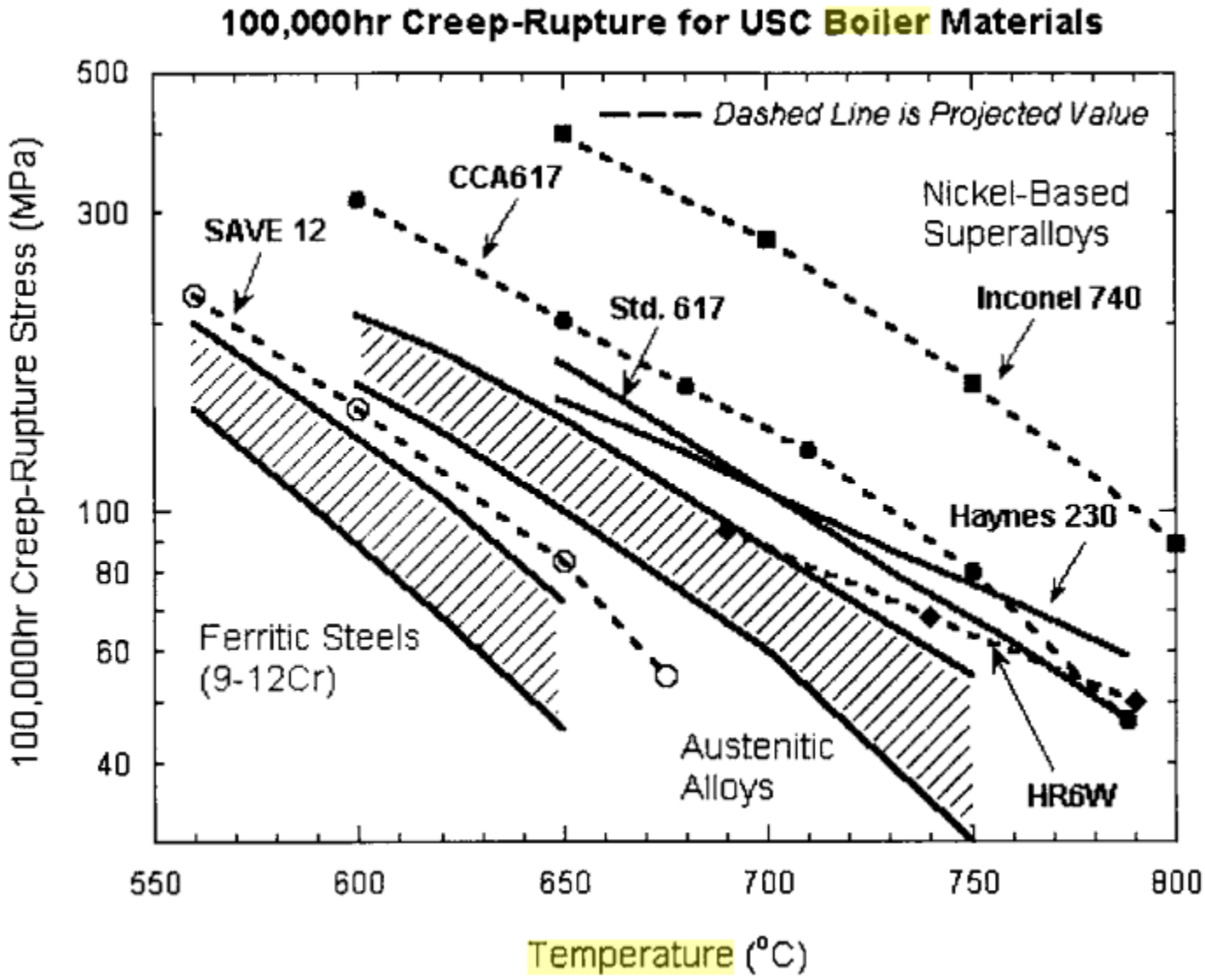
Increasing Efficiency

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

Known 9-12% Cr Ferritic Steels

Table 1. Precipitate Phases in Different Steel Phases. PT Represent for Prototype Structure and SG for Space Group Number.

			Steel Phases											
Precipitate Phase	PT	SG	P91	P92	E911	AXM	HCM12	P122	T122	NF12	FN5	TB12	VM12	X20
BCC_A2	W	229	X	x	x	x	x	x	x	x	x	x	x	x
M23C6	Cr23C6	225	X	x	x	x	x	x	x	x	x	x	x	x
LAVES	MgZn2	194	X	x	x		x	x	x	x	x	x	x	x
Z_PHASE	NaCl	225	X	x	x	x	x	x	x	x	x	x	x	x
$NbNi_3$	Al3Ti	139	X			x	x	x	x	x	x			
AlN	ZnO	194	X	x	x	x		x	x			x		
SIGMA	CrFe	136		x	x							x		
FCC_A1	Cu	225		x	x	x	x	x	x	x		x	x	x
HCP_A3	Mg	194		x	x							x		
M_2B_{Tetr}	Fe2B	140		x		x								
MU_PHASE	W6Fe7	166				x								
M6C	W3Fe3C	227				x								
Cr_2B_{Ortho}	Mg2Cu	70						x	x	x	x		x	
PI	Mo3Al2C	70										x	x	

Ferritic Steel 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

- 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 atomic structure model
  - components of very low concentration considered as point defect

- Special quasi-random structures

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*

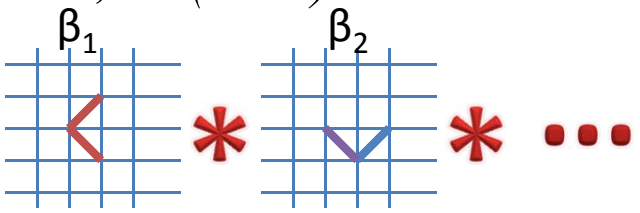
G(P,T) Module: UnitCell Expansion

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

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

$$F(\vec{\sigma},T) \cong \sum_{\alpha,s} K^{s}_{\alpha}(T)\Phi^{s}_{\alpha},$$

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

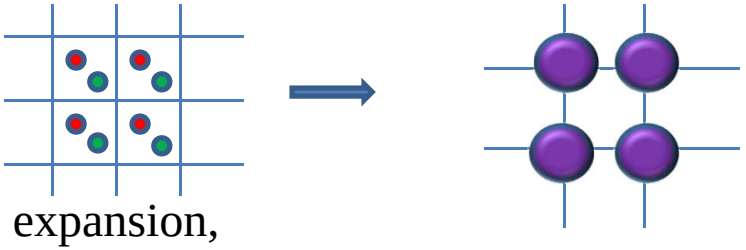


Challenge: number of cluster expansion terms n

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

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

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



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*)

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

Tasks

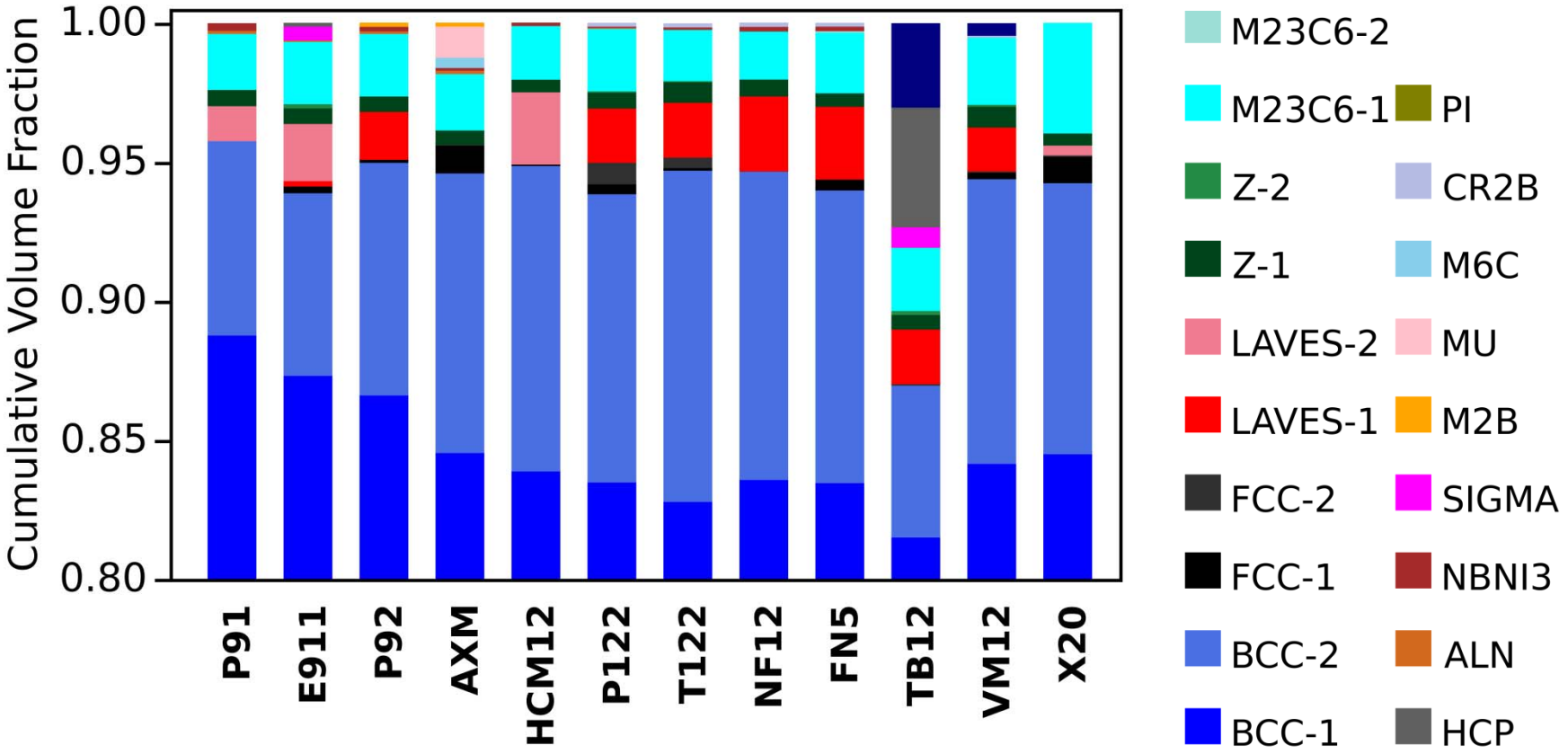
B. Application to 9-12Cr Ferritic Steels

- **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 indicative parameter of ductility.

Challenges In Structure Modeling

- **Incomplete structure information about the phases in the steel**
  - Missing information about site distribution
    - Phase compositions are known
    - Multiple non-equivalent solid solution sites
- **Composition requires exceedingly large model**
  - Dopant concentration between 1000ppm to 1ppm
    - Need to assess the dopant-dopant interaction
    - Model requires > 1000 atoms
  - Dulite dopant can be treated as point defect
  - Using the scaling law to estimate the effect of dopant with intermediate concentration: $G \sim c^{\alpha}$

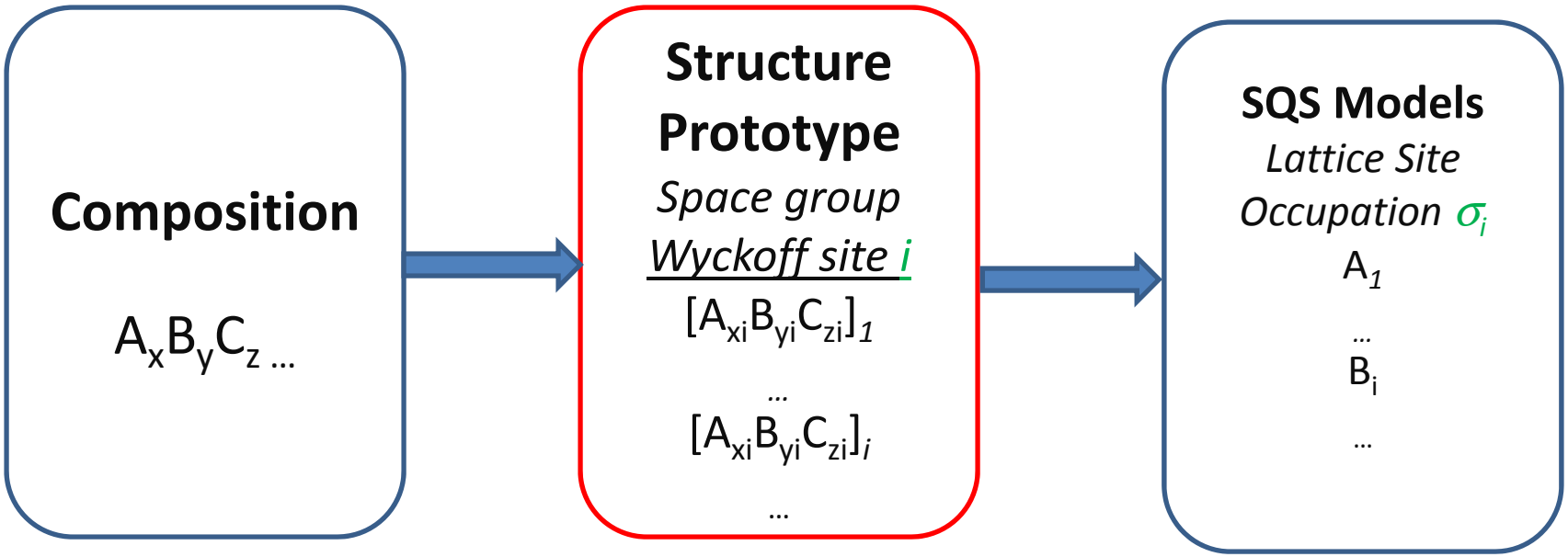
9-12 Cr Steel: Phase Volume Fraction



AXM Steel Facts

Phase	Vol fract	Composition	Crystal	Microstructure
BCC-A2#2	0.8464	$Fe_{0.996}Si_{0.002}Al_{0.001}$	cI2	Matrix phase
BCC-A2#1	0.1006	$Cr_{0.957}Mn_{0.043}$	cI2	Precipitation
$M_{23}C_6$	0.0203	$(Cr_{0.864}Mn_{0.130}Fe_{0.006})_{23}C_6$	cF116	Precipitation
μ-Phase	0.0112	$(Fe_{0.992}Cr_{0.008})_7(W_{0.650}Mo_{0.350})_6$	hR39	Precipitation
FCC-A1#1	0.0100	$Ni_{0.584}Fe_{0.370}Si_{0.046}$	cF4	Precipitation
M_6C	0.0037	$(Mo_{0.992}W_{0.008})\ MoFe_2C$	cF112	Precipitation
Z-Phase	0.0051	$(Cr_{0.898}Fe_{0.102})VN_{0.669}$	tP6	Precipitation
$NbNi_3$	0.0010	Ni_3Nb	oP8	Precipitation
AlN	0.0012	AlN	hP4	Precipitation
FCC-A1#3	0.0003	$Cu_{0.999}Ni_{0.001}$	cF4	Precipitation
M_2B	0.0003	$(Mo_{0.953}Cr_{0.047})_2B$	tI12	Precipitation

Structure Modeling



Total composition

Wyckoff site composition

Lattice site occupation

Structure Modeling

❑ Structure models generation

- Based on structure prototype
  - Limited to size < 250 atoms
  - Site-distribution based on prior knowledge

❑ Composition requires exceedingly large model

- Dopant concentration between 1000ppm to 1ppm
  - Need to assess the dopant-dopant interaction
  - Model requires > 1000 atoms
- Dulite dopant can be treated as point defect
- Using the scaling law to estimate the effect of dopant with intermediate concentration: $G \sim c^{\alpha}$

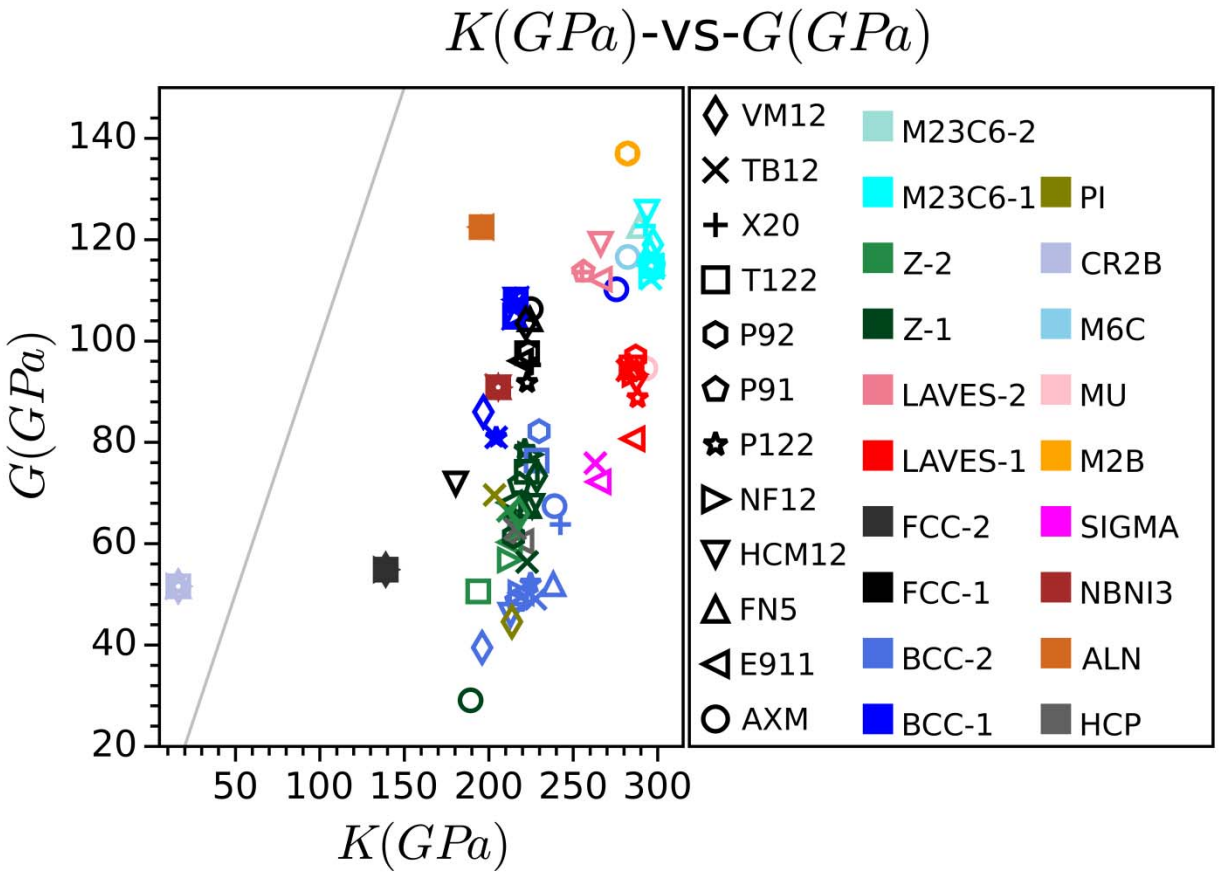
Elastic Constants Calculations

- Calculated **use the in-home developed G(p,T) package**
  - Employed VASP as the computing engine
  - Automate the calculation and fully taking advantages of symmetry
  - Both stress and energy based calculations are available
  - Accuracy setting:
    - Standard 400eV energy cutoff
    - Standard K-point sampling: metal (cutoff 35), others (cutoff 25)
    - Energy convergence 10^{-6} eV
    - Spin polarized calculation for selected phases
  - More than 100 SQS models have been calculated. Many involves more than 4 elements are for the first time been calculated.

Elastic Constants Calculations: AXM

Phases	C_{11},C_{22},C_{33}	C_{44},C_{55},C_{66}	C_{12},C_{13},C_{23}	K	G	E	ν	G/K
BCC-A2#1	396	126	215	275	110	292	0.323	0.400
BCC-A2#2	395	53	168	242	72	196	0.365	0.297
AlN	377 356	113 125	129 99	196	122	304	0.241	0.622
FCC-A1#1	322	147	179	225	106	275	0.296	0.473
Cu	181	83	121	140	56	147	0.324	0.399
$M_{23}C_6$	459	111	216	297	115	306	0.328	0.388
M_2B	440 504	141 136	199 190	282	137	353	0.291	0.486
M_6C	442	115	203	282	117	308	0.318	0.413
μ-phase	442 426 406	92 94 94	245 225 217	293	95	256	0.354	0.323
$NbNi_3$	290 305	113 111	178 153	206	91	238	0.307	0.442
*Z*phase	278 250	45 8	180 167	189	29	83	0.427	0.154

Elastic Properties: Known 9-12 Cr Steel



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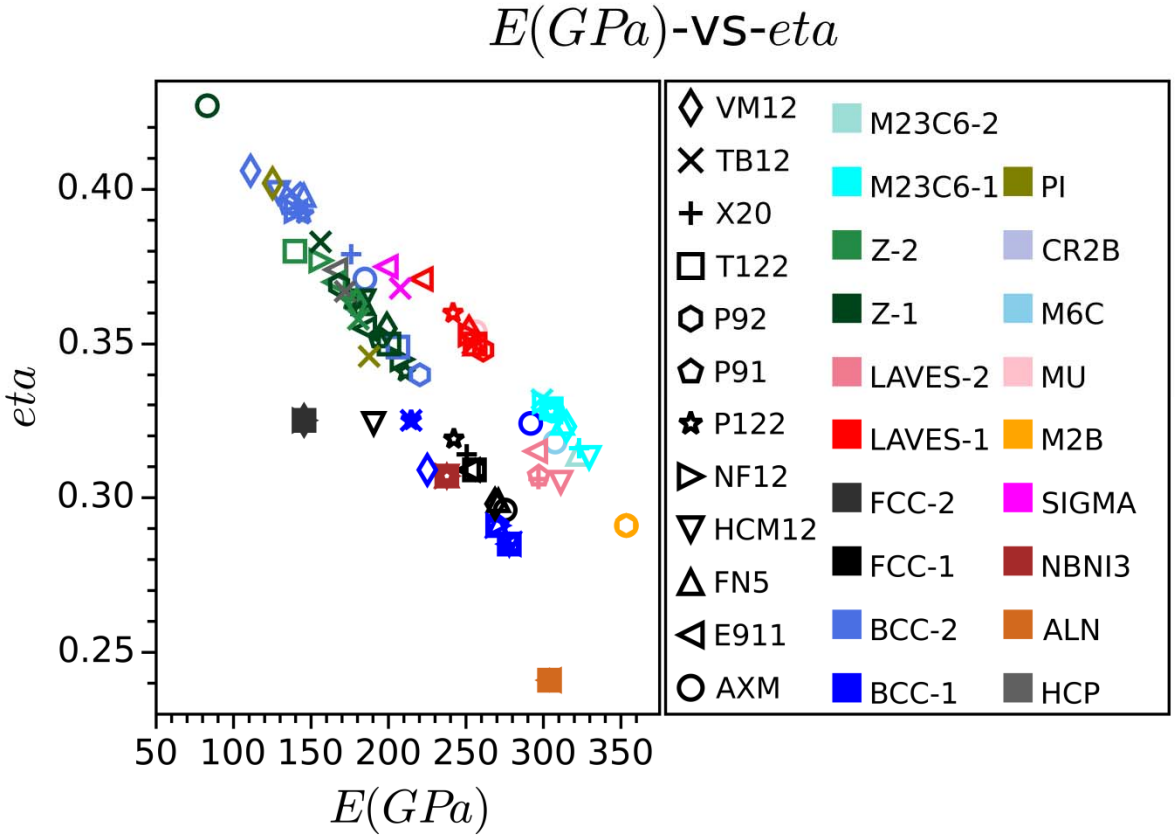
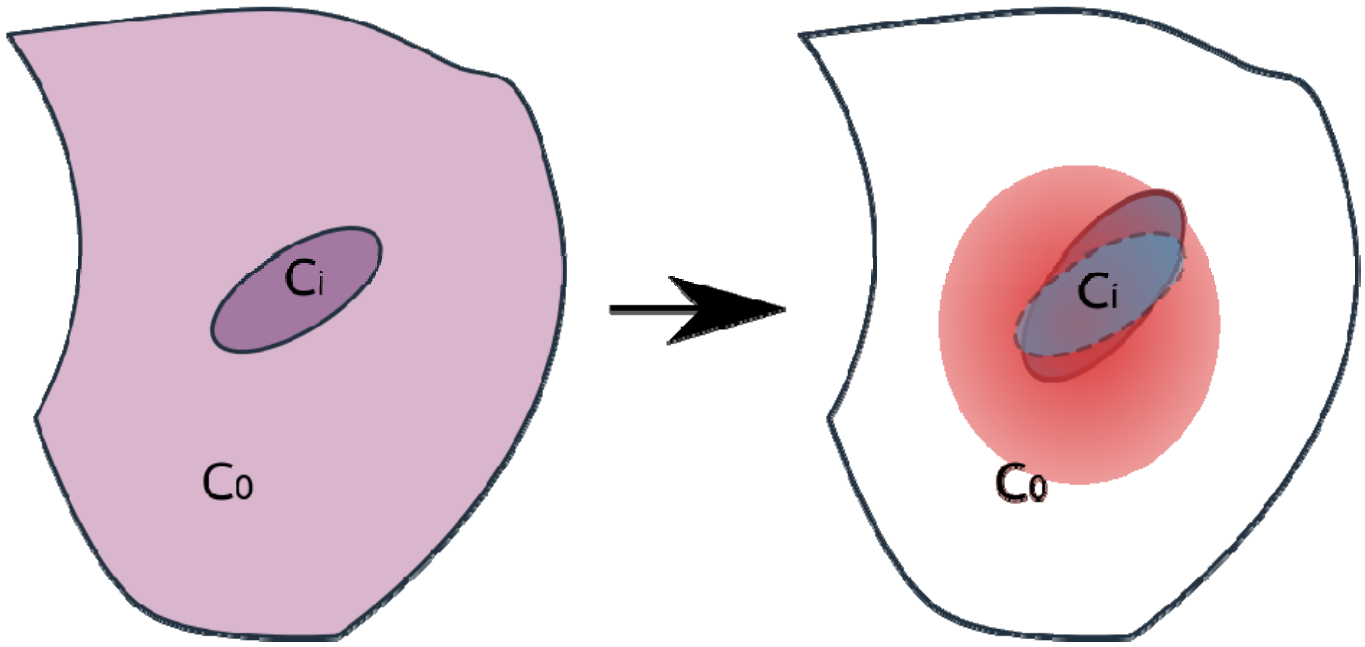


Precipitation Effects

- Homogenization scheme to assess the precipitation effects on elastic constants
  - Eshelby's inclusion theory:
    - uniform stress/strain field inside the inclusion



Effective Self Consistent Scheme

- Iterative scheme
- Multiple inclusion phases allowed
- Only volume fraction and bulk elastic constants used in the scheme *

* Precipitation size and shape information described by Eshelby tensor. Assume isotropic spherical inclusion for calculating Eshelby tensor **S**

$$C^* = (H + C_M^{-1})^{-1}$$

$$H = \text{sum}\ (H_{I,i}^{d}\ (I - \Omega_{DI,i}\ H)^{-1}$$

$$H_{I,i}^{d} = c_i \{ (C_{I,i}^{-1} - C_M^{-1})^{-1} + C_M(I - S_{I,i}^{M}) \}^{-1}$$

$$\Omega_{DI,i} = C^*(I - S_{I,i}^{*})$$

Elastic properties of steel

- Homogenized elastic modulus (GPa)

	K	G	Y	η
AXM	271	102	272	0.333
P92	251	84	228	0.349
T122	248	77	209	0.359

* Other steels have unstable precipitation phases are shown in the table.

Tasks

B. Application to 9-12Cr Ferritic Steels

- **B.2 Screening studies of Ferritic Steels**
  - Screening in a progressive manner with 4 components BCC solid solutions examined first.
  - Precipitation effects to be assessed through homogenization
  - Global optimization methods such as simulated annealing and genetic algorithm to locate the optimal ferritic design.

Search For Optimal Ferritic Design

- **From composition to phase distribution**
  - Concentration dependent chemical potential in phases estimated from computed and measured properties of known steel phases
  - Energy minimization process to compute the phase distribution

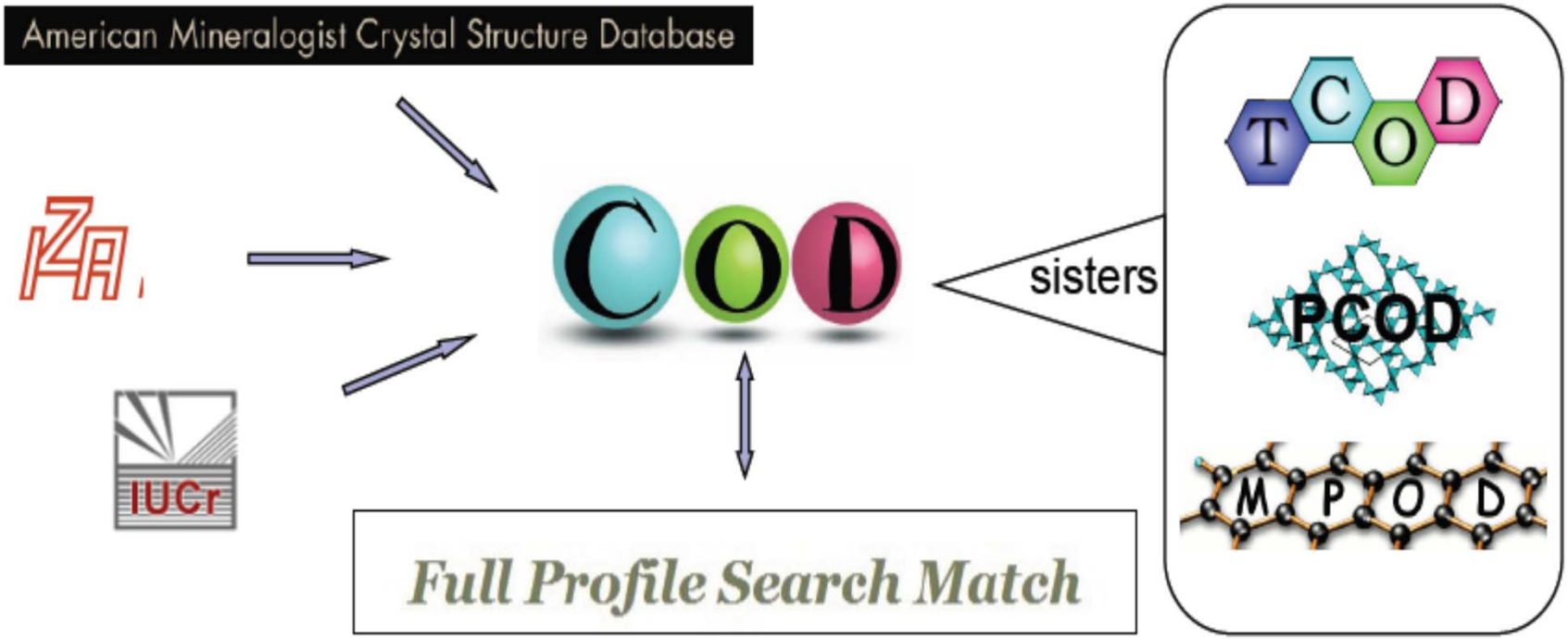
- **Homogenization to evaluate the overall mechanical properties**
  - Mechanical properties of solid solution estimated using empirical laws based on computed and measured properties for given composition.

Inverse: Properties →Structure

- First principles method brings chemical accuracy for properties calculation with known structure
- The inverse process from properties to predict structure remains the greatest challenge to material science
- Two-step approach/Material genome approach
  - Properties -> Composition -> prototype library

HCOD: A Hign Performance Database

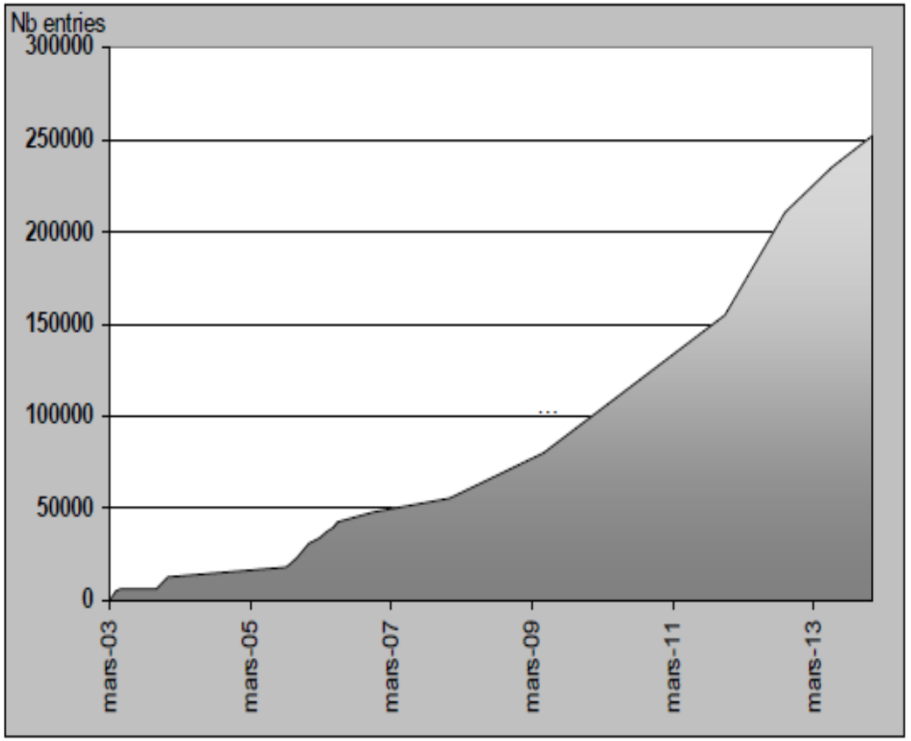
- Crystallography Open Database (COD) is an open-access database for crystal structures.
- collects the information of all small to medium sized unit cell crystallographic structures, including organic, inorganic, metal-organic compounds and minerals.



HCOD: A Hign Performance Database

- All registered users can deposit published and unpublished structures into COD database.
- COD can be extended by large number of users simultaneously, which greatly increases the grown rate of COD.
- COD stores data in a uniform format: Crystallographic Information File (CIF), one structure per file.

As Feb 2016, there are more than 350,000 entries in COD



HCOD: A High Performance Database

Research Goal:

- Build a high performance COD (H-COD) database with efficient query system on distributed computer cluster

Research Objectives:

- Design a well-organized structure for H-COD database.
- Design efficient query operations including search, updating, insertion and deletion.
- Implement the database in a distributed and parallel computing system;
- Develop Web APIs that provide user-friendly query interfaces to the database.

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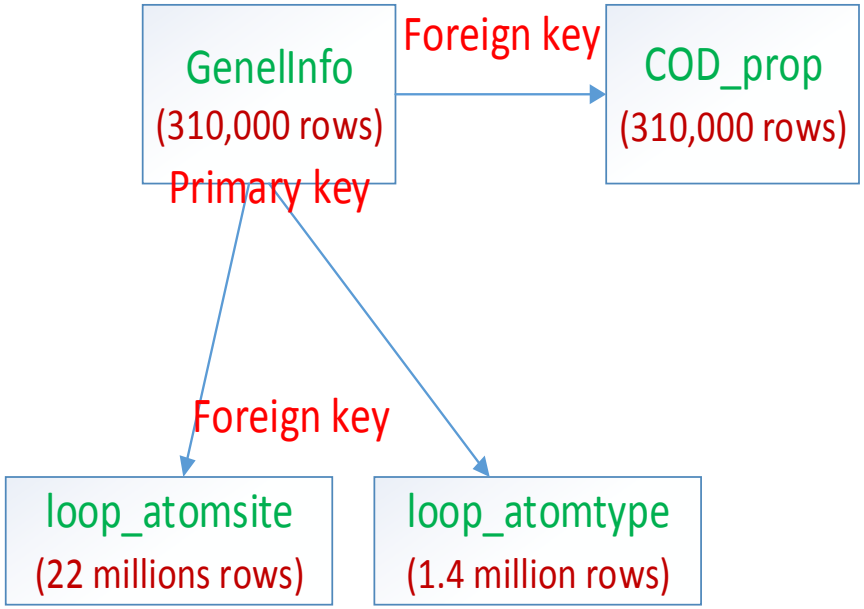
HCOD: A High Performance Database

Table design

Four tables

- First three tables are GenelInfo, Atom-site and Atom-type, where the information come from the CIFS in COD.
- The fourth table: COD_prop contains new attributes and values generated by the LatMGA method.

To maintain referential integrity of data, we use foreign key constraint: COD number is chosen as primary key in GenelInfo table and foreign key in loop tables.



HCOD:LatMGA

HCOD: Web Query Interface

Search COD CIF List by Fields

Year of Journal	2005
Journal Name	
Journal Issue	
Journal Volume	
Journal Paper DOI	
Elements	Al S ☐ Only
Number of distinct elements	Min: 2 Max: 6
Crystal type	
Composition	
Hermann-Mauguin symmetry space group	P 1 21/n 1
Space group id	
Cell length A	Min: Max:
Cell length B	Min: Max:
Cell length C	Min: Max:
Cell angle Alpha	Min: Max:
Cell angle Beta	Min: Max:
Cell angle Gamma	Min: Max:
Unit Z	Min: Max:
Wyck sites	Min: Max:
SCC NA	
SCC NB	
SCC NC	
SCC OCC	
IS ORGANIC	
	Search

LatMGA indices

LatMGA

- Ordered lattice structures:
  - Crystal and Solid Solution
  - Common description: $\{ \mathbf{L}_i \} \otimes \{ \mathbf{r}_j; \boldsymbol{\sigma}_j \}$
    where $\{ \mathbf{L}_i \}$ is the set of lattice symmetry operators
    $\{ \mathbf{r}_j \}$ is the set of lattice basis
    $\{ \boldsymbol{\sigma}_j \}$ is the set of composition vector

- Composition vector σ
  - $\boldsymbol{\sigma} = \Sigma_i \, \eta_i \, \boldsymbol{\varepsilon}_i$
  - Each vector basis ε_i represents an element or a structure unit
  - The component η_l represents the probability of the basis at the site
  - Vacancy is also a basis

Continue: LatMGA

- Material Genome Approach
  - The challenge is how to traverse the enormous configuration space
  - Uniform structure indices are highly desired for data mining

- **LatMGA**: separate lattice from composition
  - Structure = $\{ L_i \} \otimes \{ r_j; \sigma_j \} = [\{ L_i \} \otimes \{ r_j \}] \oplus [\{ L_i \} \otimes \{ \sigma_j \}]$
  - Structure is indexed by lattice prototype and composition type
  - Observation: any lattice basis sets is a subset of a fine grid insider the unit cell. For any stable structure at room temperature, **the grid needs not to be finer than the range of atomic vibration**. Therefore, any lattice may be a subset of supercell of a simple lattice such as simple cubic and hexagonal.

Continue: LatMGA

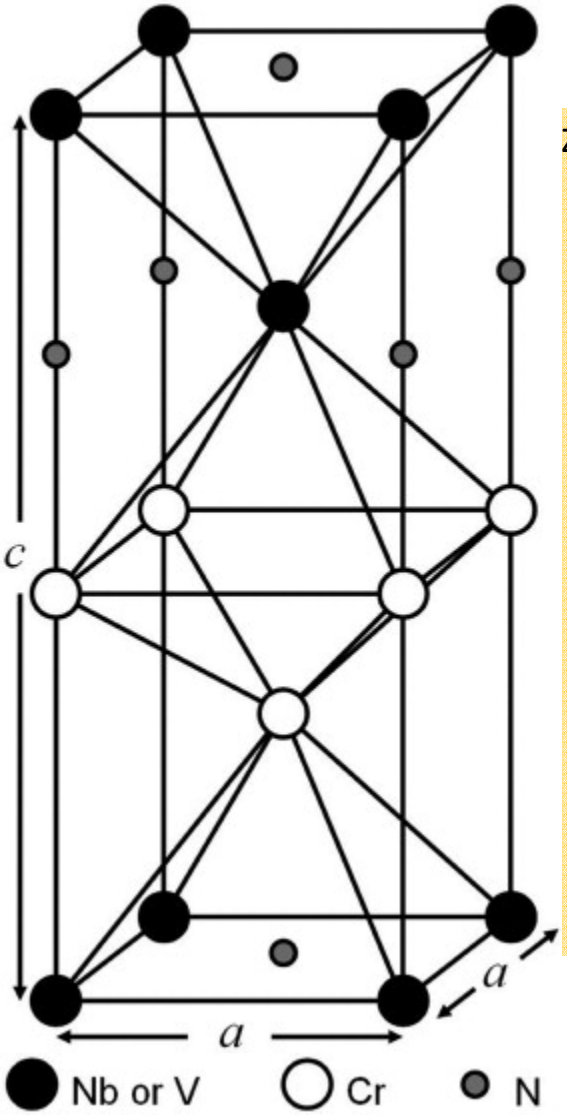
- **LatMGA:**
  - **A systematic approach to index structure prototype**
    - { # lattice prototype
    - { lattice unit type<scc,hcp, etc.>,
    - supercell <nxnxn>,
    - space group }
    - # composition space
    - { mask vector,
    - composition vectors }
    - }
  - **Three stage material configuration space exploration**

    { lattice prototype discovery:
    traverse the structure prototype indices; }

    { **composition space discovery**:
    rules based selection of mask/composition vectors;
    combine with lattice prototype to produce structure sample;
    structure sample can be evaluated using first principles methods; }

    { **material genome approach:**
    big data analysis on the high dimension structure indices space;
    inverse map from properties to structure; }

Continue: LatMGA



Z-phase indices:

Lattice prototype:

space group: 129 (P4/nmm)

irreducible sites:

Site_1	2 c	1/4	1/4	7/8
Site_2	2 c	1/4	1/4	5/8
Site_3	2 c	1/4	1/4	1/8

** it is a subset of 2x2x8 supercell of 1 basis simple cubic lattice casted into space group 129

Composition:

ε_1=Nb, ε_2=V ε_3=Cr ε_4=N ε_5=Vac

Site_1	(0.5 0.5 0 0 0)
Site_2	(0 0 1 0 0)
Site_3	(0 0 0 1 0)

casted from scc-2-2-8 to 129 :: size= 2 2 8

space_group 129
origin 2

Aa1	0.75000	0.25000	0.00000 #	2 a Aa
Ab1	0.75000	0.25000	0.87500 #	4 f Ab
Ac1	0.75000	0.25000	0.75000 #	4 f Ac
Ad1	0.75000	0.25000	0.62500 #	4 f Ad
Ae1	0.75000	0.25000	0.50000 #	2 b Ae
Af1	0.25000	0.25000	0.00000 #	2 c Af
Ag1	0.25000	0.25000	0.12500 #	2 c Ag
Ah1	0.25000	0.25000	0.25000 #	2 c Ah
Ai1	0.25000	0.25000	0.37500 #	2 c Ai
Aj1	0.25000	0.25000	0.50000 #	2 c Aj
Ak1	0.25000	0.25000	0.62500 #	2 c Ak
Al1	0.25000	0.25000	0.75000 #	2 c Al
Am1	0.25000	0.25000	0.87500 #	2 c Am

Continue: LatMGA

Progress:

- Tested against all cubic phases found in the Crystallography Open Database

- Excluding wrongful data, all can be indexed using supercell of simple cubic casted into the specific space group and a mask to indicate the closely matched Wyckoff sites (for example, Ag1, Ak1, Am1 sites, a mask vector [0 0 0 0 0 0 1 0 0 0 1 0 1])

- We are now in the process of implementing programs to search for un explored masks by combining with basis information of atoms such as atomic size, charge, etc.

Continue: LatMGA

Progress:

- All cubic phases with space group 225 (total 689 valid structures)
  - 304 mapped to SCC-2-2-2
  - 229 mapped to SCC-4-4-4
  - 10 mapped to SCC-6-6-6
  - 133 mapped to SCC-8-8-8
  - 13 mapped to SCC-12-12-12

Conclusions and Discussions

- Mechanical properties of all phases found in the 9-12% Cr ferritic steels computed; several phases were found unstable using the SQS models.

- Homogenization method developed to evaluate overall mechanical properties. Search for optimal composition/phase distribution/volume fraction is in progress.

- Development of the LatMGA method for:
  - Prototype library generation based on composition
  - Automate structure model generation based on prototype library
  - Automate the properties calculation of structure models
  - Data mining to map properties to composition

- Developed a high performance distributed database system for data mining of crystal structure information.