CODATA2006, beijing, China, Oct. 23-25, 2006

Integrating Data and Modeling In Materials Design

Ying CHEN[1], Yasunori KANETA[1], Shuichi IWATA[2]

[1] School of Engineering, The University of Tokyo, Japan

[2] Frontier Sciience, The University of Tokyo, Japan

Outline

I. Introduction: materials design

II. Approach

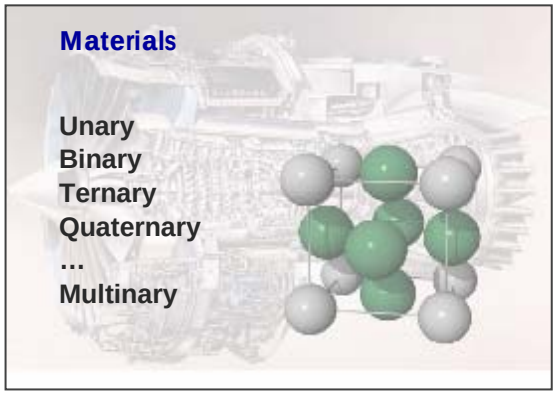
- Data-driven approach: data mining
- Modeling-driven approaches
- Data/Modeling-driven approach

III. Examples

- Structural map: empirical, computational
- Discovery of new H-storage materials
- 1st principles calculation of phase diagram

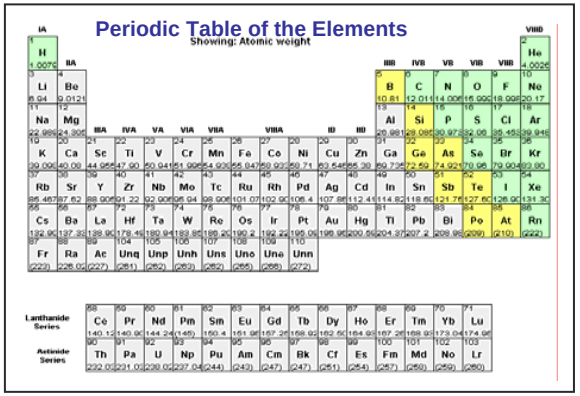
IV. Summary

I. Introduction: Materials Design



Too many Possibilities…?

Design



Prediction (Production Line)

Regularities ?

Needs — **Functions** — **Properties** — **Structures** — **Atomic Constituents**

Specification design | **Function design** | **Structure design** | **Process design**

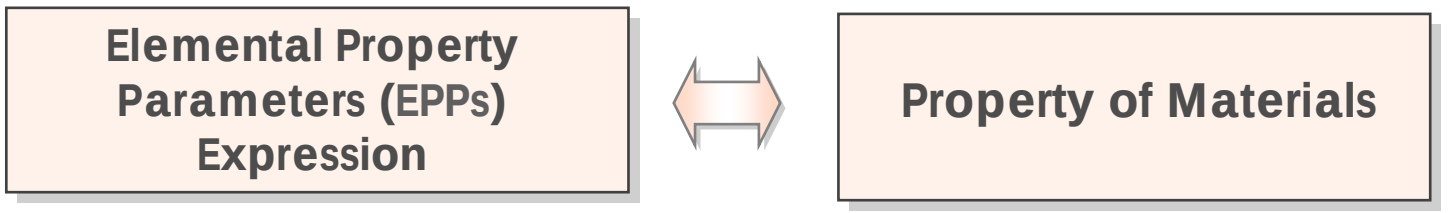
Materials Design (Resolution Line)

II. Approaches: Data-Driven Approach

Basic Idea

- **Based on the comprehensive materials database to reveal regularities:**
  - **Formation** of compound in a given binary system
  - **Composition** of stable compounds in "compound formers"
  - **Structures** of a given compound
  - **Properties** of a given compound

- **Postulation**



- **Tool: Materials Databases: Pauling File**

II. Approaches: Data-Driven Approach

Mapping

- **Purpose of Mapping**

  Proper **Elemental Properties** as **Axes**

  ↓

  Substances in same/similar structure/properties → **Groups**

- **Twbo key points in mapping**

  Characterization: To find optimal coordinates

  Classification: To define meaning of domainsb

II. Approaches: Modeling-Driven Approach

Calculations based on various physical models provide:

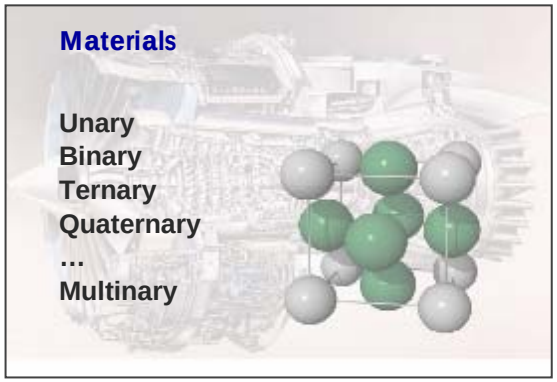
- **Complement** to empirical data, provide **new** data;
- **Further screening** and prediction of hypothesis;
- Understanding of **insight** into the origin;
- **Prediction** of materials with required properties.

II. Approaches: Modeling-Driven Approach

Theoretical Approaches

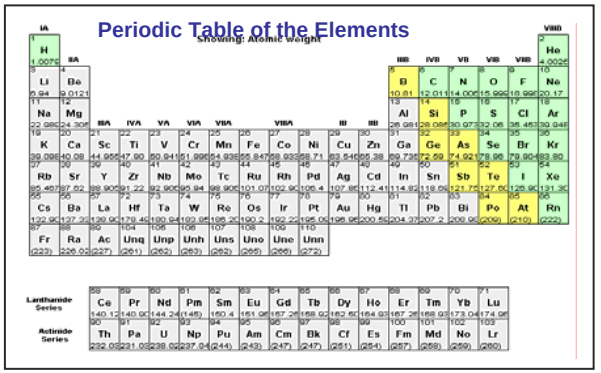
- First Principles Electronic Structures (FLAPW, Wien)
- Car-Parrinello Molecular Dynamics (CPMD, VASP)
- Cluster Expansion Method (CEM)
- Cluster Variation Method (CVM)
- Phase Field Method (PPM)
- Classical Molecular Dynamics (MD)
- ……

II. Approaches: Data/Modeling-Driven Approach

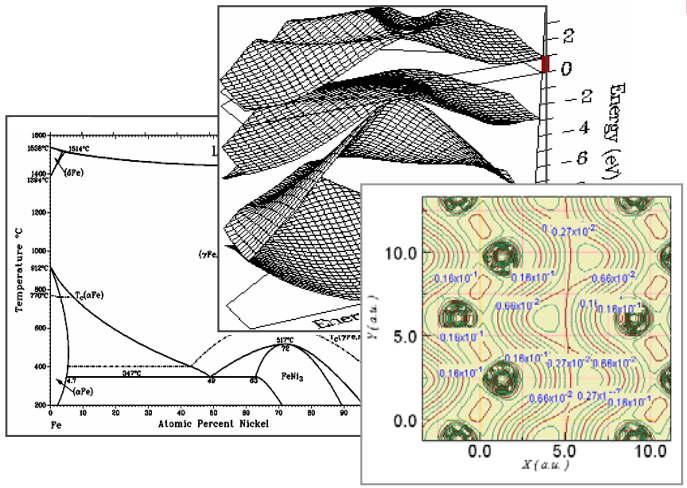


Too many Possibilities…?

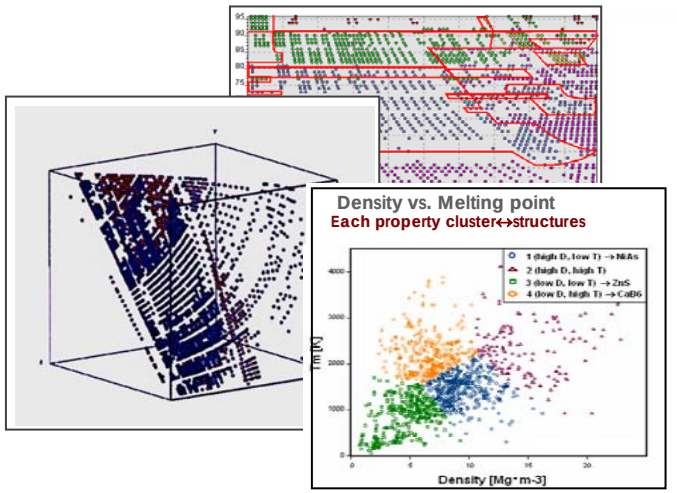
Design



Regularities



Model-Driven Approach → Origin



Data-Drive Approach → Discovery

Purpose: Regularity between Crystal structure & Element properties

Optimal coordinates
56
Element Property Parameters

?

- 6 most distinct EPP groups
  - Group number
  - Mendeleev number
  - Cohesion energy
  - Electrochemical factor
  - Size
- Operations
  - Sum EP(A)+E
  - Difference EP(A)-E
  - Product EP(A)*E
  - Ratio EP(A)/EP(B)
  - Maximum Max(EP(A),EP(B))

Decreasing pc

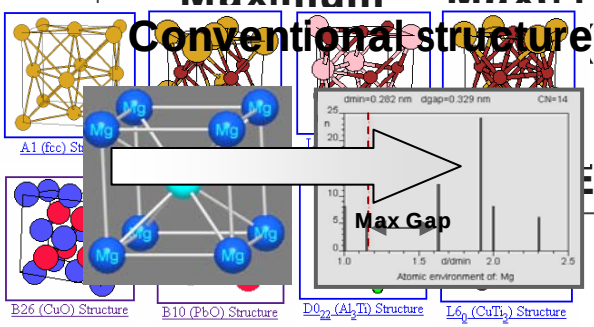
2-3
Optimal EPP Expressions

Distribution, Patterns, ... ☺

Definition of domains
~3,500
Conventional Structures Types

Conventional structure types

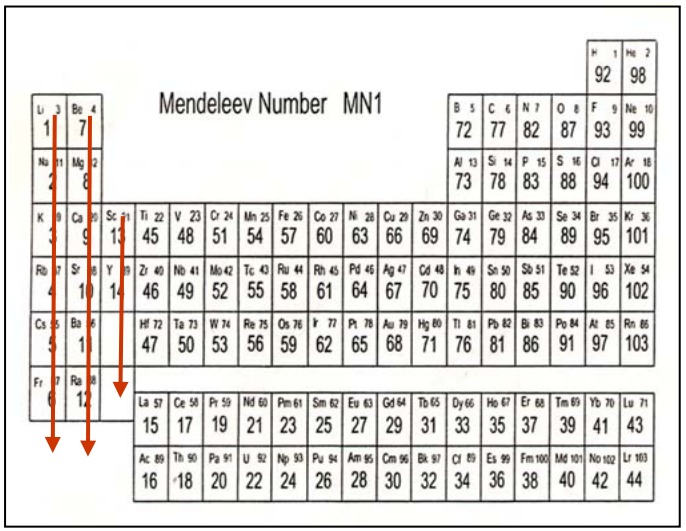
Conventional structure types

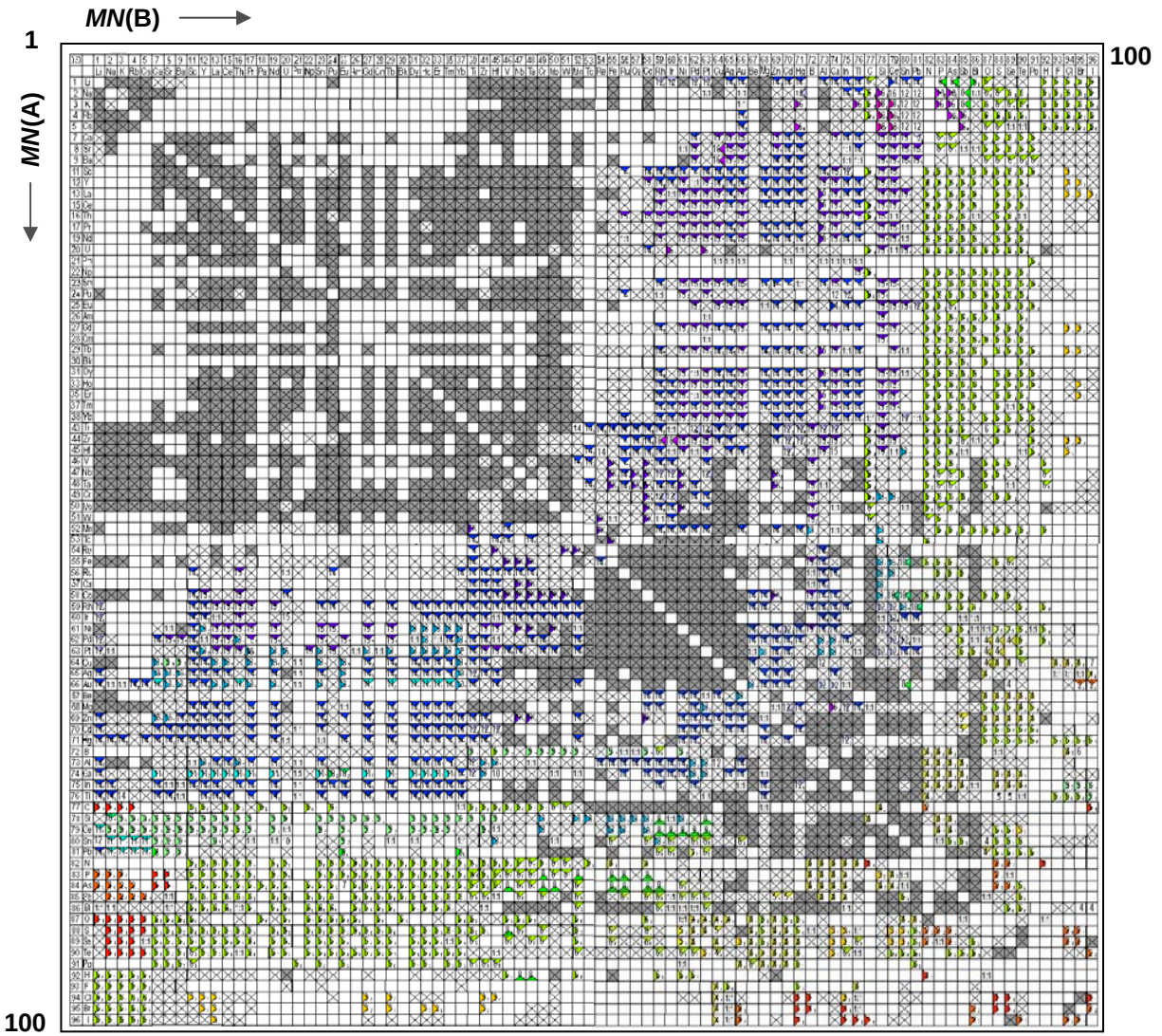


~30
Environment Types

MN(A) *vs.* *MN*(B) Map for 1:1 Binary Compounds (RT)

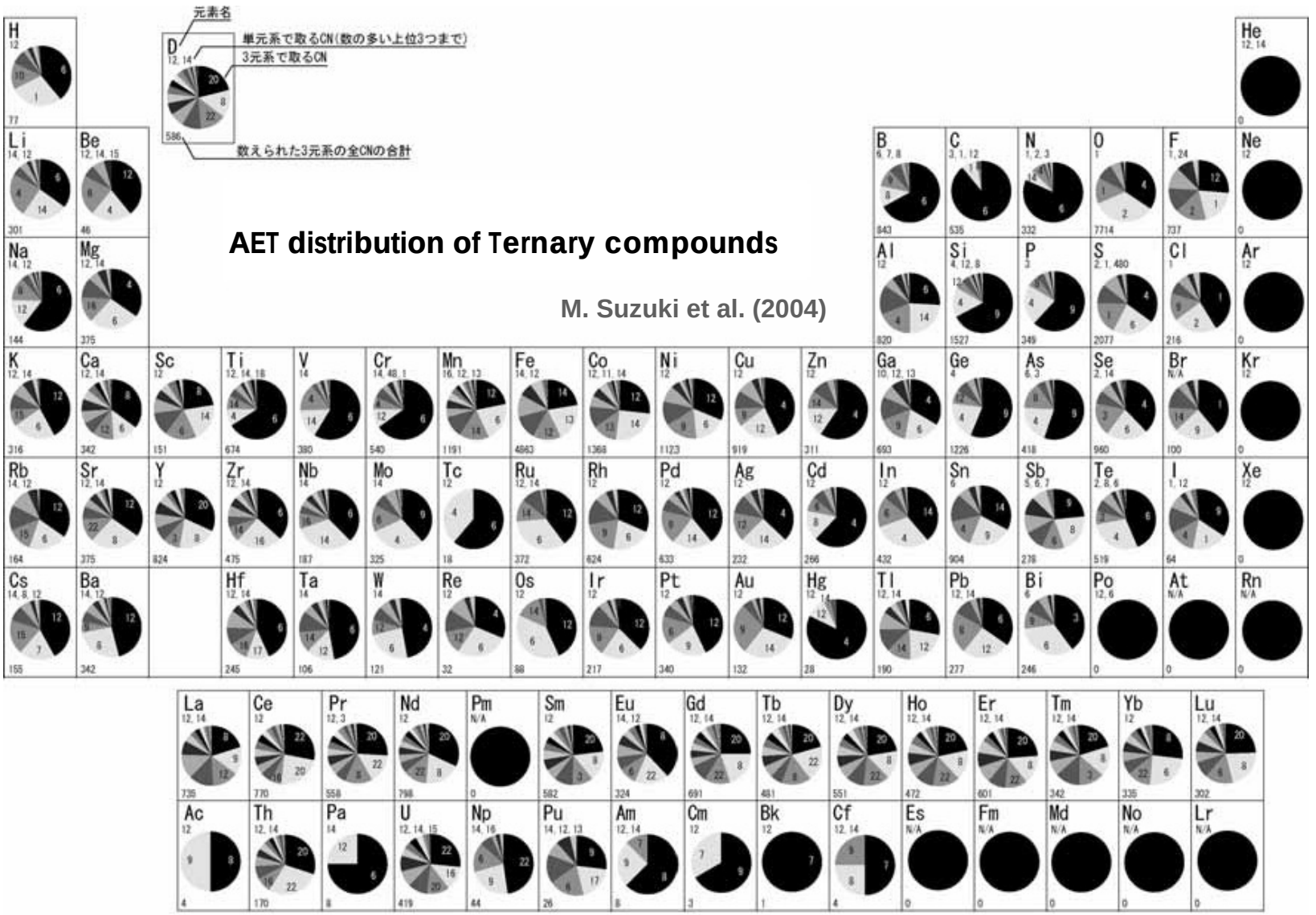
- CN 1-3
- CN 4-6 (-11)
- CN 12 (-13)
- CN 14(-18)
- Non-former
- Former (no 1:1)



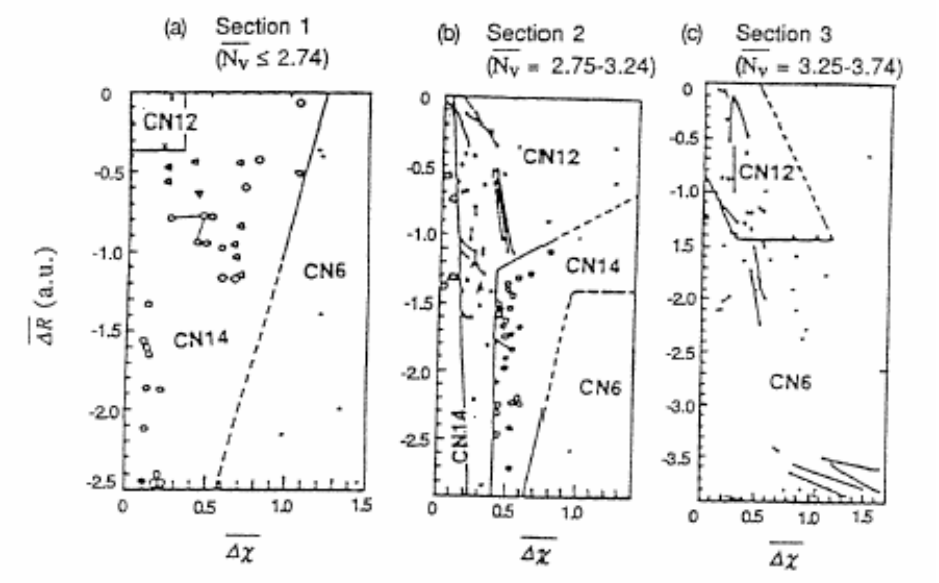


P. Villars, Y. Chen et al. (2004)

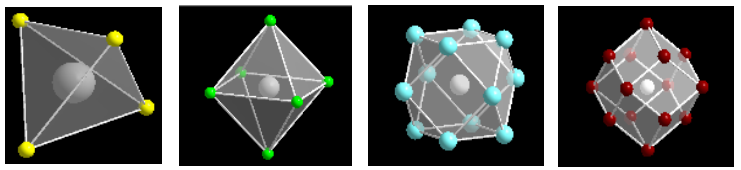
III-1 AET distribution of Ternary compounds



Successfully separated ~2,500 intermetallic compounds with single AET



4 most popular AETs



CN4 CN6 CN12 CN14

P. Villars at el. (1983)

x: difference of Zunger's radii
y: difference of M-B elentronegativity
z: sum of valence electrons number

Problems

- Microscopic mechanism ?
- Uncompleted separation

Theoretical Modeling:
Calculated map

Model Systems

AB intermetallic compound

- Nv < 3.5, s-s, s-p, p-d

- Martynov-Batsanov's electronegativity χ

$$\overline{\Delta\chi} = \chi_A - \chi_B$$

→ Atomic valence electronic energy level $\{\varepsilon_i\}$

$$\Delta E = E_A - E_B$$

- Pseudopotential radii's sum $R = r_s + r_p$

→ Bond length d

Calculation Approaches

Tight-binding approach $E_{tot} = E_{bond} + E_{rep}$

E_{bond} - attractive bond energy: $E_{bond} = \sum_{k}^{occ.} q_k \varepsilon_k$

$$H = \sum_{i\alpha} \varepsilon_{i\alpha} |i\alpha\rangle\langle i\alpha| + \sum_{i\alpha j\beta} h_{i\alpha j\beta} |i\alpha\rangle\langle j\beta| + |j\beta\rangle\langle i\alpha|$$

$$h_{\alpha\beta\gamma}(d) = \eta_{\alpha\beta\gamma} \frac{h}{md^2}$$

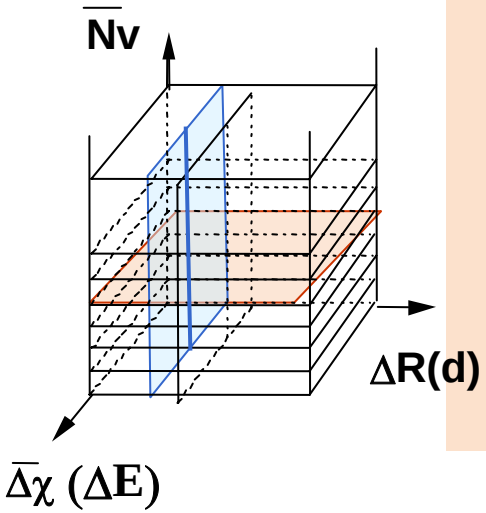
E_{rep} **- pairwise repulsive contribution**

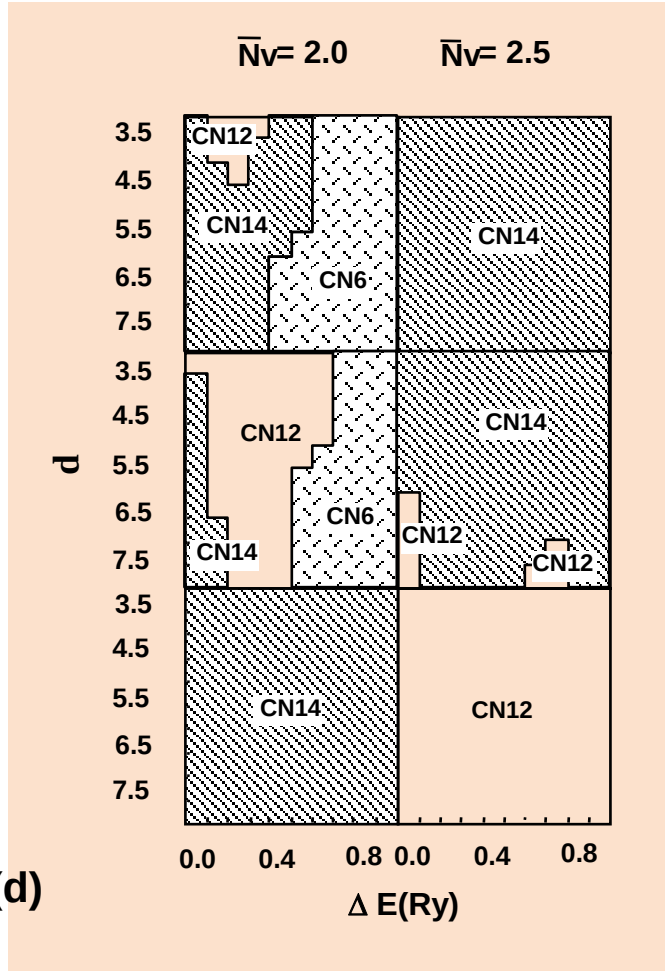
$$E_{rep} = \frac{1}{2N} \sum_{i,j} \Phi(R_{ij}) \qquad \mathbf{\Phi_{AA}(R) = h^{\lambda}(R)}$$

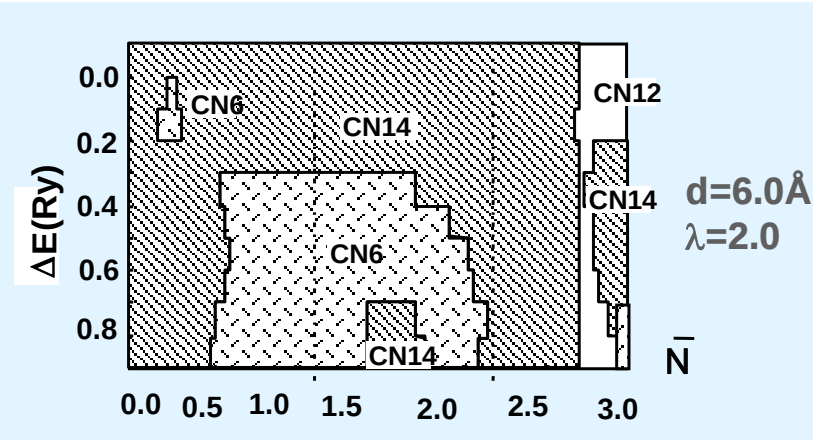
Structural energy difference theorem

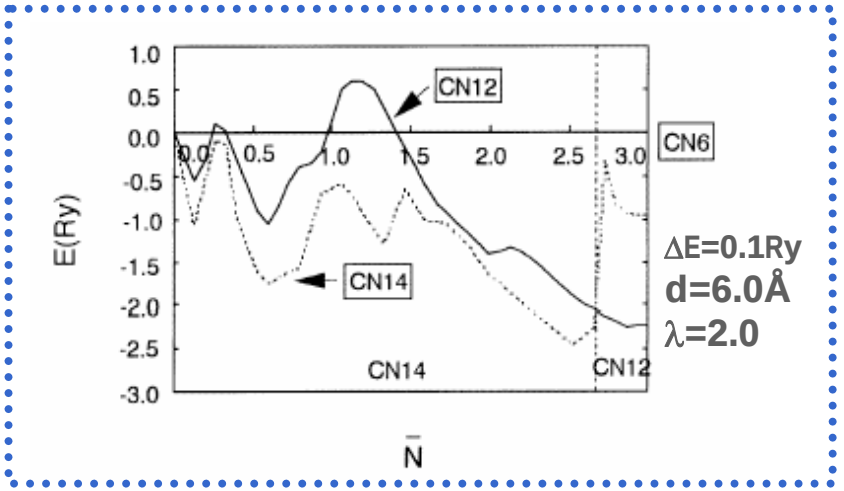
$$\Delta E_{tot} = (\Delta E_{bond})_{\Delta E_{erp}=0}$$

Y. Chen et al. (1996)

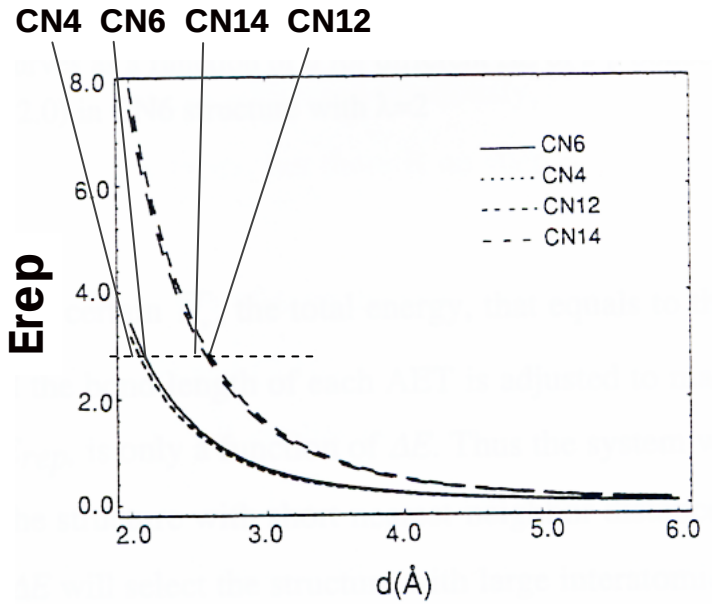

$\bar{N}v$
$\Delta R(d)$
$\bar{\Delta}\chi$ (ΔE)

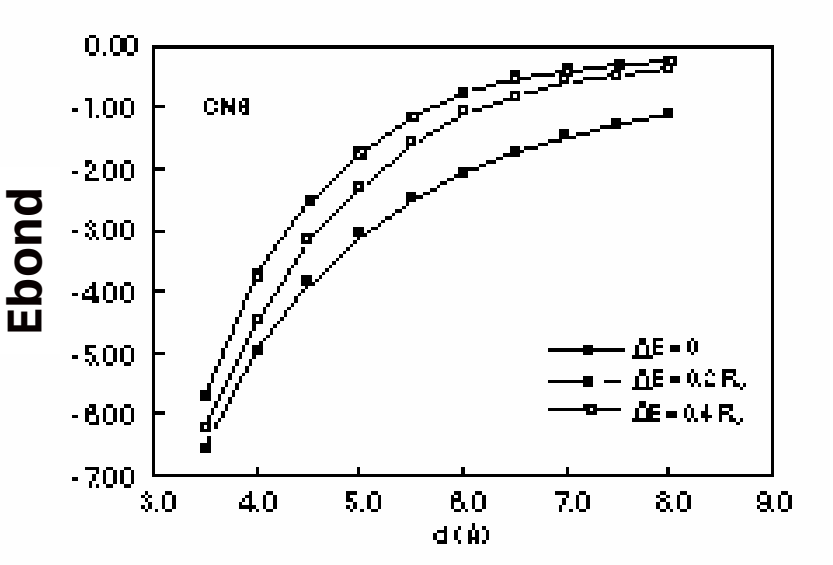

$\bar{N}v$= 2.0
$\bar{N}v$= 2.5
d
3.5
4.5
5.5
6.5
7.5
CN12
CN14
CN6
0.0
0.4
0.8
Δ E(Ry)


ΔE(Ry)
0.0
0.2
0.4
0.6
0.8
CN6
CN14
CN12
0.0
0.5
1.0
1.5
2.0
2.5
3.0
$\bar{N}$
d=6.0Å
λ=2.0


E(Ry)
1.0
0.5
0.0
-0.5
-1.0
-1.5
-2.0
-2.5
-3.0
CN12
CN6
CN14
CN14
CN12
0.0
0.5
1.0
1.5
2.0
2.5
3.0
$\bar{N}$
ΔE=0.1Ry
d=6.0Å
λ=2.0

Investigate Origin of structural preference





Influence of ΔE: large $\Delta E \rightarrow$ small $d \rightarrow$ CN4 and CN6

$\rightarrow$ CN4 requires very large ΔE, no in low Nv.

Influence of λ: higher $\lambda \rightarrow$ higher CN: CN12, CN14

III. Example-2: Discovery of New H-Storage Materials

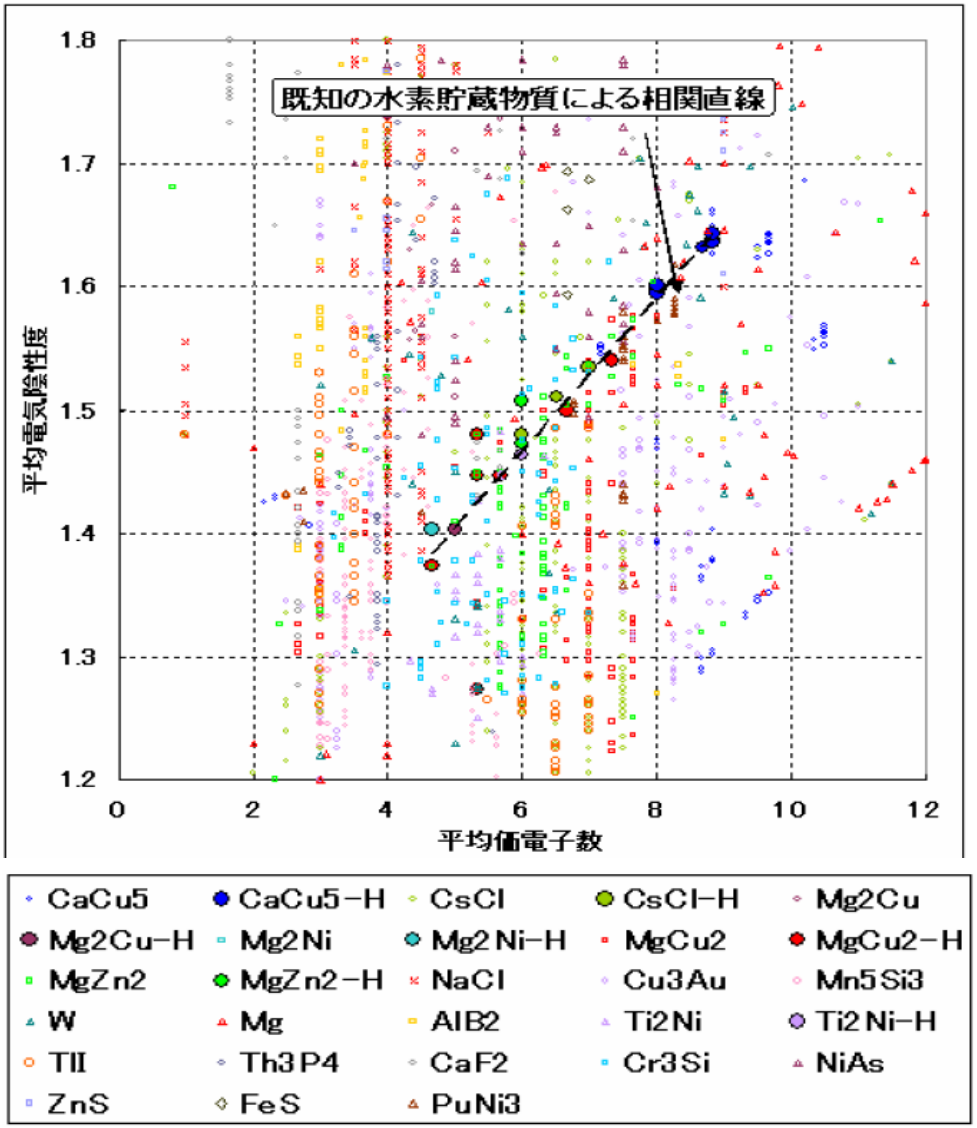
Starting point

32 Already known H-storage alloys (HAS)

結晶構造	水素貯蔵物質
$CaCu_5$	$LaNi_5$,$LaCo_5$,$CaNi_5$,$CeCo_5$, $CeNi_5$,$GdCo_5$,$GdNi_5$,$NdCo_5$, $NdNi_5$,$PrCo_5$,$PrNi_5$,YNi_5,YCo_5, $ThCo_5$,$YbNi_5$,$SmNi_5$,$SmCo_5$
CsCl	TiFe,TiNi,TiCo
Mg_2Cu	Mg_2Cu
Mg_2Ni	Mg_2Ni
$MgCu_2$	ZrV_2,$ZrCo_2$,$ZrMo_2$,$ZrFe_2$, $ZrCr_2$,$TiCr_2$,Ti_2Co
$MgZn_2$	$ZrMn_2$, $TiMn_2$
Ti_2Ni	Ti_2Ni

Surprising

linear relation for known HSAs
Average valence electrons
vs.
Average electronegativity



M. Ono et al. (2005)

III-2. Discovery of New H-Storage Materials

linear relation for known HSAs → Candidates of new HSAs

From small distance To the straight line

化学式	結晶構造	L	化学式	結晶構造	L
ErNi5	CaCu5,hP6,191	0.000	Nd5Sb3	Mn5Si3,hP16,193	0.001
ThNi5	CaCu5,hP6,191	0.000	Pr5Sb3	Mn5Si3,hP16,193	0.001
TmNi5	CaCu5,hP6,191	0.000	Sm5Sb3	Mn5Si3,hP16,193	0.001
TaFe2	MgZn2,hP12,194	0.000	Ce5Sb3	Mn5Si3,hP16,193	0.001
TaCo2	MgCu2,cF24,227	0.000	Yb5Sb3	Mn5Si3,hP16,193	0.001
La5Si3	Mn5Si3,hP16,193	0.000	HoFe5	CaCu5,hP6,191	0.001
NpFe2	MgCu2,cF24,227	0.001	Mn3Rh	Cu3Au,cP4,221	0.001
PuFe2	MgCu2,cF24,227	0.001	WFe2	MgZn2,hP12,194	0.001
UFe2	MgCu2,cF24,227	0.001	ScCo2	MgCu2,cF24,227	0.001
NdFe5	CaCu5,hP6,191	0.001	ErIn	CsCl,cP2,221	0.001
DyNi5	CaCu5,hP6,191	0.001	GdIn	CsCl,cP2,221	0.001
HoNi5	CaCu5,hP6,191	0.001	TmIn	CsCl,cP2,221	0.001
TbNi5	CaCu5,hP6,191	0.001	YIn	CsCl,cP2,221	0.001
HfMn2	MgZn2,hP12,194	0.001	LuNi5	CaCu5,hP6,191	0.001
CeBi	CsCl,cP2,221	0.001	Sc5Pb3	Mn5Si3,hP16,193	0.001
BaPo	NaCl,cF8,225	0.001	DyCo5	CaCu5,hP6,191	0.001
CeBi	NaCl,cF8,225	0.001	HoCo5	CaCu5,hP6,191	0.001
AmNi2	MgCu2,cF24,227	0.001	TbCo5	CaCu5,hP6,191	0.001
NpNi2	MgCu2,cF24,227	0.001	RuAl	CsCl,cP2,221	0.001
PuNi2	MgCu2,cF24,227	0.001	GdFe5	CaCu5,hP6,191	0.001
UNi2	MgZn2,hP12,194	0.001	ThFe5	CaCu5,hP6,191	0.001
NpCo2	MgCu2,cF24,227	0.001	ScFe2	MgCu2,cF24,227	0.001
PuCo2	MgCu2,cF24,227	0.001	ScFe2	MgZn2,hP12,194	0.001
UCo2	MgCu2,cF24,227	0.001			

Further screening by large unfilled volume

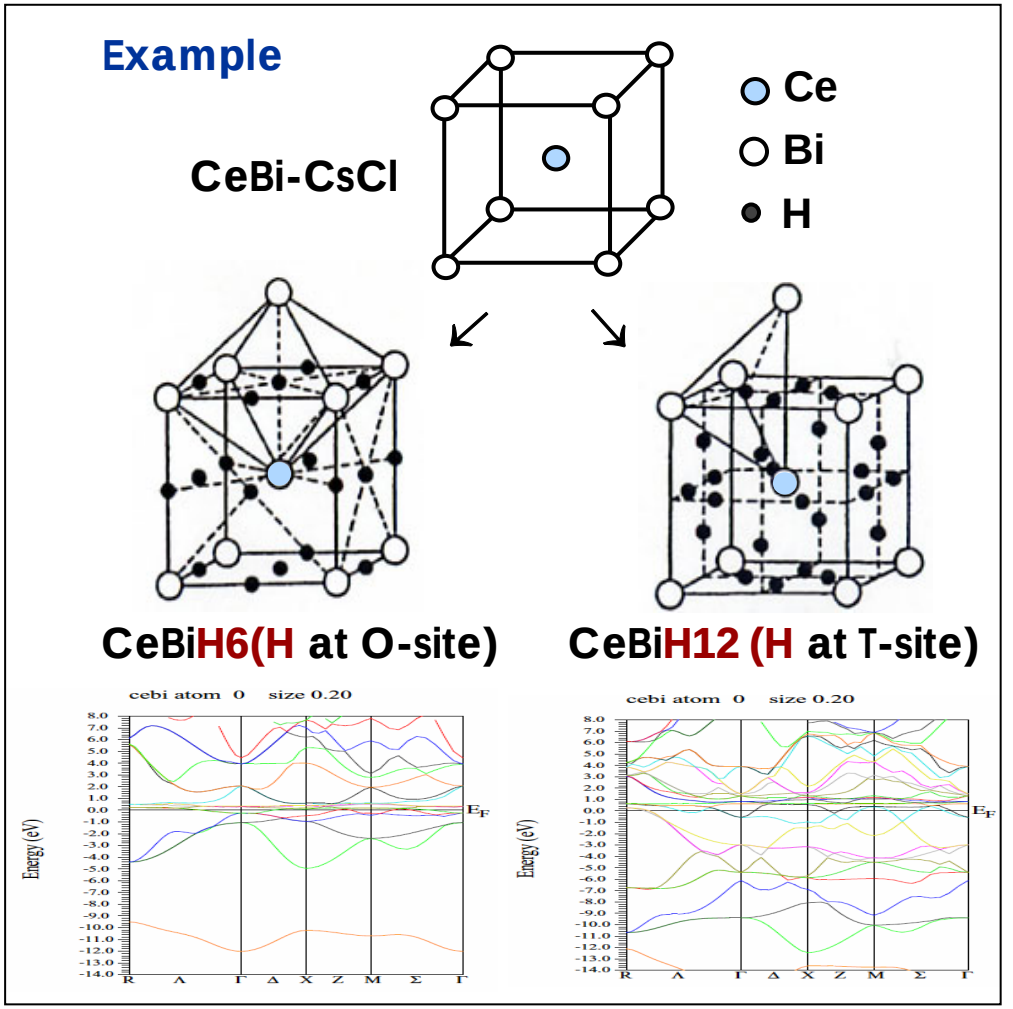
	結晶構造	隙間の体積 [Å^{3}Atom^{-1}]	直線との距離
$PuCo_3$	$PuNi_3$	4.36	0.0157
$PuNi_3$	$PuNi_3$	4.30	0.0120
$ThFe_3$	$PuNi_3$	4.12	0.0120
HfV_2	$MgCu_2$	4.37	0.0037
$NpFe_2$	$MgCu_2$	4.41	0.0108
$NpNi_2$	$MgCu_2$	4.25	0.0076
$PuFe_2$	$MgCu_2$	4.20	0.0120
$PuNi_2$	$MgCu_2$	4.23	0.0105

III-2. Discovery of New H-Storage Materials

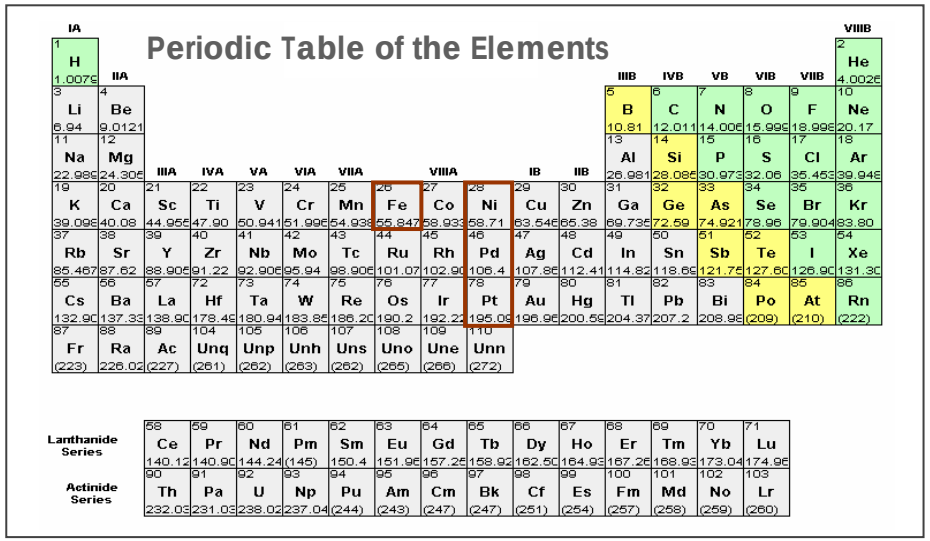
First Principles Calculation: A_nB_m , $A_nB_mH_q$

- Possible H sites, structures stability
- Interstitial volume, Storage capacity
- Electronic structure, binding
- Formation energy, Dissociation pressure

→ **Candidates of New H-storage alloys !**



III-3 Phase Equilibrium of Fe-Pt, Pd, Ni

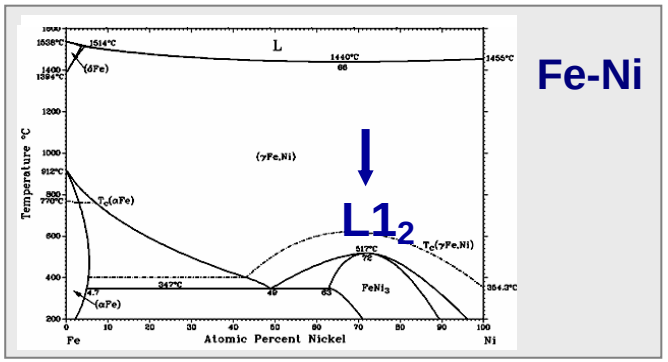


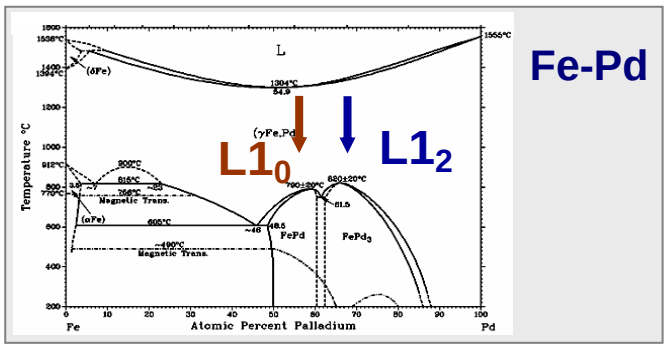
Fe-Ni, Pd, Pt

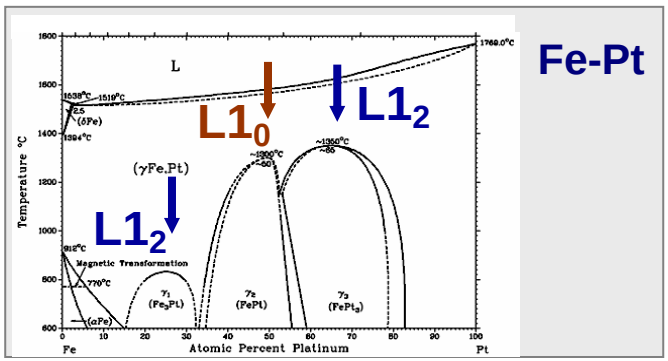
- Systematic variation of phase stability
- $L1_0$: Mechanical, Magnetic properties

Objectives

- Phase stability: Reproducing, Origin?





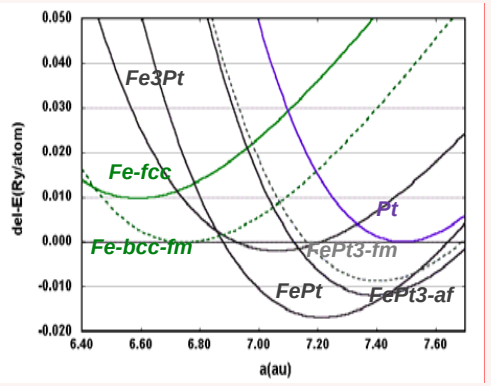


Y. Chen, T. MOhri (2002-2005)

III. Example 3: First Principles Cal. of Phase Diagram

Atomic Information

Internal Energy (First principles): E



Cluster Expansion Method (CEM)

$$E^{(k)}(r) = \sum_{i=1}^{m} v_i(r) \cdot \xi_i^k$$

$$v_i = \sum_{k=0}^{m-1} \left\{ \xi_j^k \right\}^{-1} \cdot E^{(k)}$$

Thermodynamics: $F = E - T \cdot S$

$$F(T, V(r), \{\xi_i\}) = \sum_j v_j(V(r), \{\xi_i\}, T) - T \cdot S(\{\xi_i\})$$

Cluster Variation Method (CVM): S

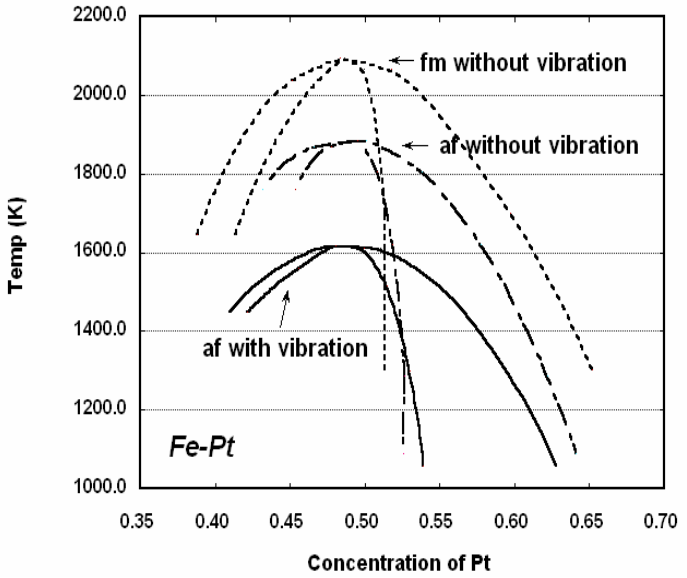
$$S = k_B \cdot \ln \frac{\prod_{i,j} \{Ny_{ij}!\}^6 \cdot \{N!\}}{\prod_i \{Nx_i!\}^5 \cdot \prod_{i,j,k,l} \{Nw_{ijkl}!\}^2}$$

Phase Equilibria, Thermo-dynamic properties

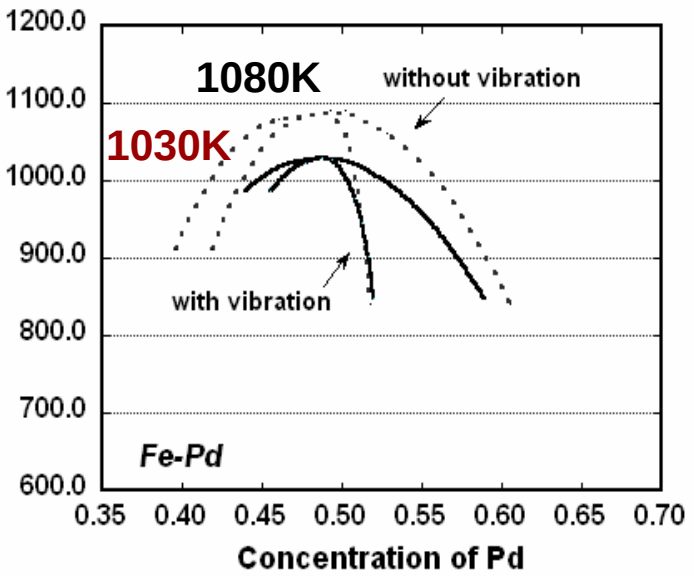
III-3 Phase Equilibrium of Fe-Pt, Pd, Ni

Phase Diagram: L10-disorder, Phase Separation

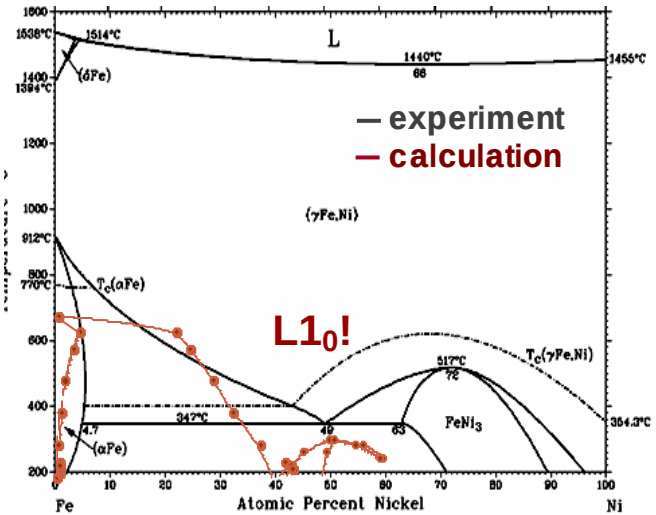
Fe-Pt



Fe-Pd



Fe-Ni



Experiment 1600K

Experiment 1023K

No L10 in exp. Phase diagram: Stable L10?

Exp.(1962), MC(1996): 590K～775K

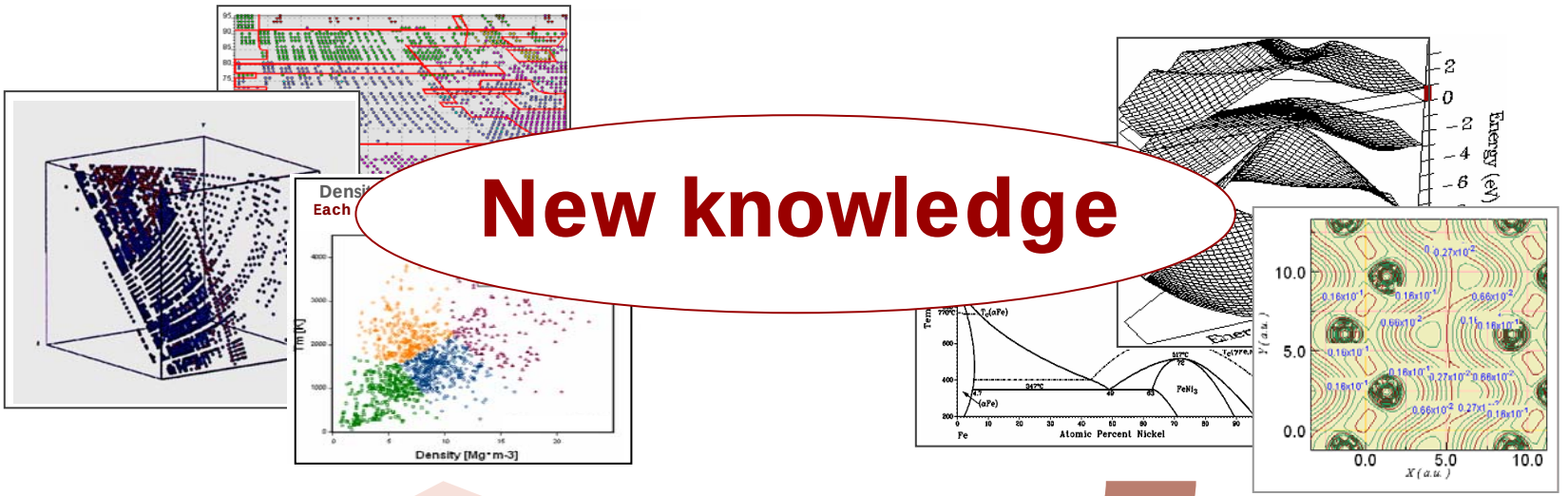
Ohuma et al. ASM meeting 2004

IV. Summary

Interaction of Data & Modeling

Data Mining
→ Regularity

Modeling
→ Physical Origin



New knowledge


Thank you very much !

Element Properties (56 properties, …182 data sets)

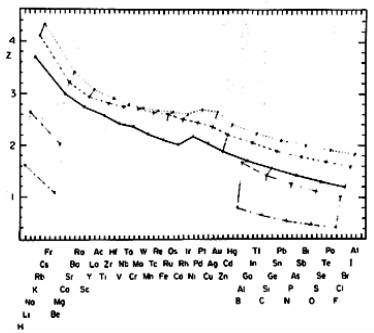
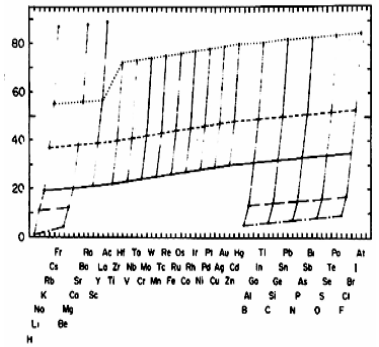
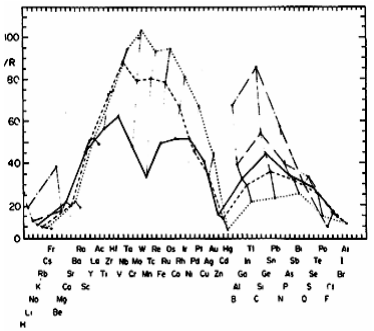
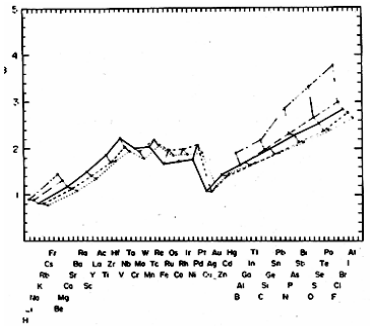
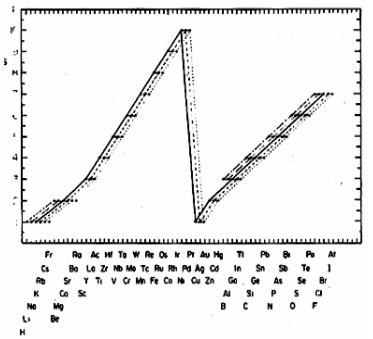
atomic number
atomic weight
density
molar volume
atomic energy level (Herman-Skillman)
valence electron number
atomic radius
Van der Waals radius
covalent radius
first ionic radius
second ionic radius
third ionic radius
metallic radius
Zunger pseudo-potential radius
Pauling electronegativity
Alfred-Rochow electronegativity
Martynov-Batsanov electronegativity
absolute electronegativity
first ionization energy
second ionization energy
third ionization energy
molar electronic affinity

Young's modulus
rigidity modulus
bulk modulus
Poisson's ratio
mineral hardness
Brinell hardness
melting temperature
Vickers hardness
boiling temperature
Debye temperature
thermal conductivity
molar heat capacity
coefficient of linear thermal expansion
atomization enthalpy
fusion enthalpy
vaporization enthalpy
electrical resistance
magnetic susceptibility
reflectivity
refractive index
…

Optimal coordinate for certain problem?

Element property vs. Atomic number

→ 5 Patterns
→ 5 Element property groups
→ 1 representative for 1 group

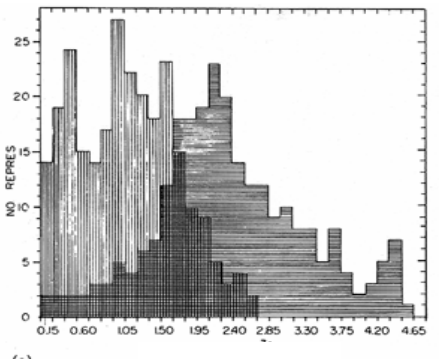
Size factor	Atomic No. factor	Cohensive energy factor	Electro-chemical factor	Valence electron factor
Zunger's pseudopotential radii (r_s, r_p, …)	Atomic No.	Atomization energy	Electro-negativity	valence electron

Expression of compound property by element property

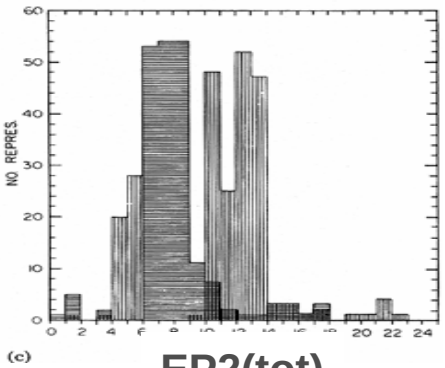
EP(tot) = EP(A) op EP(B)

Operations		
	Sum	EP(A)+EP(B)
	Difference	EP(A)-EP(B)
	Product	EP(A)*EP(B)
	Ratio	EP(A)/EP(B)
	Maximum	Max(EP(A),EP(B))
	Minimum	Min(EP(A),EP(B))

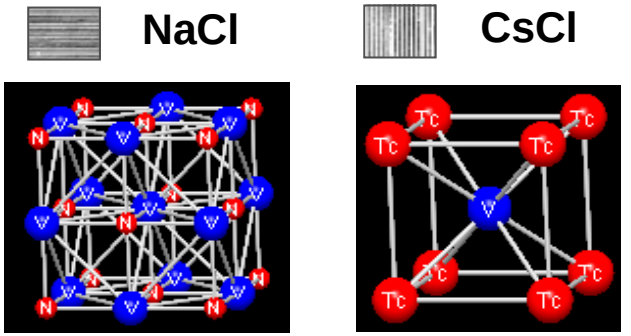
Test separating capacity for NaCl-type and CsCl-type structures



EP1(tot)



EP2(tot)



31 Most common Atomic Environment Types (AETs)