Status of Interatomic Potentials Models for Hydrogen Energy Applications

SAND2018-2004C

European Advanced Energy Materials and Technology Congress
Stockholm, Sweden, March 25-28, 2018

X. W. Zhou, R. B. Sills, M. E. Foster, R. A. Karnesky, M. D. Allendorf, V. Stavila
[1]Sandia National Laboratories, USA
T. W. Heo, B. C. Wood, S. Kang
[2]Lawrence Livermore National Laboratory, Livermore, California 94550, USA

Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525. Lawrence Livermore National Laboratory is operated for the US Department of Energy under Contract DE-AC52-07NA27344. The authors gratefully acknowledge research support from the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Fuel Cell Technologies Office, under Contract Numbers DE-AC04-94AL85000 and DE-AC52-07NA27344.

Outline

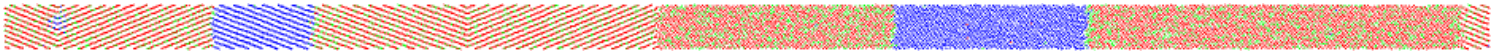
1. Embedded-atom method potential for Fe-Ni-Cr-H
2. Analytical bond order potential for Al-Cu-H
3. Analytical bond order potential for Mg-H

Four criteria of fidelity of Fe-Ni-Cr interatomic potential

1. permit stable high temperature MD simulations
2. capture the correct stacking fault energy (γ_{sf})
3. prescribe well the elastic constants
4. give reasonable energy and volume for various compositions

Issues of literature potentials

1. The potential we published (CALPHAD 1993, 17, 383) did not consider the four criteria
2. Smith and Was' potential (PRB 1989, 40, 10322) was fitted to effective atoms and did not consider stacking fault energy
3. The 2013 version of Bonny et al's potential (MSMSE 2013, 21 085004) incorrectly predicts phase separation



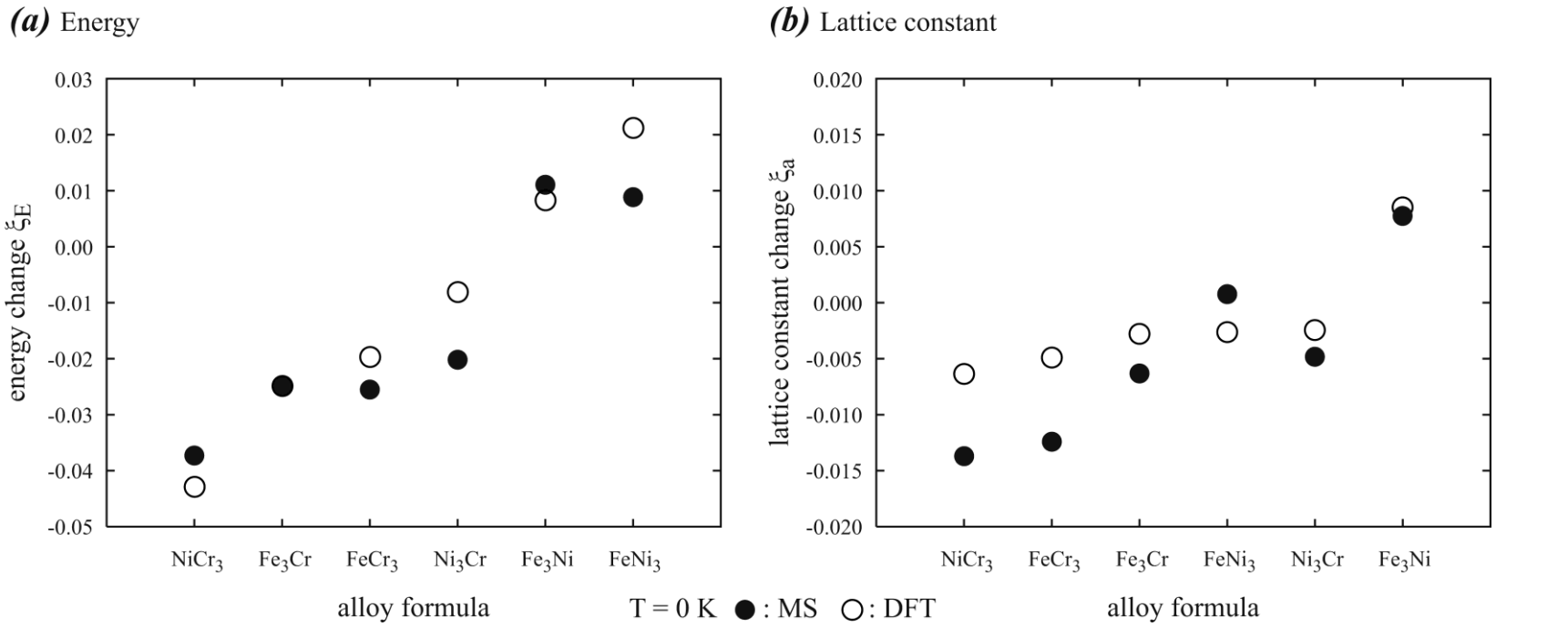
4. The 2011 version of Bonny et al's potential (MSMSE 2011, 19, 085008) incorrectly predicts negative slope of stacking fault energy with Ni composition
5. Tong et al's potential (Mol. Sim. 2016, 42, 1256) incorrectly predicts large negative stacking fault energy (~ -200 mJ/m^2)

Sandia National Laboratories

Current status of our Fe-Ni-Cr-H embedded-atom method potential

.....

Energy and volume trends

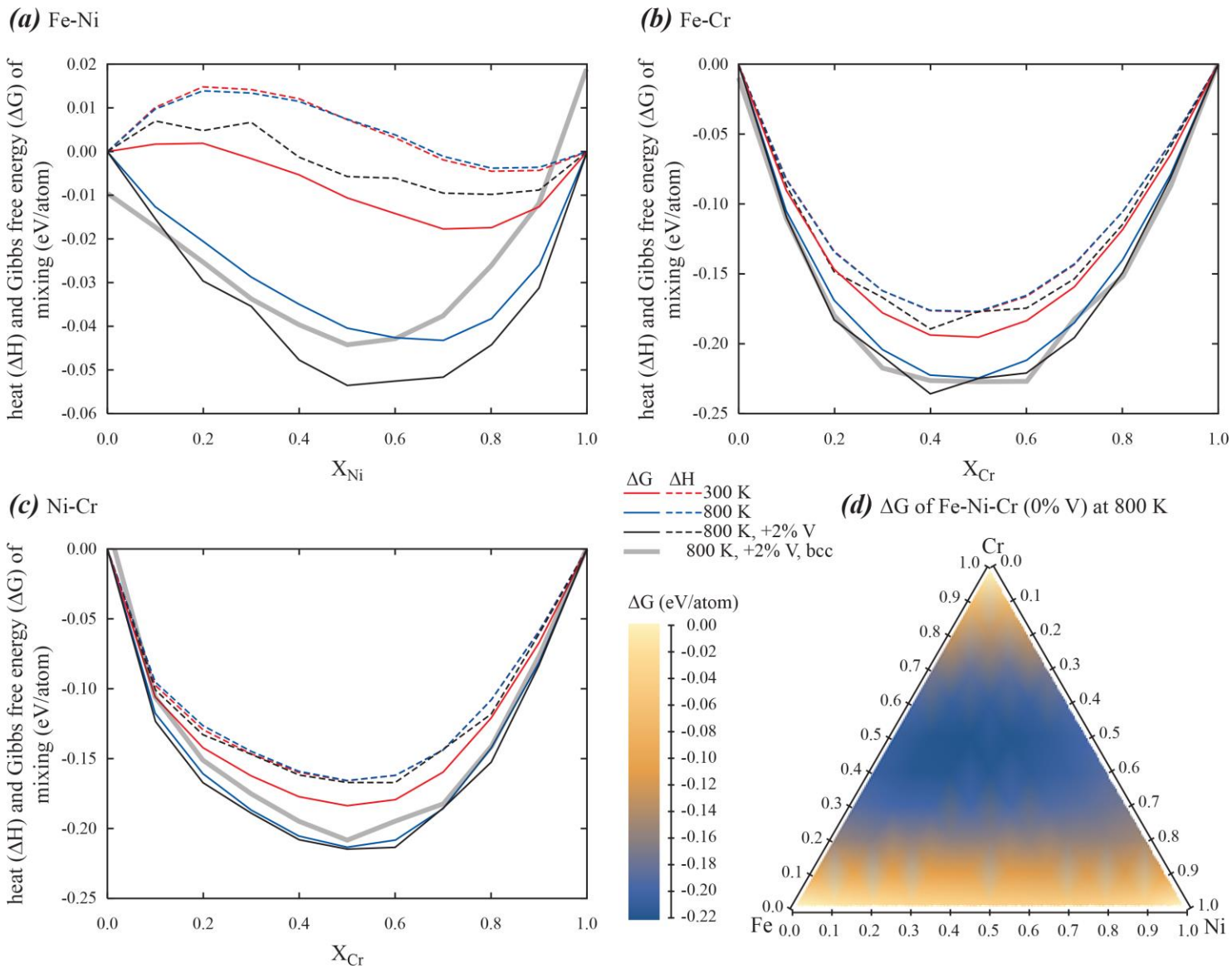


Swelling parameter comparison with experiments

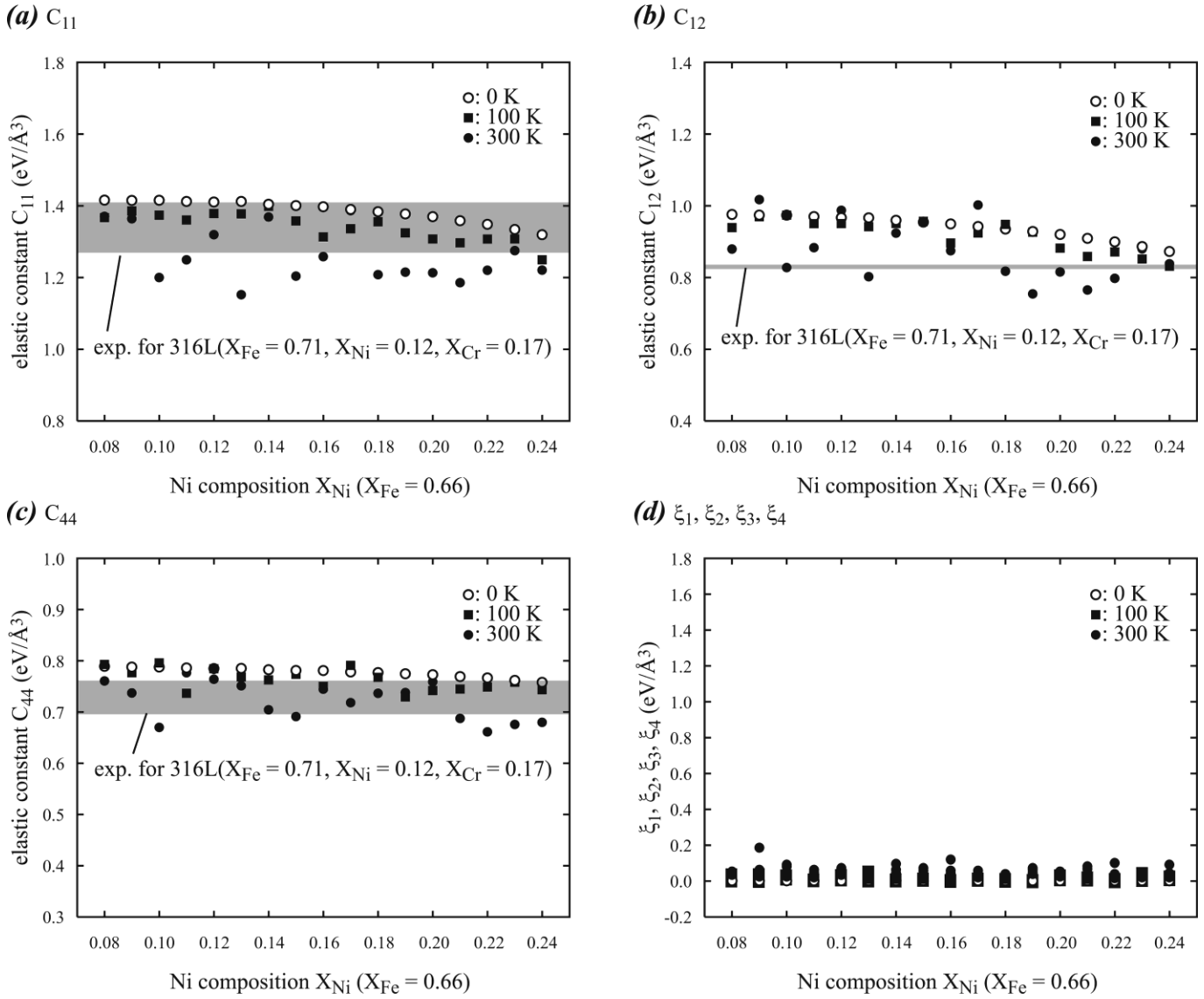
solute	fcc Fe		bcc Fe		
	MS	DFT	MS	DFT	exp.
Ni	3.46%		10.09%		4.65%
Cr	-20.82%		8.06%		4.36%

Experimental data from King, J. Mater. Sci. 1966, 1, 79

Heats and Gibbs free energy of solution

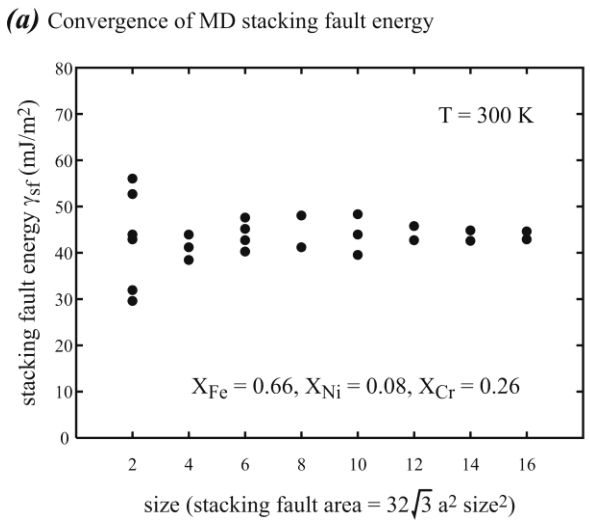


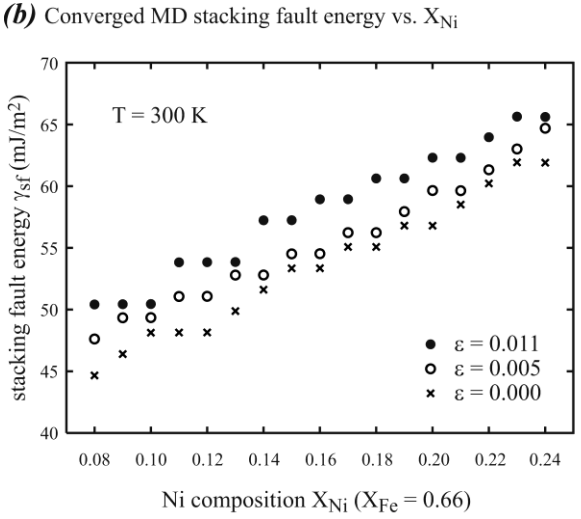
Elastic constants

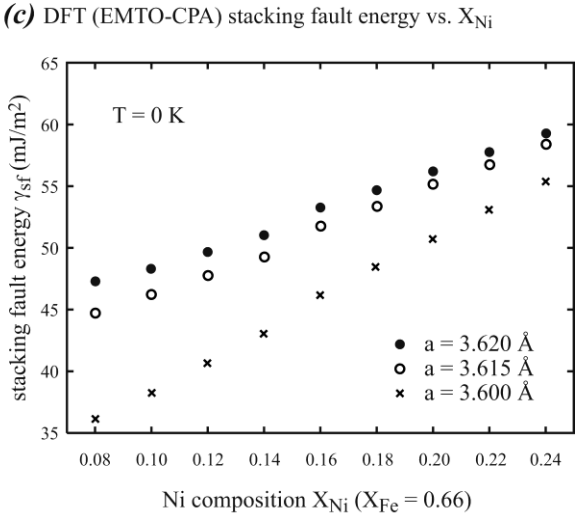


Experimental data for 316L from Ledbetter, Ultrasonics 1985, 23, 9; Bonny et al, MSMSE 2011, 19, 085008; Bonny et al, MSMSE 2013, 21, 085004.

Stacking fault energy

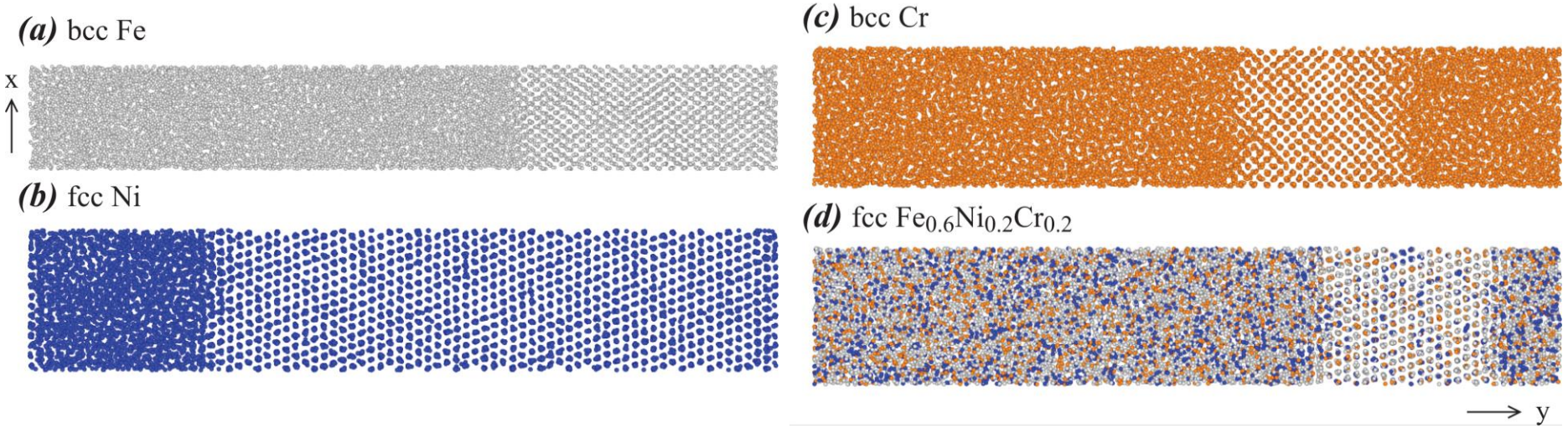






The predicted stacking fault energies match well with experimental results (see, for example, Vitos et al, PRL 2006, 96, 117210, and references therein).

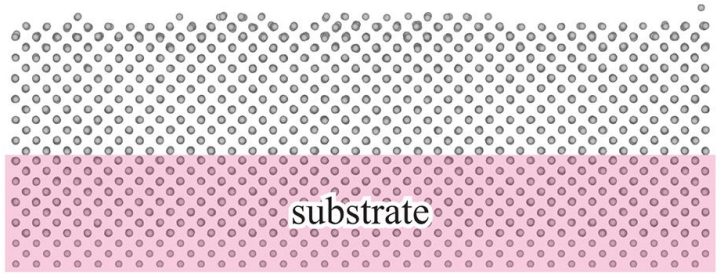
Melting



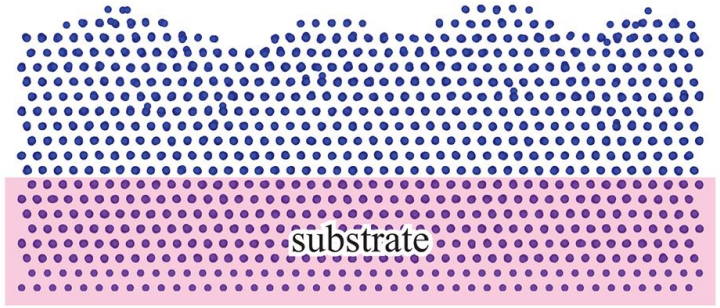
Growth simulations

Sandia National Laboratories

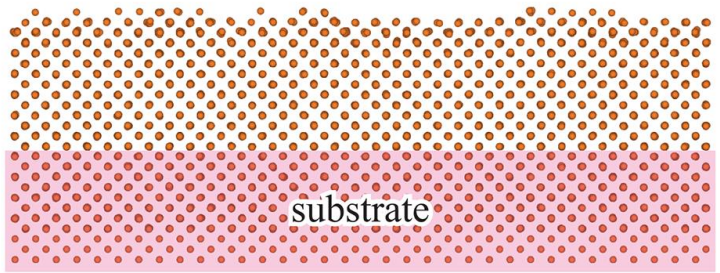
(a) Fe on bcc Fe, atom map



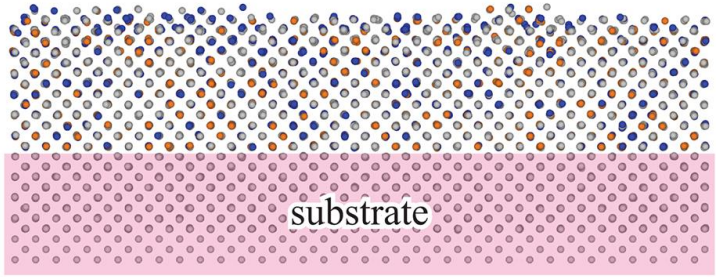
(b) Ni on fcc Ni, atom map



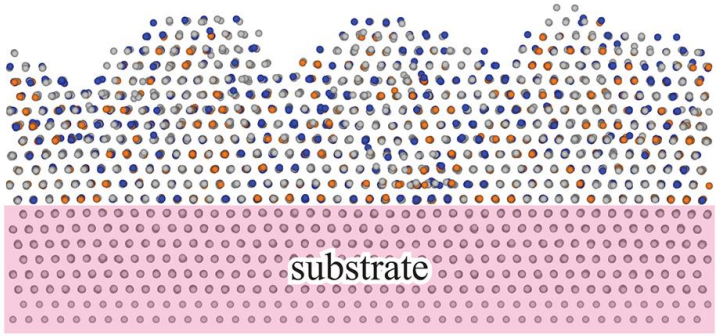
(c) Cr on bcc Cr, atom map



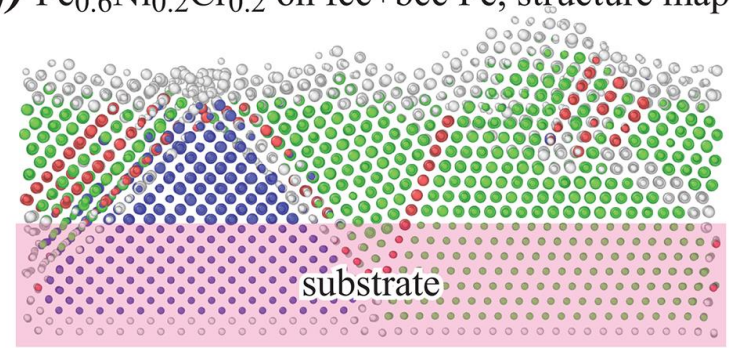
(d) $Fe_{0.6}Ni_{0.2}Cr_{0.2}$ bcc Fe, atom map



(e) $Fe_{0.6}Ni_{0.2}Cr_{0.2}$ on fcc Fe, atom map



(f) $Fe_{0.6}Ni_{0.2}Cr_{0.2}$ on fcc+bcc Fe, structure map



atom: Fe Cr Ni

structure: fcc bcc hcp undefined

bcc: x [100], y [010], z: [001] fcc: x [11$\bar{2}$], y [111], z [1$\bar{1}$0]

1 nm

T = 300 K, E_i = 0.1 eV, R ~ 0.5 nm/ns

Current status of our Al-Cu-H analytical bond order potential

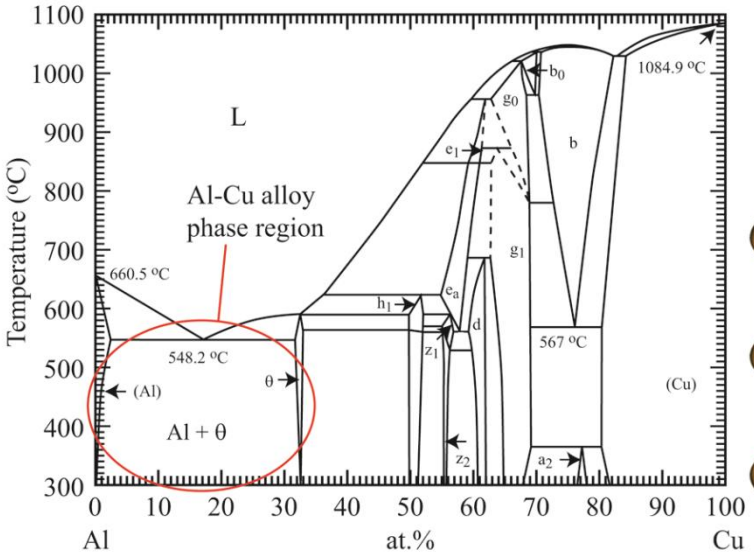
……

1. X. W. Zhou, D. K. Ward, and M. E. Foster, J. Alloys Compds. 2016, 680, 752;
2. X. W. Zhou, D. K. Ward, M. Foster, J. A. Zimmerman, J. Mater. Sci., 2015, 50, 2859.

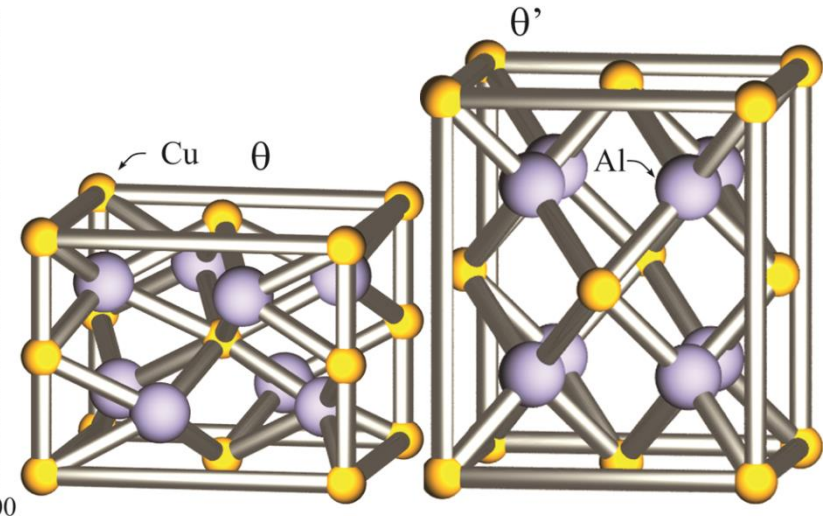
Wish list for Al-Cu-H potential

1. A high stacking fault energy of Al observed in experiments;
2. Properties trends of a variety of stable and metastable structures;
3. Al-rich side of the Al-Cu phase diagram;
4. A reasonable positive heat of solution of Cu in Al;
5. H2 $\Leftrightarrow$ 2H chemical reaction;
6. $Al_{1-x}H_x \rightarrow Al + H_2$ and $Cu_{1-x}H_x \rightarrow Cu + H_2$ phase separations;
7. Robust MD simulations.

(a) Al-Cu phase diagram



(b) crystal structure of the θ and θ' phases



Stacking fault energy of Al

Model/Exp.	γ_{100}	γ_{110}	γ_{111}	γ_{sf}
EAM-CY	583	631	526	1
EAM-Mishin1	947	1013	873	141
EAM-BAM	1017	1154	1003	85
EAM-VC	862	969	829	71
EAM-MSAH	194	328	138	126
EAM-Zhou	868	958	832	44
EAM-MKBA	495	582	427	125
EAM-JNP	977	1055	910	0
MEAM	903	944	599	141
REAX-LJGS	481	483	427	0
REAX- Ojwang	810	848	711	1
BOP	979	1069	850	133
DFT [38]	1063	1098	987	------
Exp. [59,60,61,62]	980-1140	980-1140	980-1140	120-144

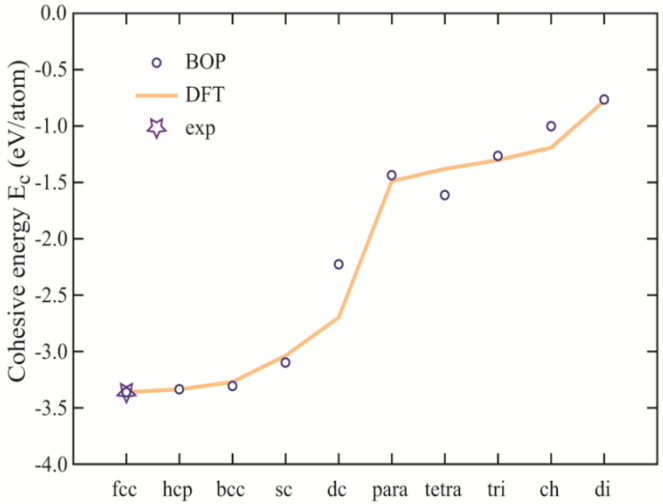
BOP captures a high stacking fault energy of Al.

Property trends of Al and Cu

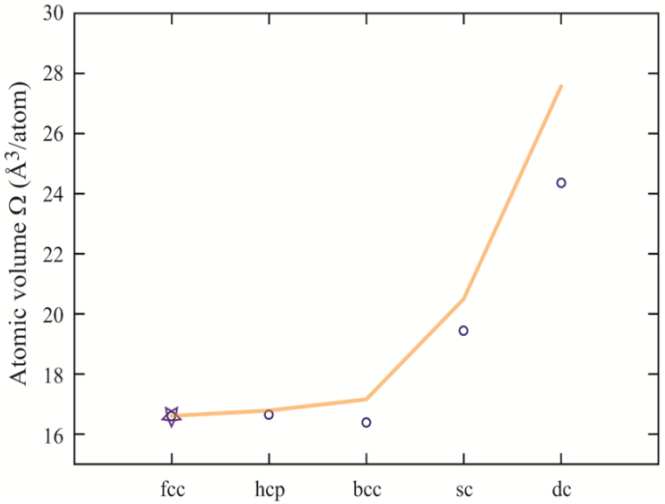
Sandia National Laboratories

Al

(a) Cohesive energy E_c

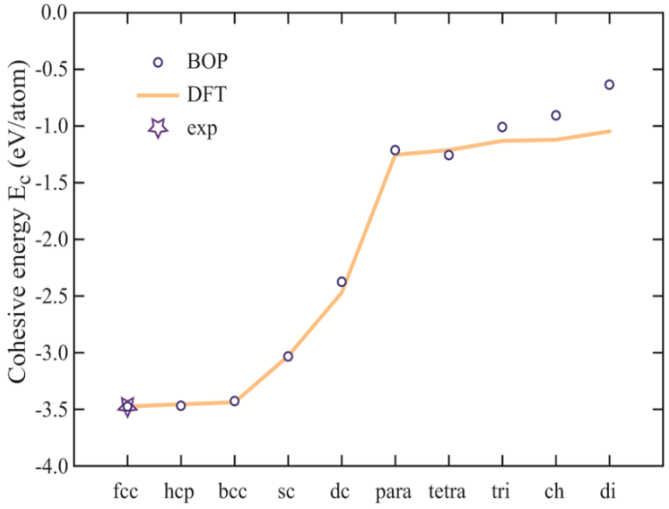


(b) Atomic volume Ω

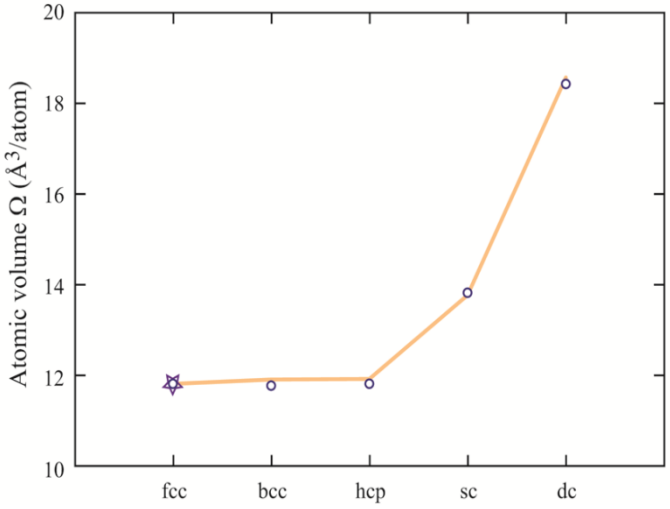


Cu

(a) Cohesive energy E_c

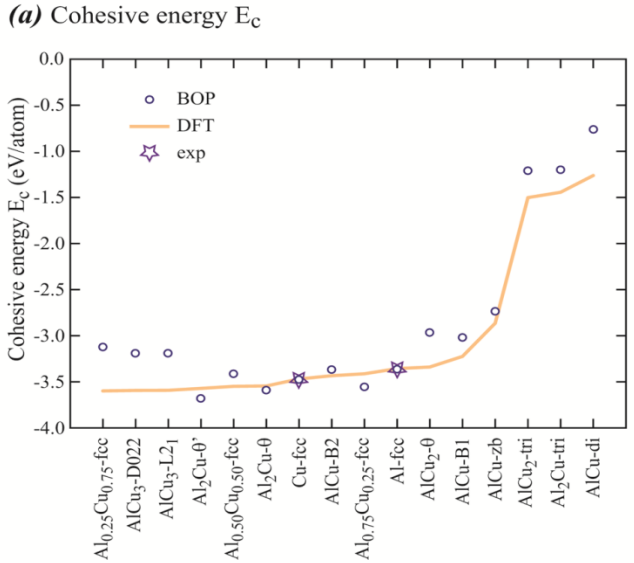


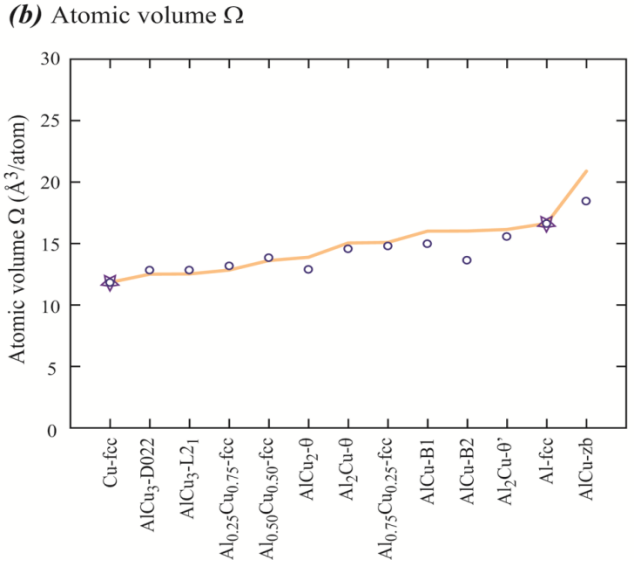
(b) Atomic volume Ω



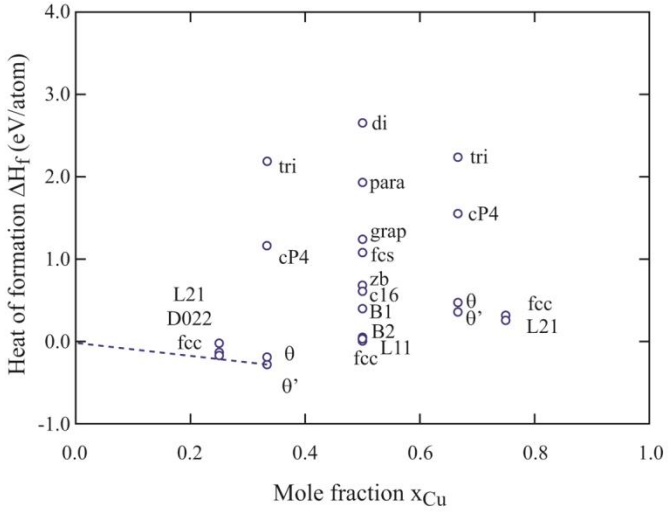
Pretty good property trends for Al and Cu.

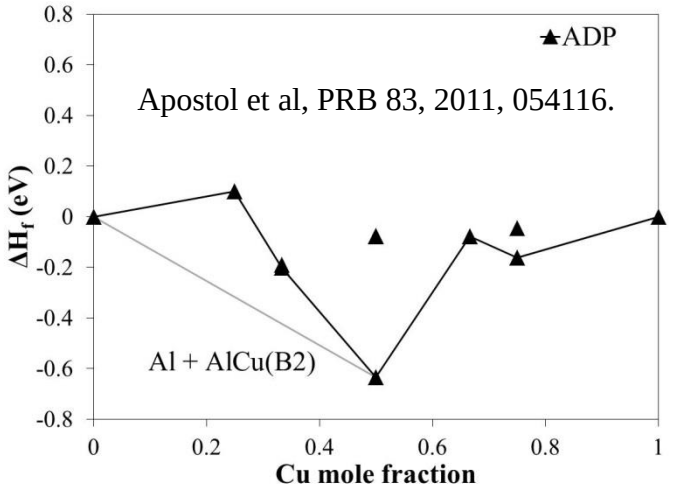
Property trends of Al-Cu





Heat of formation vs. composition



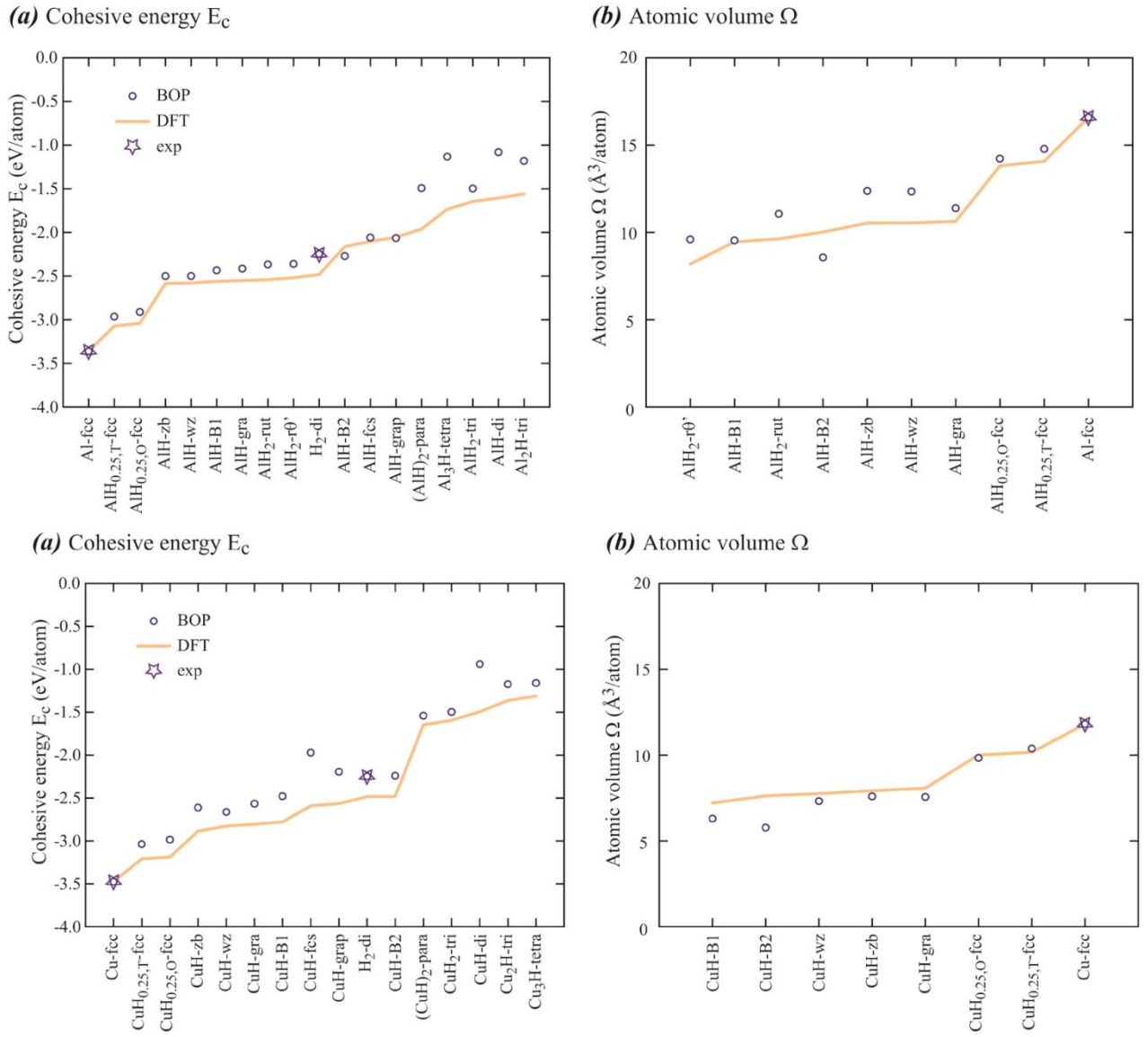


Pretty good property trends for AlCu.

Property trends of Al-H and Cu-H

Sandia National Laboratories

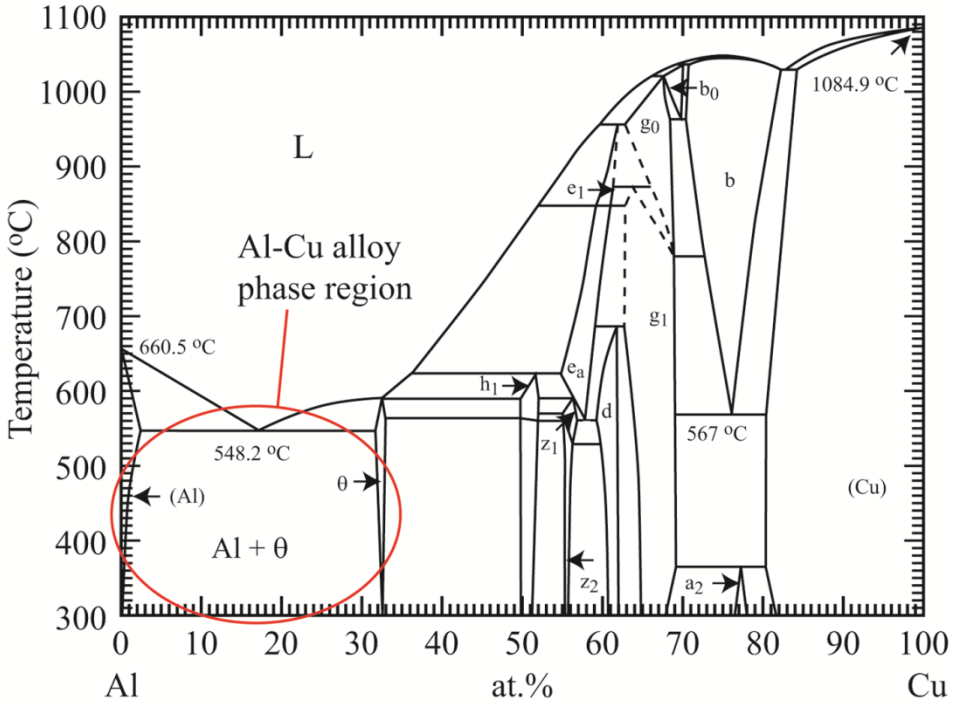
AlH

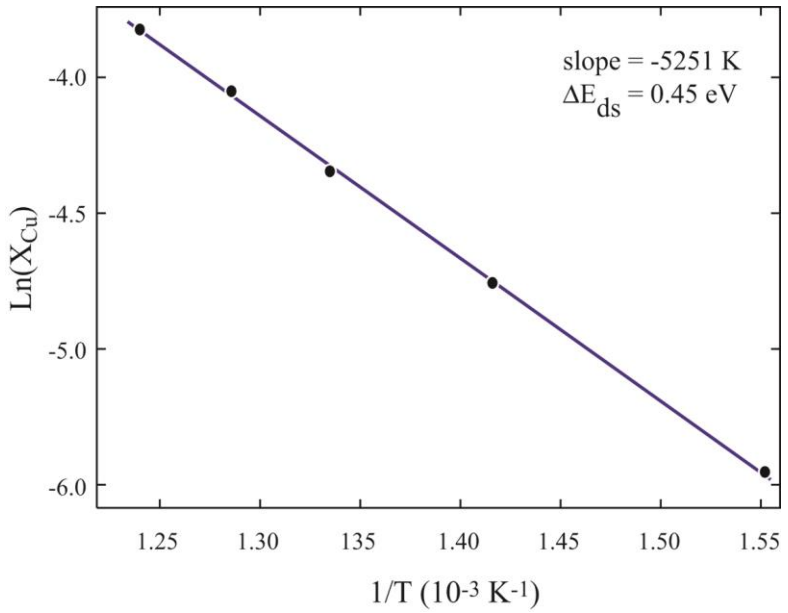


CuH

Pretty good property trends for AlH and CuH.

Dilute heat of solution of Cu in Al





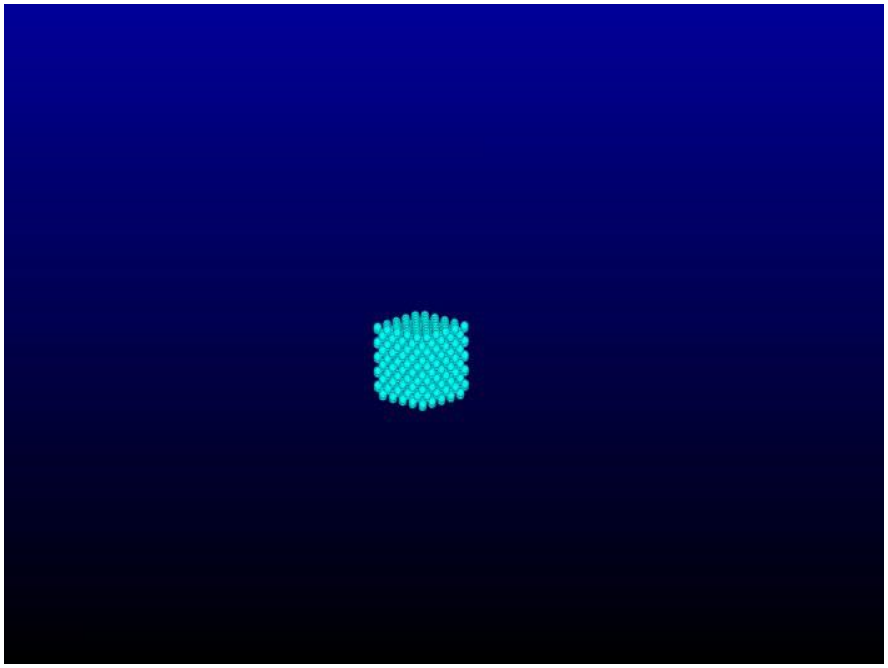
1. Traditionally, heat of solution of Cu in Al is taken as energy change due to taking a Cu atom from Cu pool and putting it in Al pool;
2. Should really be the energy change due to taking a Cu atom from Al_2Cu pool and putting it in Al pool;
3. Must be a positive number

Literature comparison

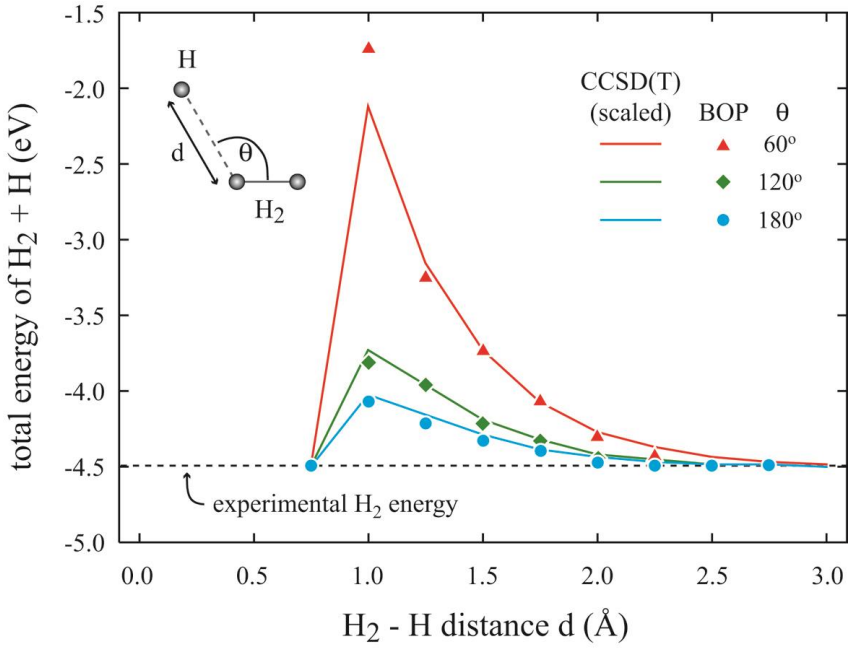
ADP	1.09
EAM-CY	-0.06
BOP	0.14
DFT	0.40
Exp.	0.45

$H_2+H \rightarrow H+H_2$ chemical reaction

Hydrogen crystal to H_2 gas



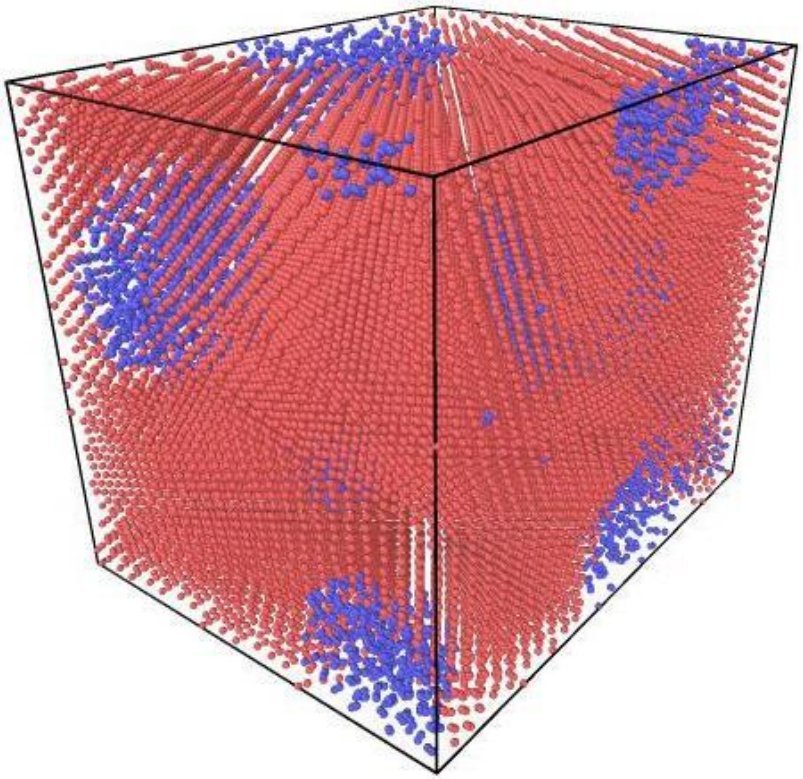
$H_2+H \rightarrow H+H_2$ energy profiles



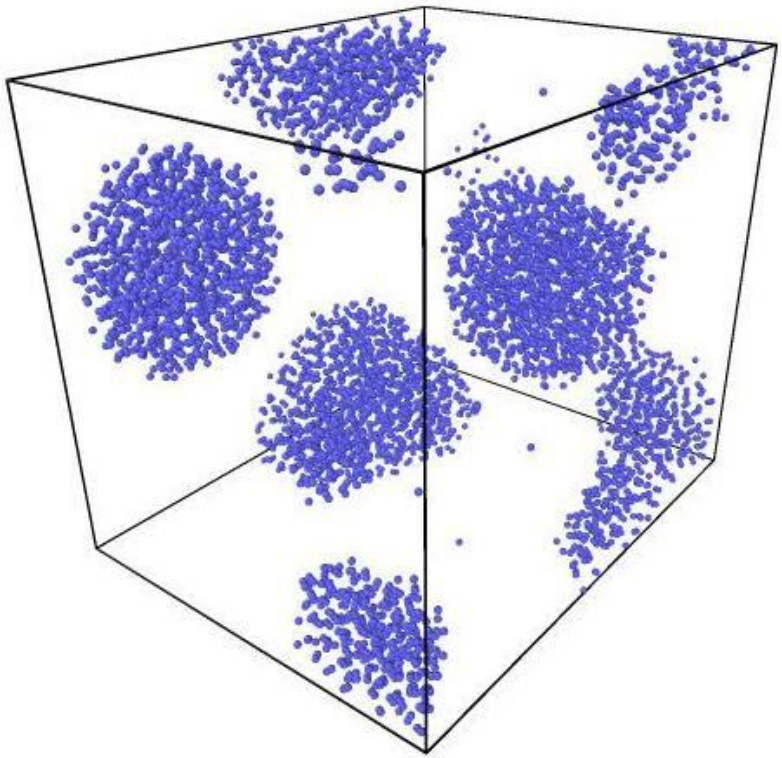
Our BOP captured the $H_2+H \rightarrow H+H_2$ reaction

Al + H_2 and Cu + H_2 Phase Separation

(a) N_H/N_{Cu} = 0.20 with both Cu and H shown



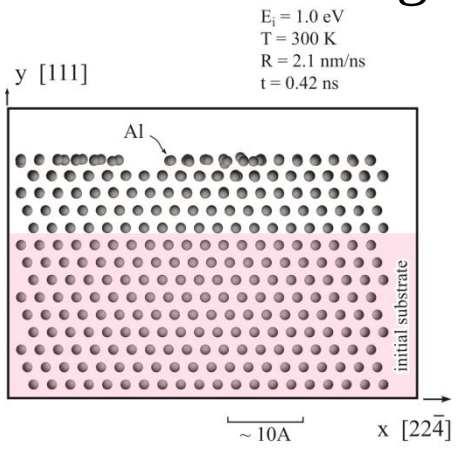
(b) Only H shown

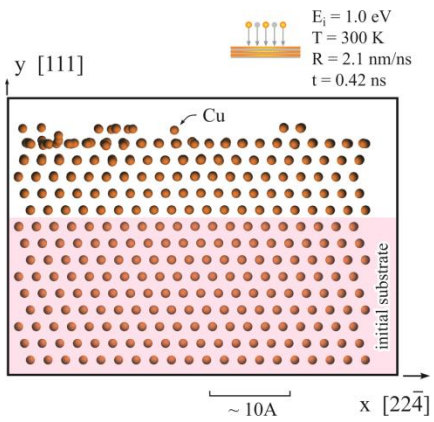


MD growth simulations

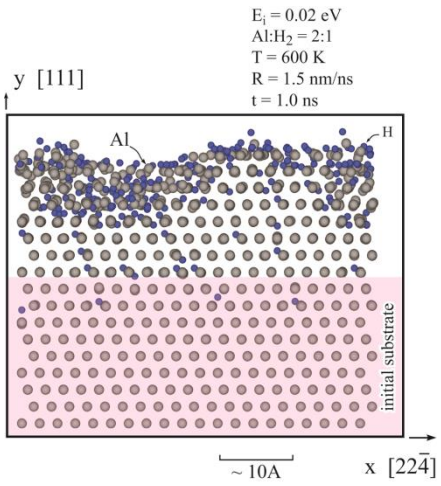
Sandia National Laboratories

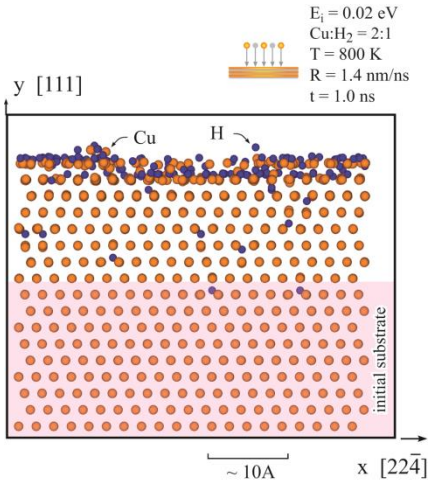
Al and Cu growth without H



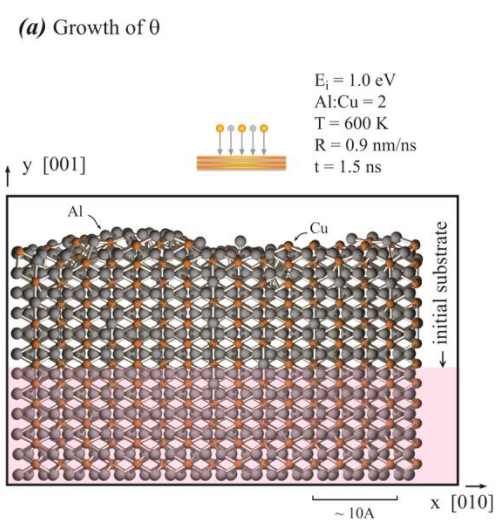


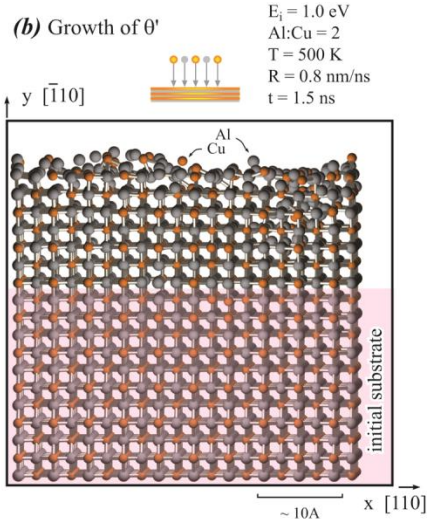
Al and Cu growth with H



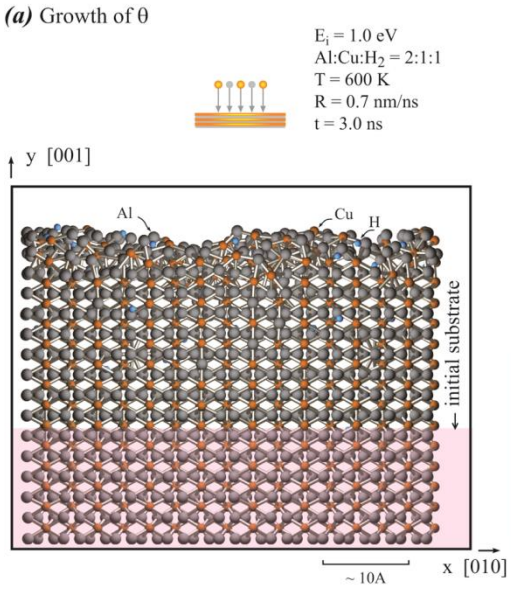


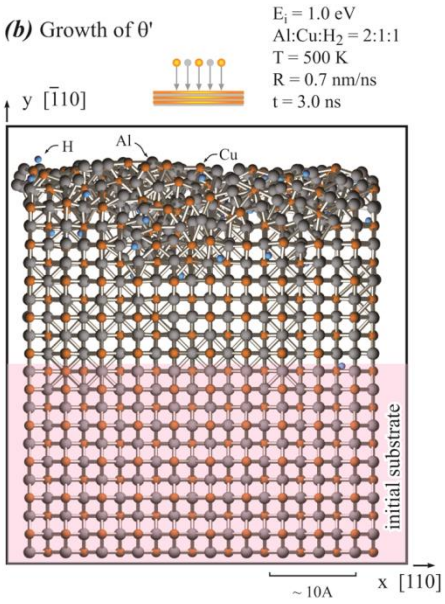
Al_2Cu (θ and θ') growth without H





Al_2Cu (θ and θ') growth with H





Sandia National Laboratories

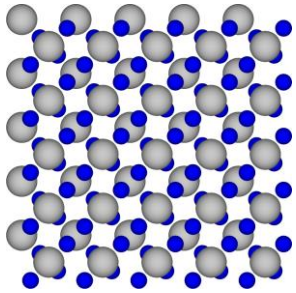
Current status of our Mg-H analytical bond order potential

.....

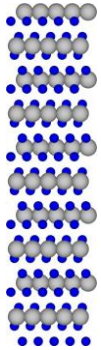
Issues of literature potentials

1. Literature Mg-H **EAM** potential developed by Ruda et al (ANALES DE LA ASOCIACION QUIMICA ARGENTINA, 84, 393, 1996) cannot maintain the correct MgH_2 crystal structure even during 0 K;

Rutile Crystal for MgH_2

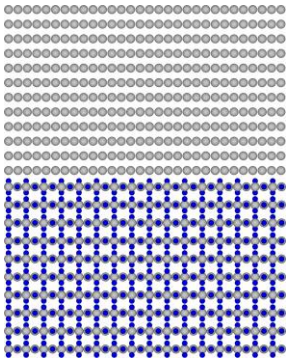


Molecular Statics At 0 K ⇒

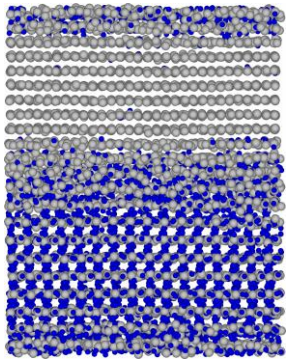


2. Literature Mg-H **ReaxFF** potential developed by Cheung et al (S. Cheung, W. Q. Deng, A. C. T. van Duin, W. A. Goddard, J. Phys. Chem., 109, 851 2005) cannot maintain the correct MgH_2 crystal structure during 300 K molecular dynamics (MD) simulations;

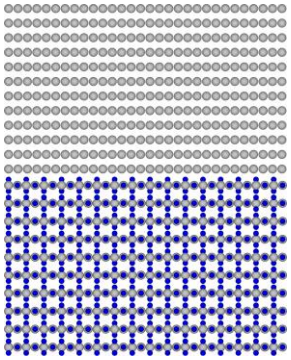
$(110)MgH_2$ // (0001) Mg, [110] MgH_2 // [2-1-10] Mg
Phil. Mag. Lett., 90, 1, 2010



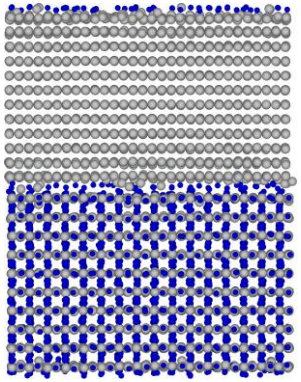
Molecular Dynamics T = 300 K t = 0.01 ns ⇒



3. Our preliminary Mg-H **bond order potential** enables MD simulations of MgH_2 crystals.

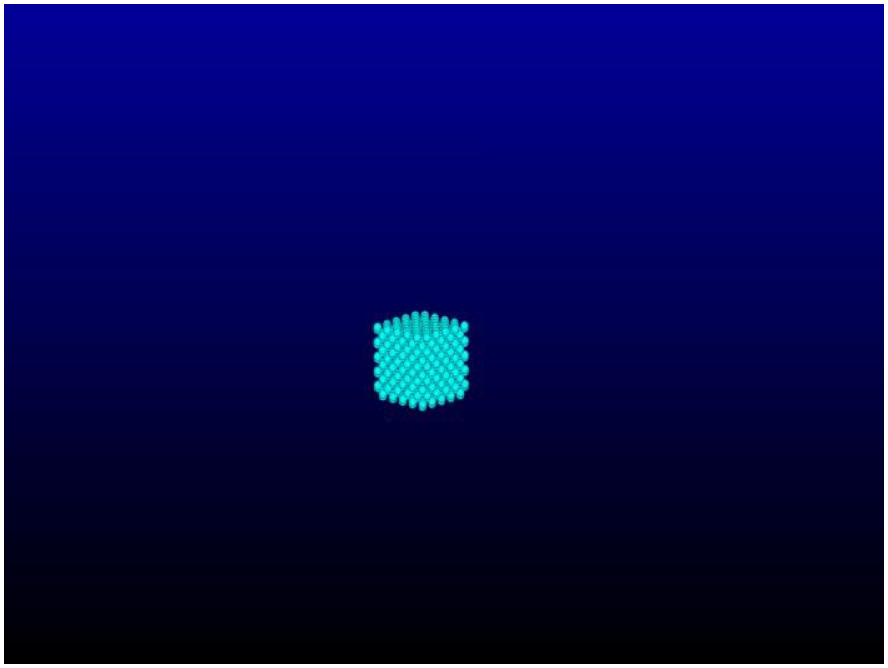


Molecular Dynamics T = 300 K t = 1.0 ns ⇒

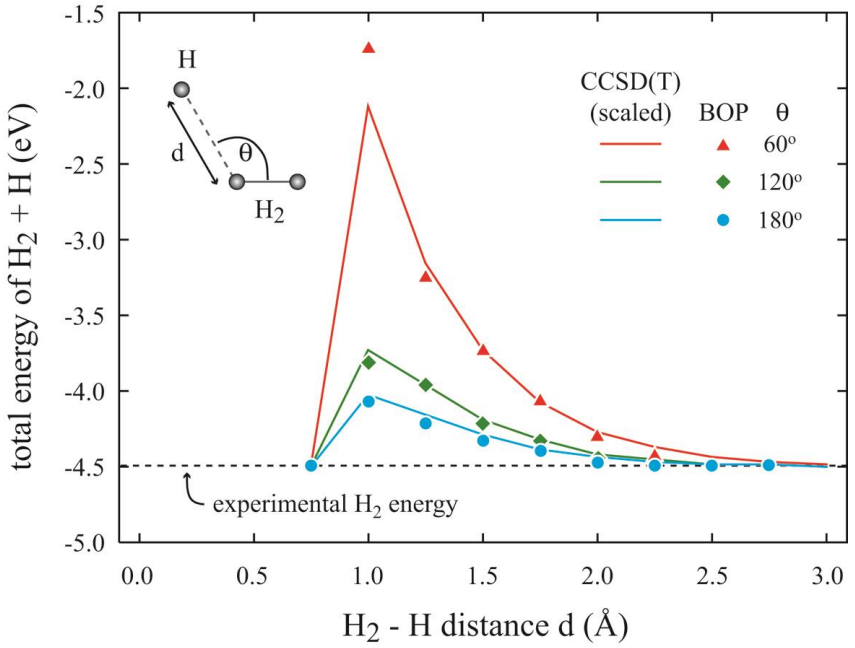


$H_2+H \rightarrow H+H_2$ chemical reaction

Hydrogen crystal to H_2 gas



$H_2+H \rightarrow H+H_2$ energy profiles

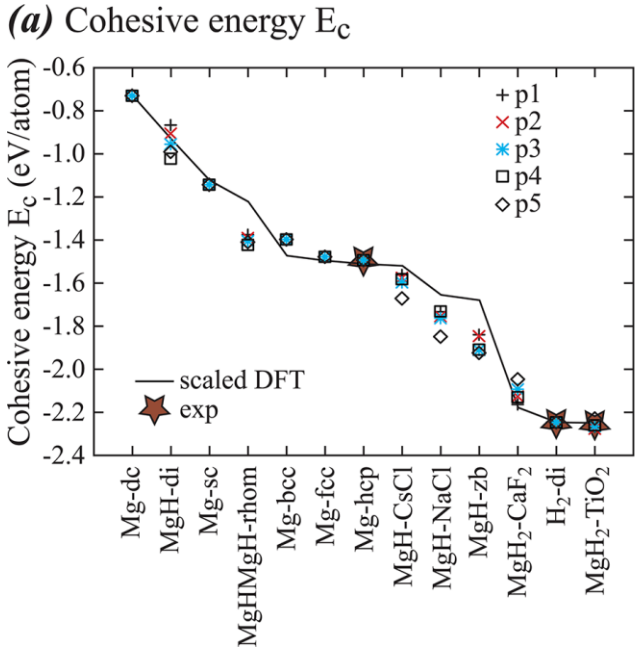


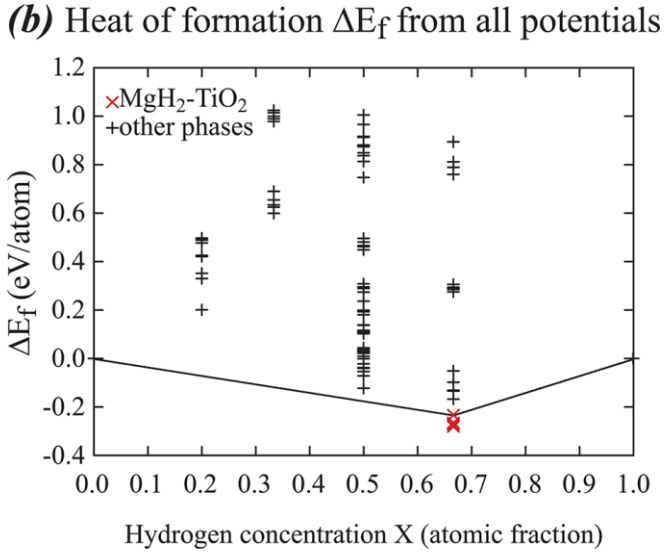
Our BOP captured the $H_2+H \rightarrow H+H_2$ reaction

Transferability

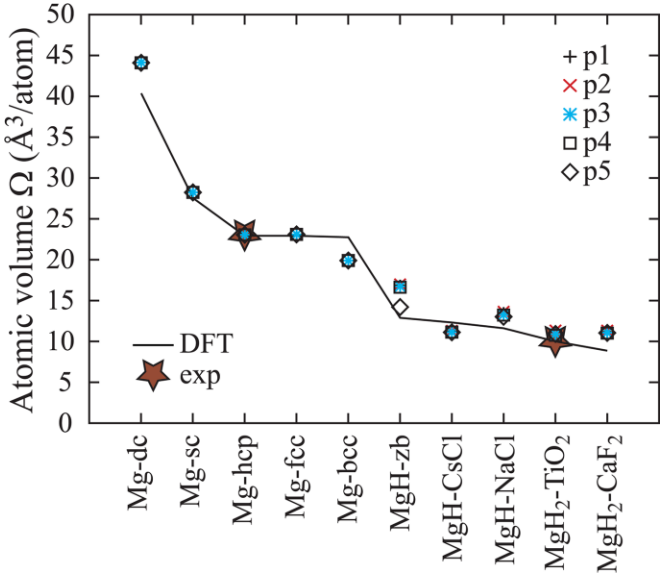
Sandia National Laboratories

Energy trends





Volume trends



Elastic constants of Mg-hcp

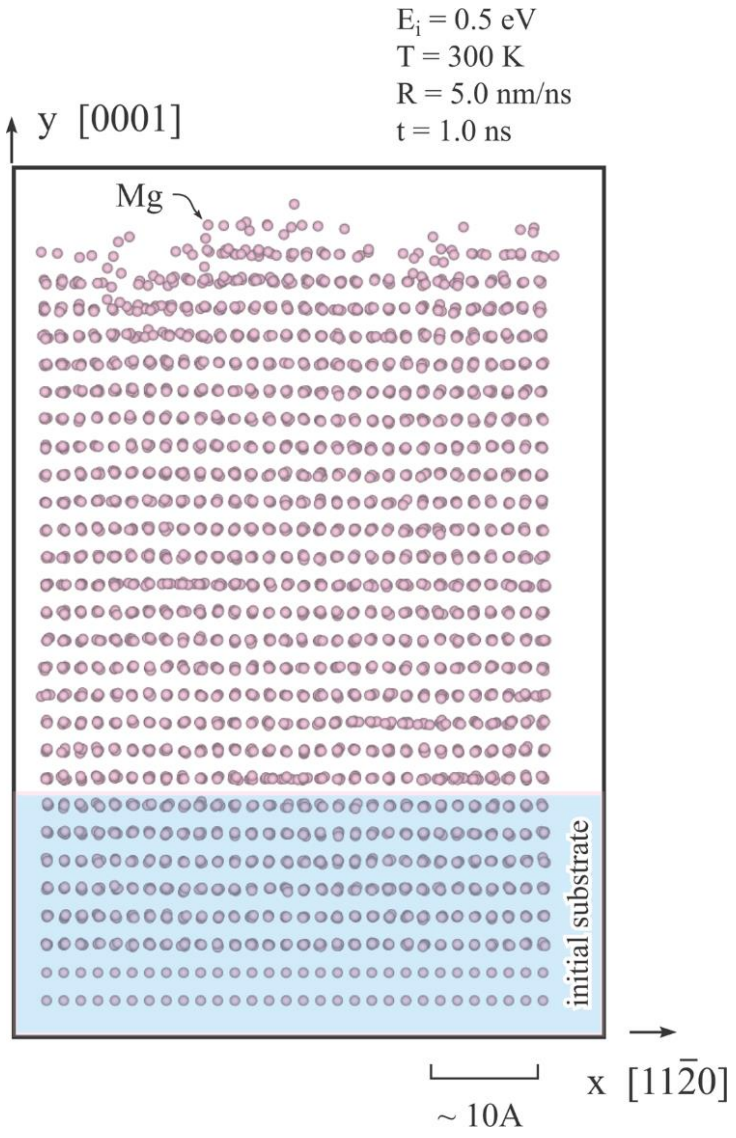
Table I. Elastic constants C_{11}, C_{33}, C_{44}, C_{13}, and bulk modulus $B = (2C_{11} + 2C_{12} + C_{33} + 4C_{13})/9$ in unit eV/Å^3.

C_{11}		C_{33}		C_{44}		C_{13}		B	
Cal.	Exp.	Cal.	Exp.	Cal.	Exp.	Cal.	Exp.	Cal.	Exp.
0.40	0.40	0.42	0.41	0.11	0.12	0.14	0.14	0.23	0.23

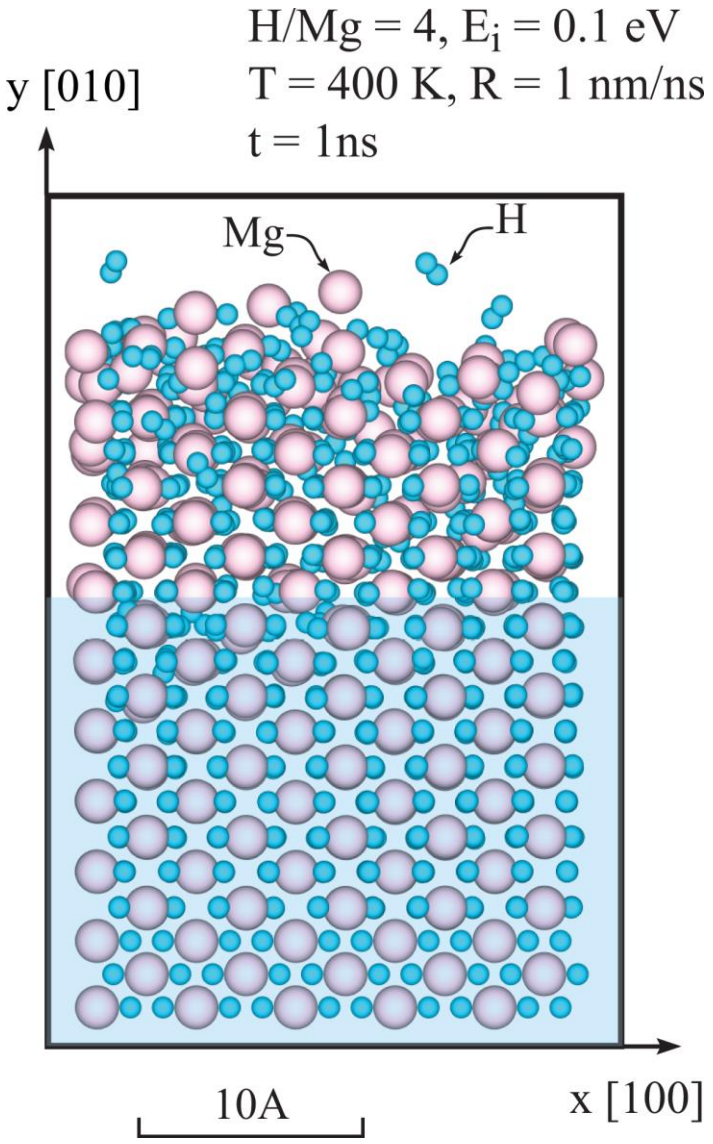
MD Growth Simulations

Sandia National Laboratories

Crystalline growth of Mg-hcp

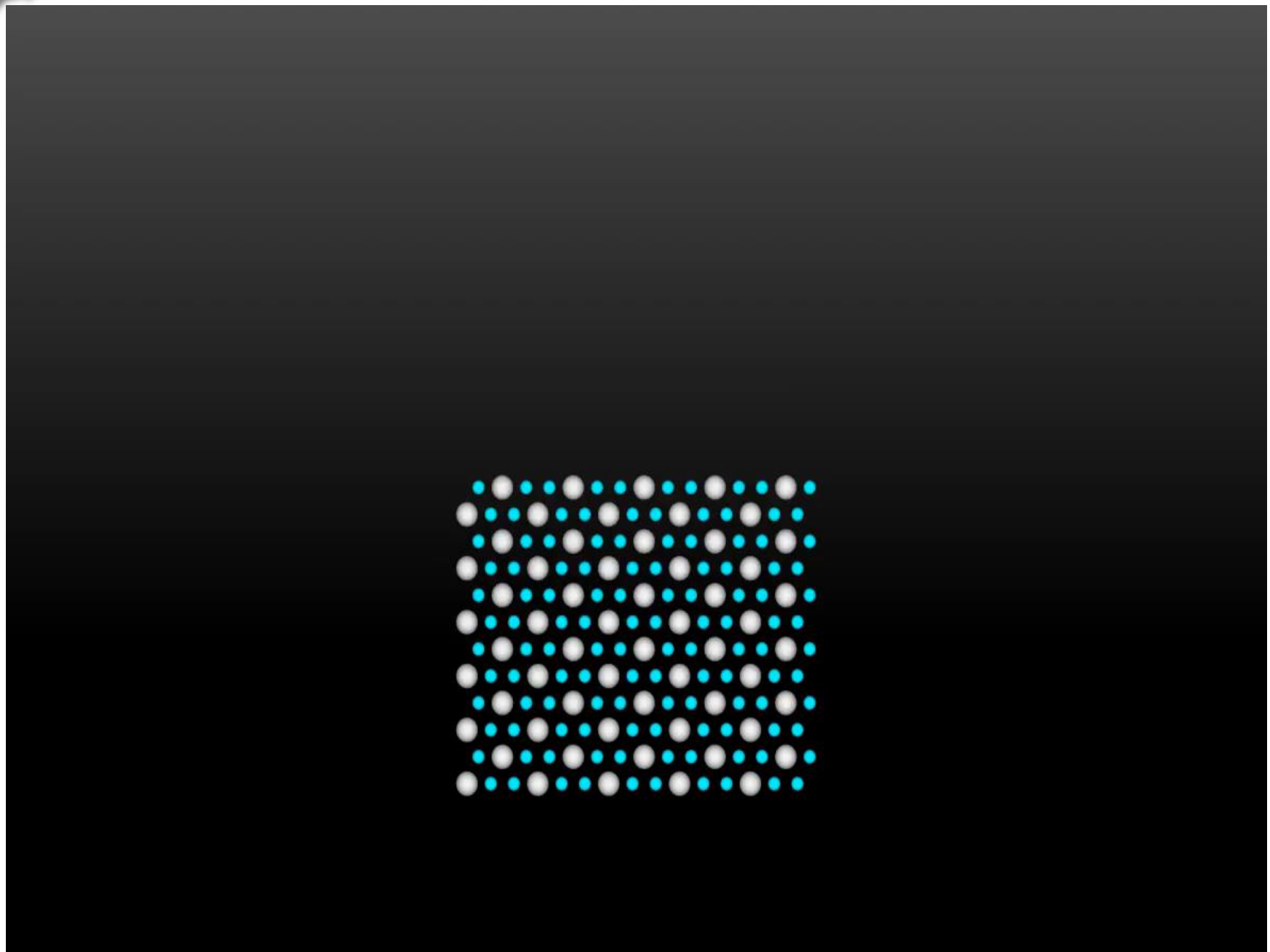


Crystalline growth of MgH_2-rutile



Time-evolution of growth

Sandia National Laboratories



Summary

1. Our Fe-Ni-Cr EAM captures the four criteria critical for simulations of stainless-steels. Hydrogen is yet to be added;
2. Our Al-Cu-H BOP captures the high Al stacking fault energy, the stability of Al_2Cu compound, and the H_2/H chemical reaction;
3. Our Mg-H BOP captures the crystalline growth of the rutile phase of MgH_2 hydride.