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Atomic scale investigation of hydrogen diffusion in nickel plated zirconium

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Milestones

M1. Evaluate existing potentials for Ni-Zr, Zr-O and Zr-H – 28 February 2019

M2. Create DFT data to add H interactions to the Ni-Zr and Zr-O potentials – 30 April 2019

M3. Calculate H/T diffusion in the bulk systems Ni, Ni-Zr, Zr and ZrO_2 – 31 July 2019

M4. Calculate H/T diffusion in the interface systems Ni:Zr and ZrO_2:Zr – 31 August 2019

M5. Complete analyses and final report and/or journal article – 30 September 2019

U.S. DEPARTMENT OF ENERGY

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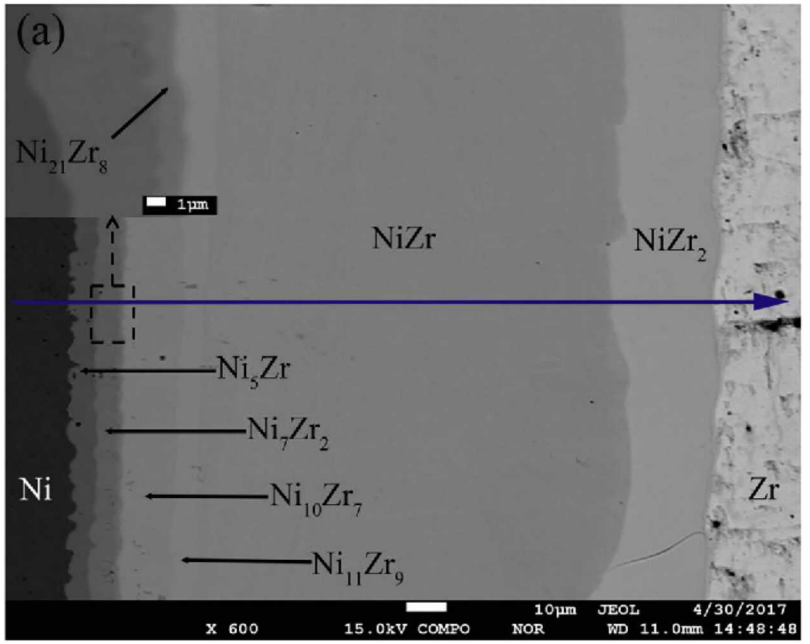
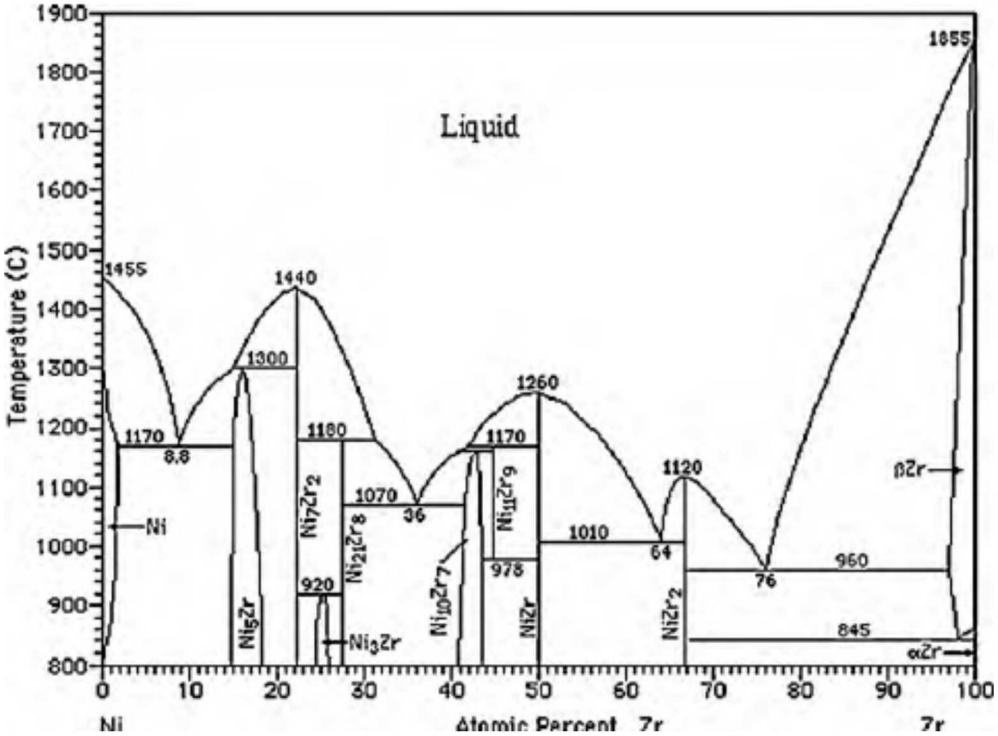
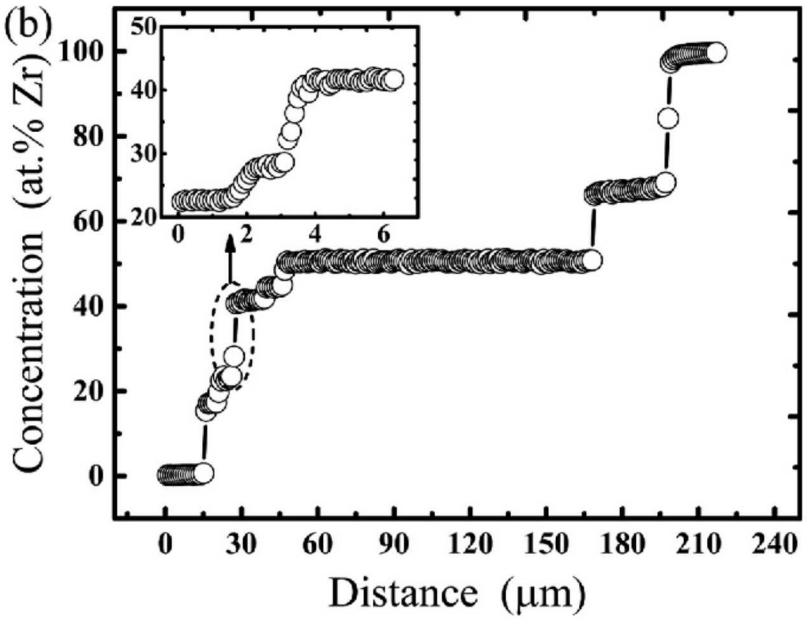
National Nuclear Security Administration

Motivation

- Zyr-4 is a getter of H due to its ability to absorb and transport H
- Zr forms a surface oxide layer that inhibits its H permeability
- Coating with Ni prevent oxide formation
- **How does the Ni layer affect H permeability?**
- Understanding can inform experiments/ design/testing



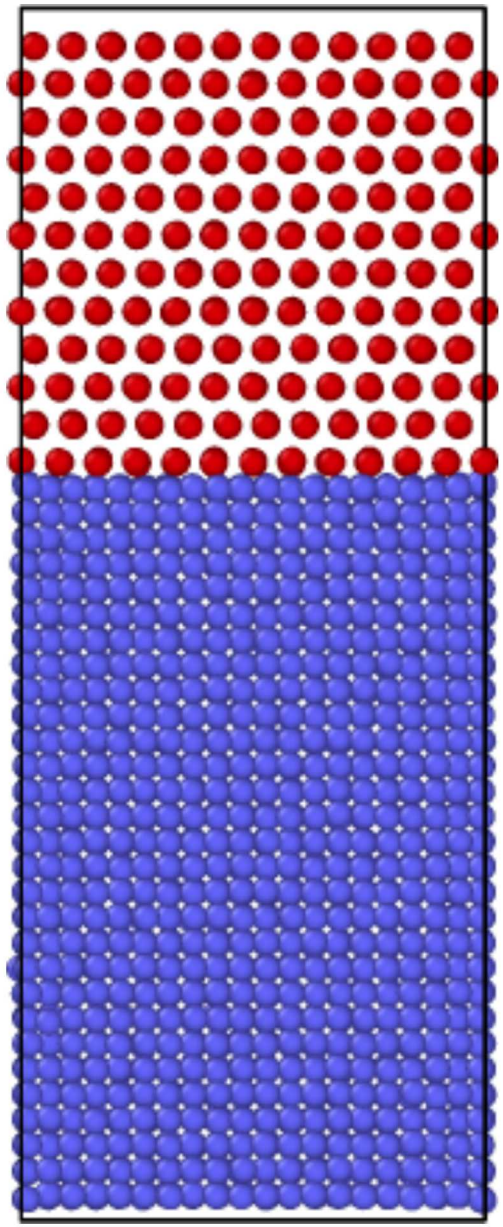




[Tao, J.AlloysComp., 2018]

Project plan

1. Build an interatomic potential for Zr-Ni-H based on new DFT training data and existing potentials
2. Use high temperature molecular dynamics and Arrhenius behavior to estimate the H diffusion coefficient in bulk Zr, Ni, and Zr-Ni bimetals.
3. Build an interatomic potential for Zr-O-H based on new DFT training data and existing potentials
4. Use high temperature molecular dynamics and Arrhenius behavior to estimate the H diffusion coefficient in bulk ZrO_2, and Zr-ZrO_2 bimaterials.
5. Explore the effects of bulk and interface defects on the H-diffusion
6. Use results to understand the change in kinetics of Ni plated Zr.



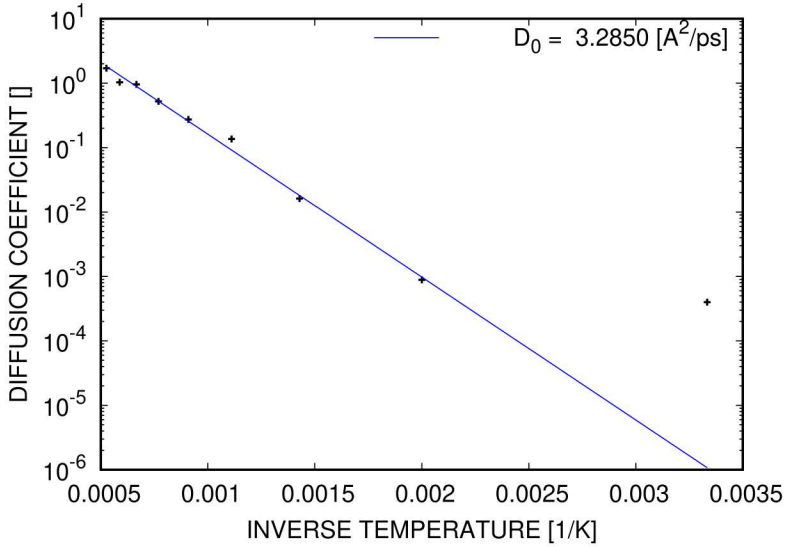
Existing potentials

A number of binary potentials have been developed that include two of Zr/Ni/H/O in the appropriate phases.

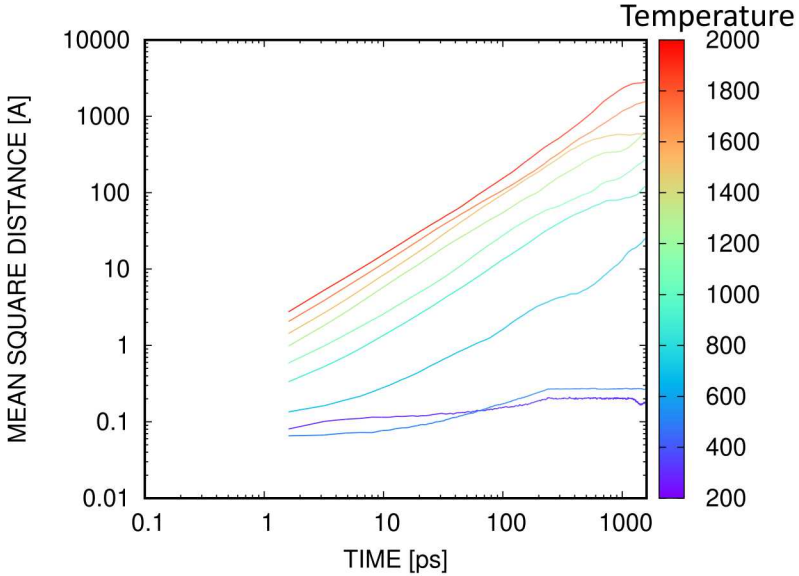
Notable potentials:

- Zr-H by Byeong-Moon Lee and Byeong-Joo Lee, *Metallurgical and Materials Transactions*, 2014
- Ni-Zr by Mendelev *et al., Philosophical Magazine,* 2012

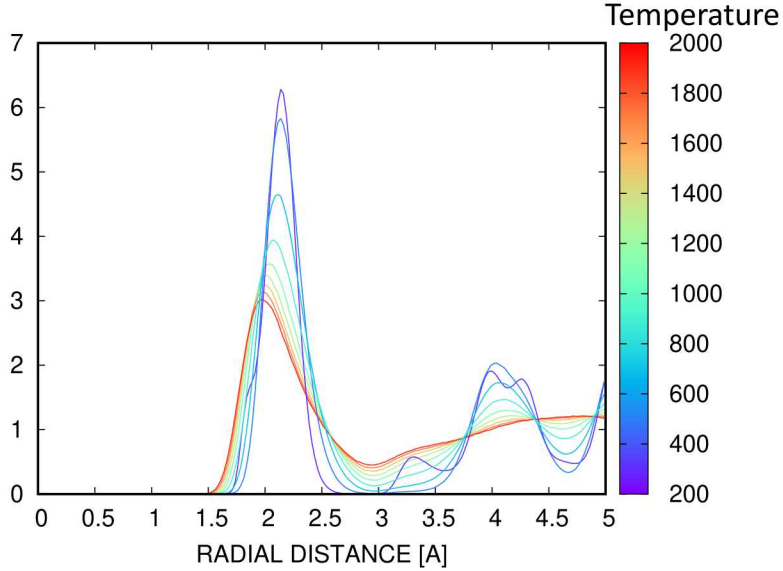
Zr-H Arrhenius behavior



H in Zr random walk

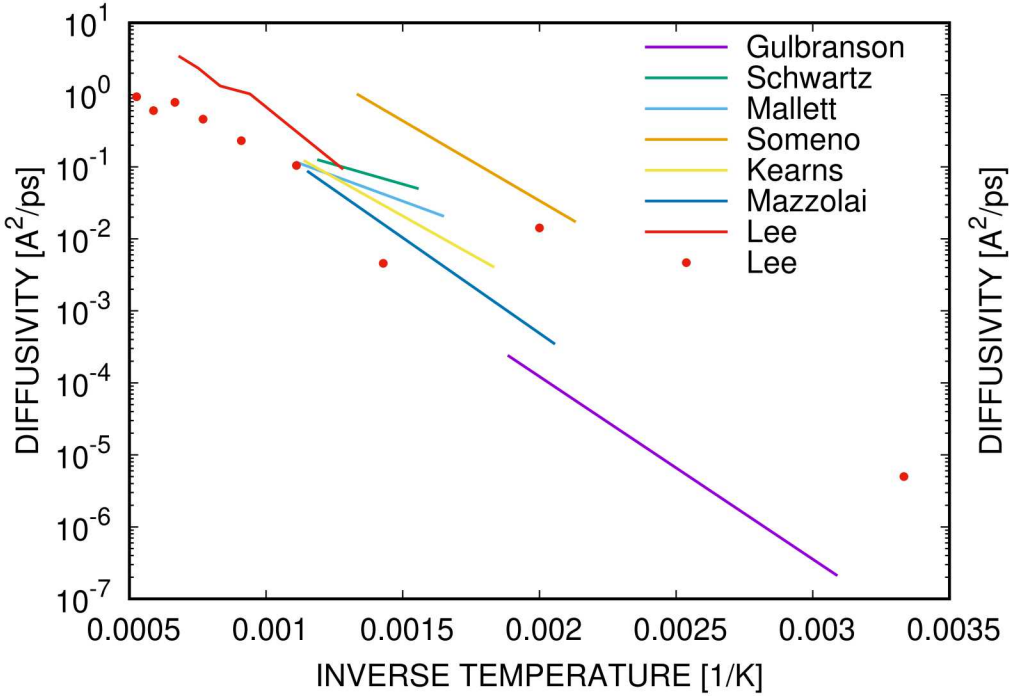


Zr-H radial distribution

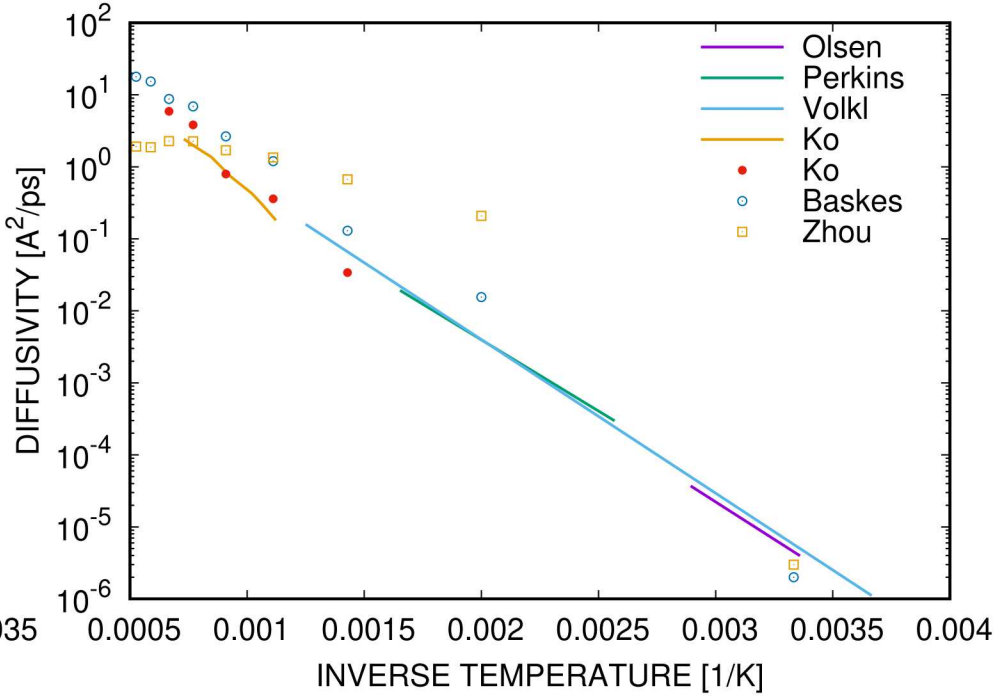


Diffusion

Zr-H Arrhenius behavior



Ni-H Arrhenius behavior



We pulled data from the literature (lines) and also ran independent calculations (points, sometimes needing to translate papers to potential files and test translations)

Ko&Lee MEAM NiH is notable: apparently unstable for more than one H

Potential selection

Lee MEAM

	Zr	Ni	H
Zr	X	?	X
Ni		X	X
H			?

Mendelev EAM

	Zr	Ni	H
Zr	X	X	?
Ni		X	?
H			?

- Byeong-Moon Lee and Byeong-Joo Lee. *A comparative study on hydrogen diffusion in amorphous and crystalline metals using a molecular dynamics simulation*. Metallurgical and Materials Transactions A, 45(6):2906–2915, 2014
- MI Mendelev, MJ Kramer, SG Hao, KM Ho, and CZ Wang. *Development of interatomic potentials appropriate for simulation of liquid and glass properties of NiZr2 alloy*. Philosophical Magazine, 92(35):4454–4469, 2012

EAM:

- no hydride formation
- optimized to study the liquid and amorphous structure of the $NiZr_2$ alloy.
- simpler to parameterize
- peculiar behavior of Ko & Lee Ni-H MEAM potential for more than 1 H in a simulation

Embedded atom (EAM) potential

$$\Phi(r_{\alpha\beta}) = \sum_{a,\alpha\in\mathcal{G}_a} F_a \left(\sum_{b,\beta\in\mathcal{G}_b\neq\alpha} \rho_{ab}(r_{\alpha\beta}) \right) + \sum_{a,\alpha\in\mathcal{G}_a} \sum_{b,\beta\in\mathcal{G}_b\neq\alpha} \frac{1}{2}\phi_{ab}(r_{\alpha\beta})$$

Multibody embedding energy

Pair potential

Two potentials built upon the NiZr Mendelev potential:

- **NiZr+H:** set F_H = 0 and only tune the *-H pair potentials in Morse form (3x3 parameters):

  $$\phi_{ab}(r) = E_{ab}\left(\exp(-2\alpha_{ab}(r - r_{ab})) - 2\exp(-\alpha_{ab}(r - r_{ab}))\right)$$

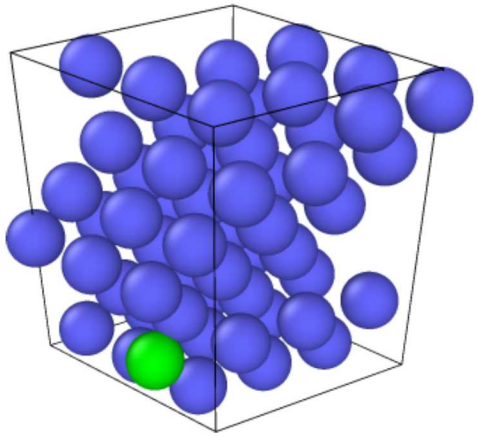
- **NiZrH:** calibration of the full EAM interactions (20+ parameters)

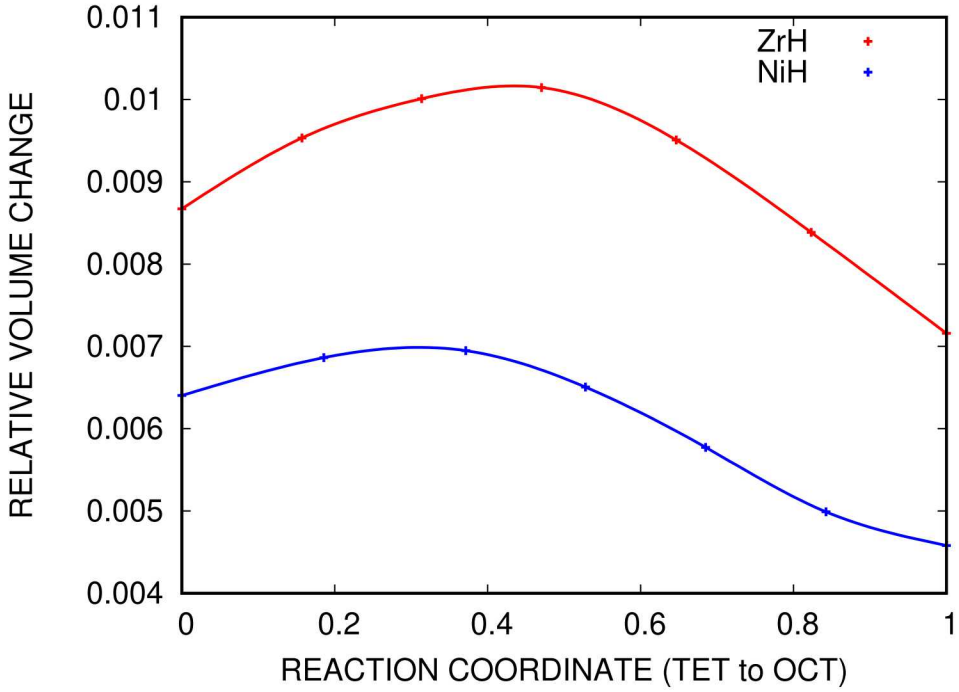
Ab initio training data

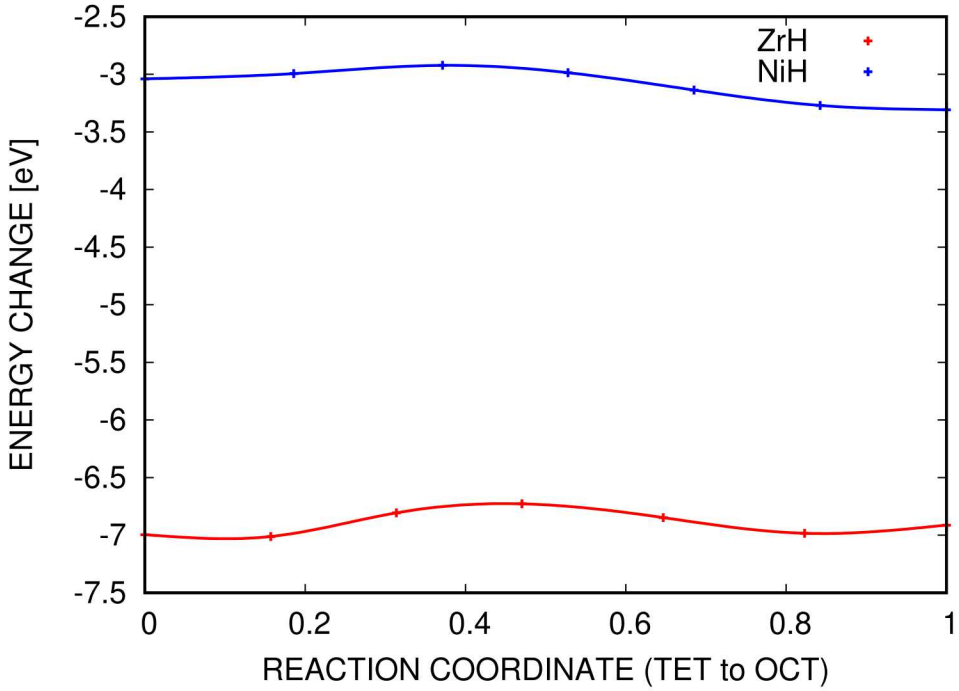
We have training data for
Ni with H and Zr with H

- H swelling volume
- H insertion energy
- H tetrahedral to octahedral diffusion barrier
- H-H (in metal) interaction energy

DFT cell







We have similar training data for H in ZrO_2

NiZr+H potential development

In addition to calibrating a unified EAM potential we have added H-Zr, H-Ni, and H-H interactions to Mendelev's potential using the DFT data with a pairwise Morse potential form (3 parameters for each pair)

$$\sum_{a,\alpha\in\mathcal{G}_a}\sum_{b,\beta\in\mathcal{G}_b\neq\alpha}\frac{1}{2}\phi_{ab}(r_{\alpha\beta})$$

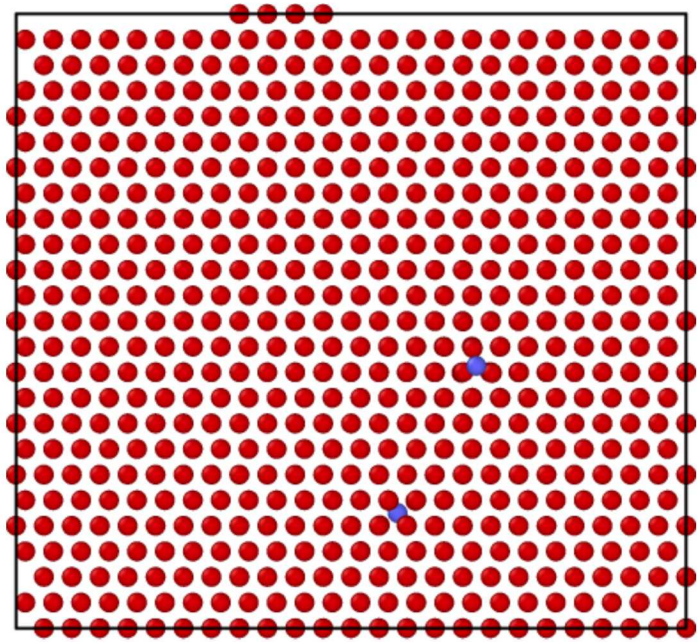
Pair potential

This can be done independently

$$E = D_0\left[e^{-2\alpha(r-r_0)} - 2e^{-\alpha(r-r_0)}\right] \qquad r < r_c$$

1. Calibrate H-Zr/Ni parameters using the (single H) swelling volume and energy change along the TET --> OCT diffusion path

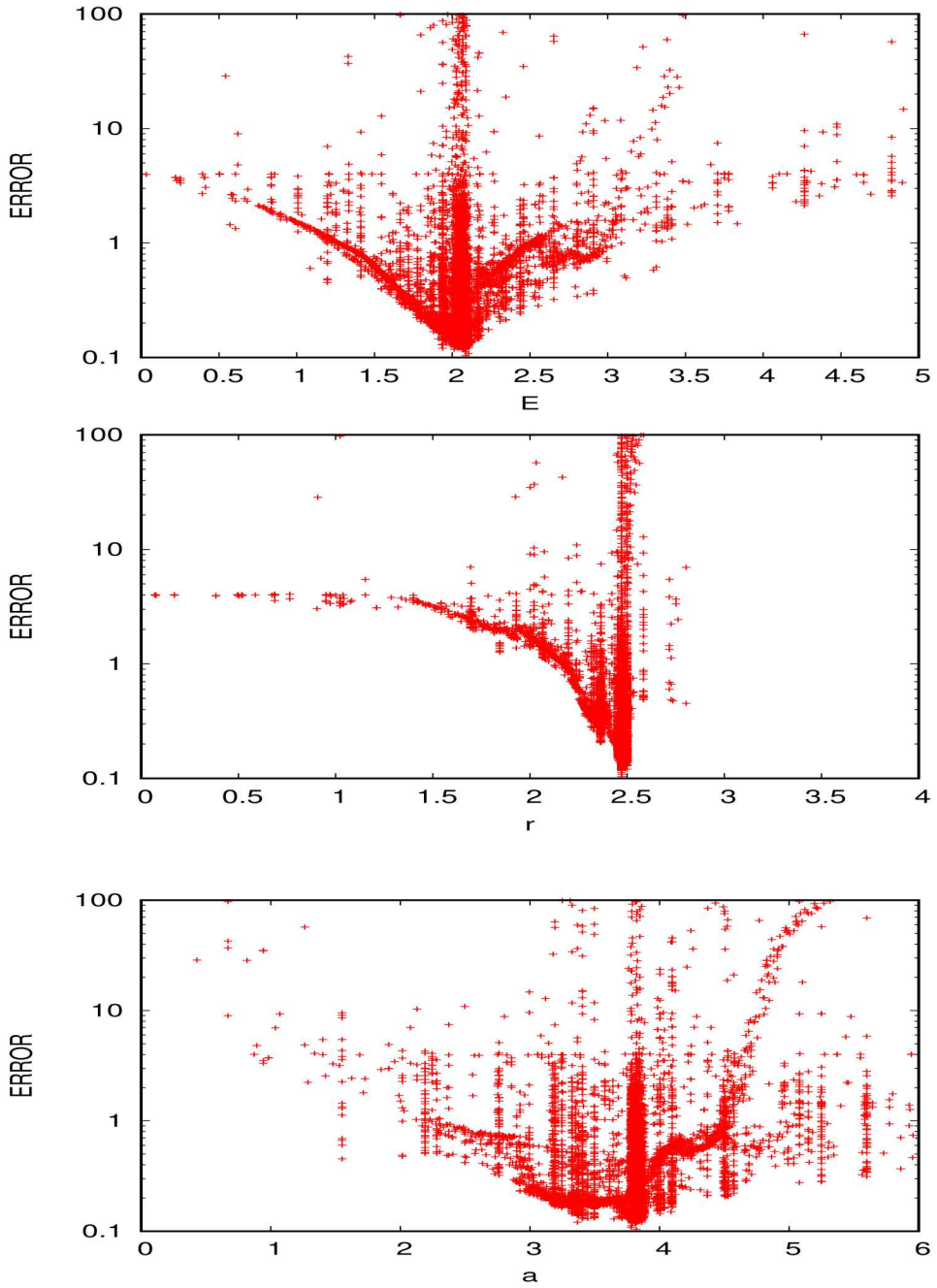
2. Calibrate H-H parameters using the two H insertion energy and volume change



Calibration

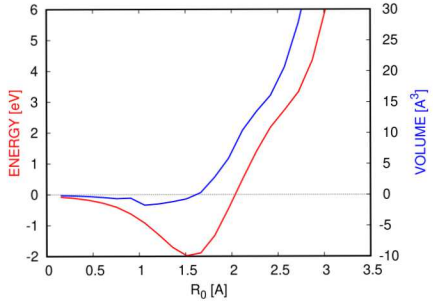
Ni-H pair interaction calibration with SOGA genetic algorithm. The genetic algorithm is a hybrid between a sampler/explorer and an optimizer.

Calibration based on the energy and volume changes along the DFT diffusion path appears to be well-posed – the are is a deep single well in the error

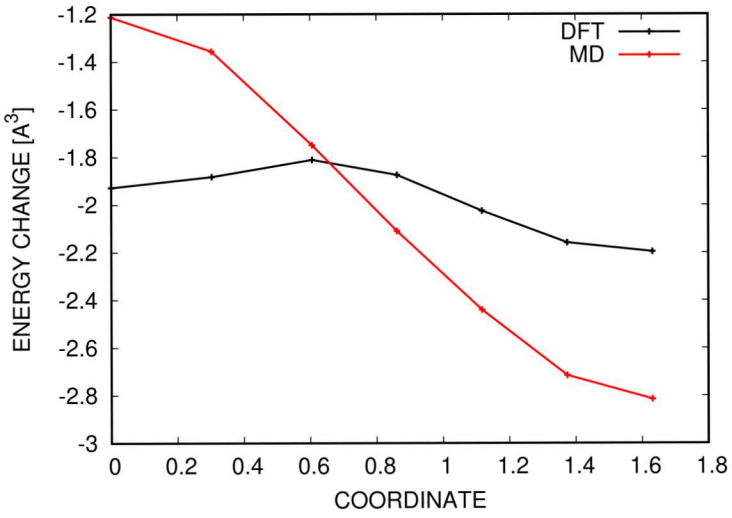


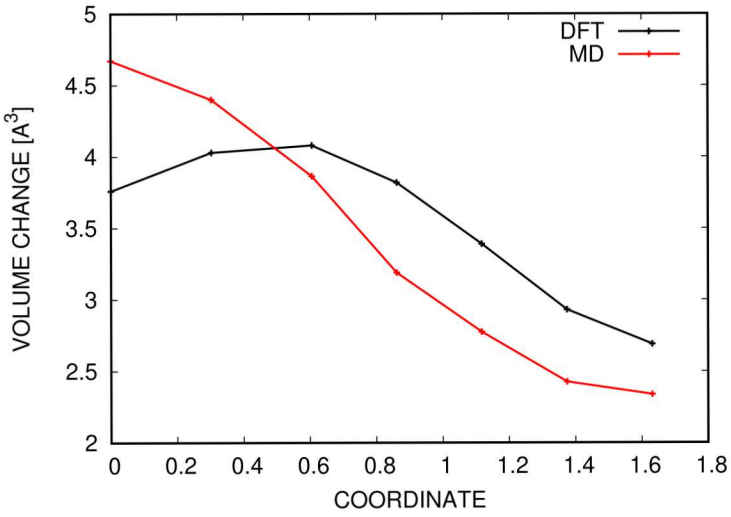
NiZr+H potential

For the pair interactions only there is model discrepancy with the data

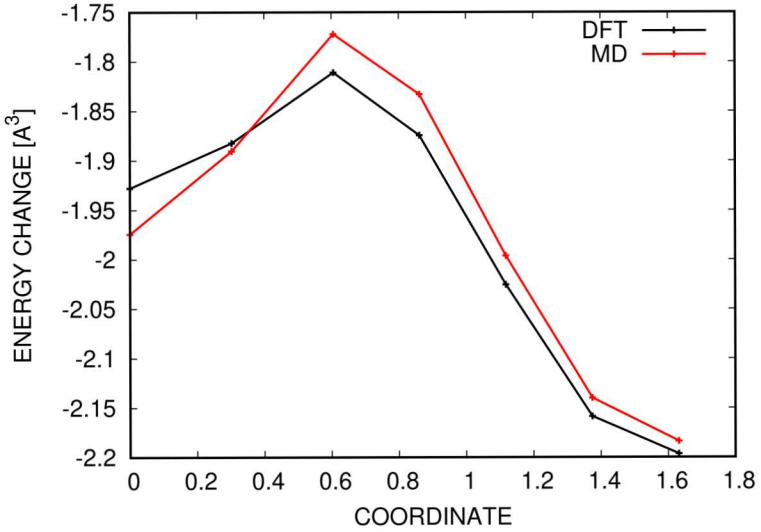


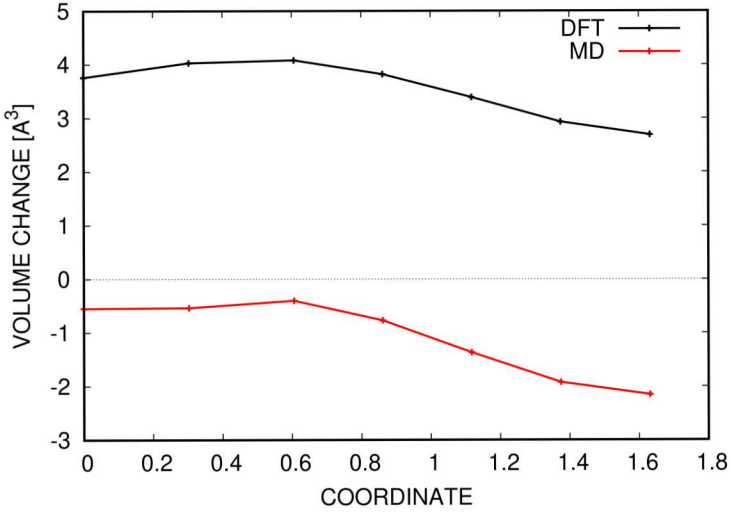
No barrier if try to fit V & E



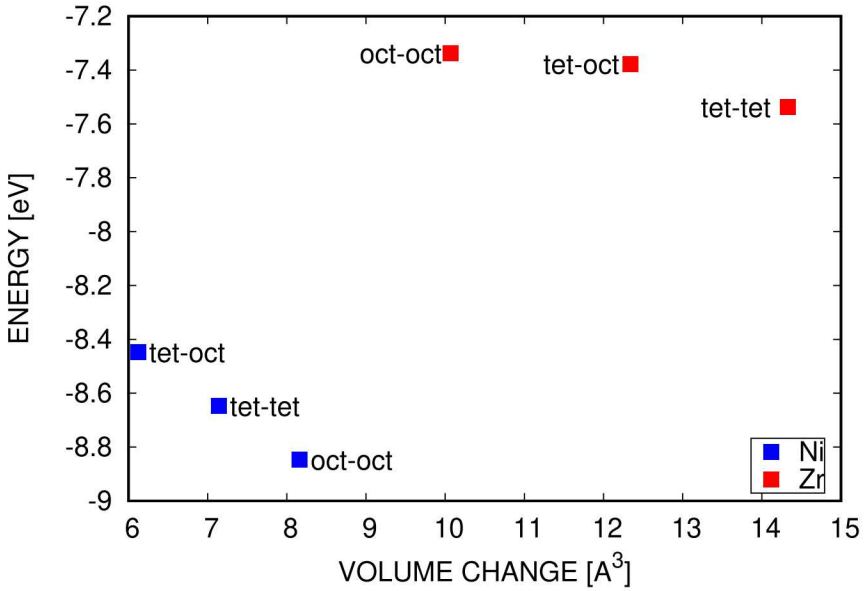


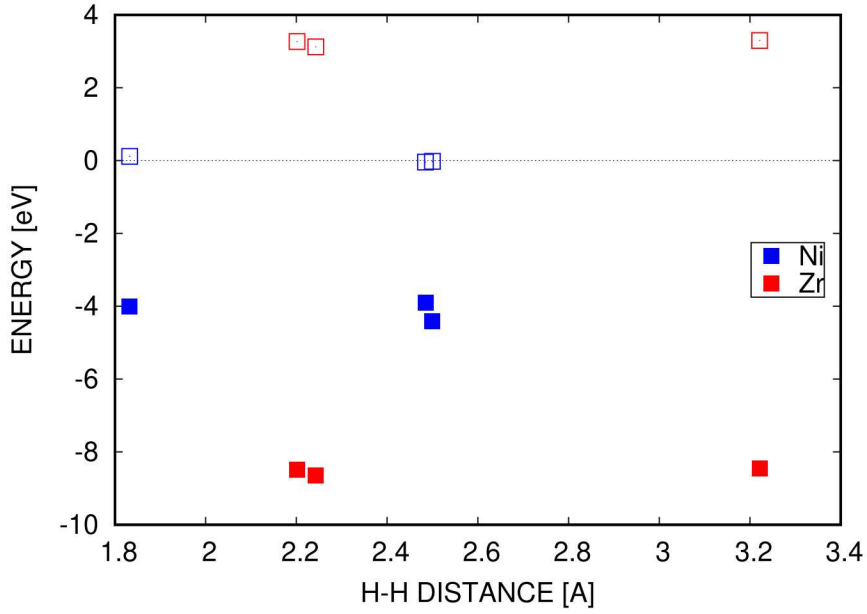
Good barrier if fit E only



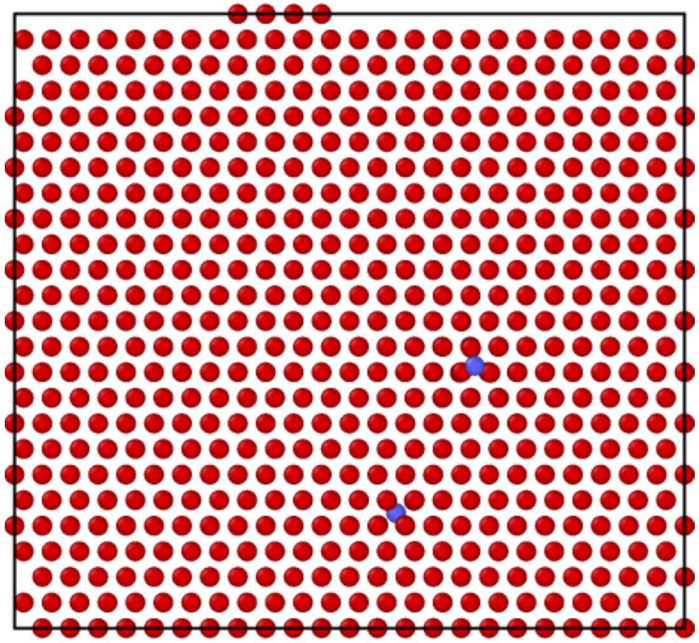


H-H potential



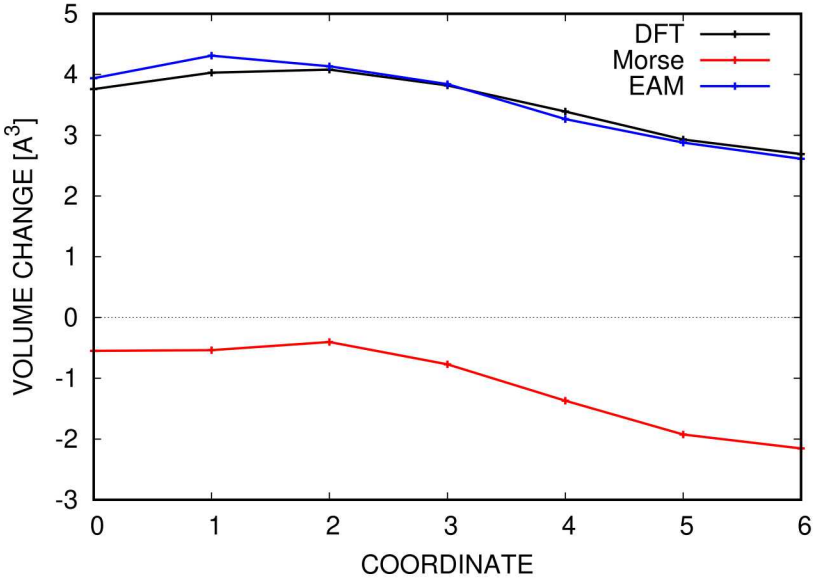


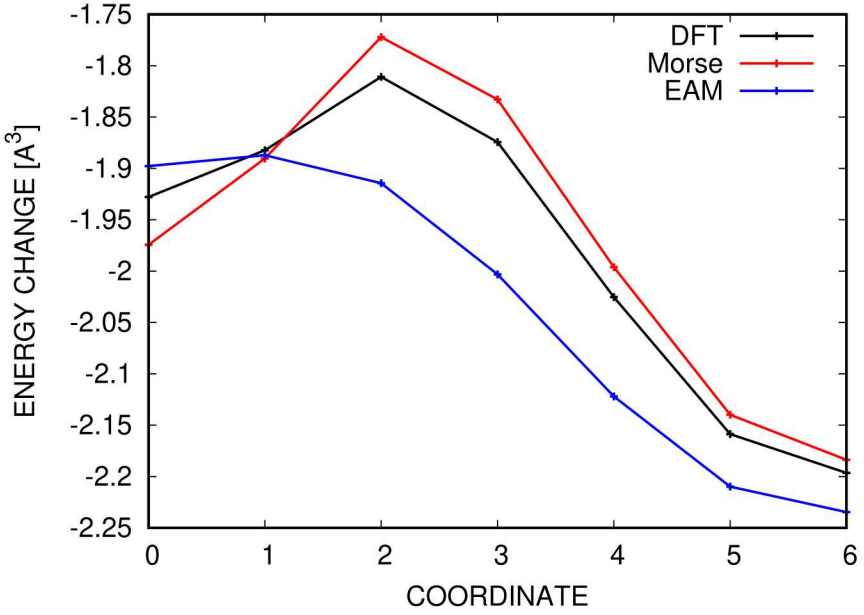
- There is a clear trend in volume-energy but the ordering is different for Ni vs Zr
- **There is no clear trend in H-H distance vs energy**
- Ni insertion is effectively additive (interactions very short range)
- Zr insertion has an additional cost (interactions long range compared to cell size?)



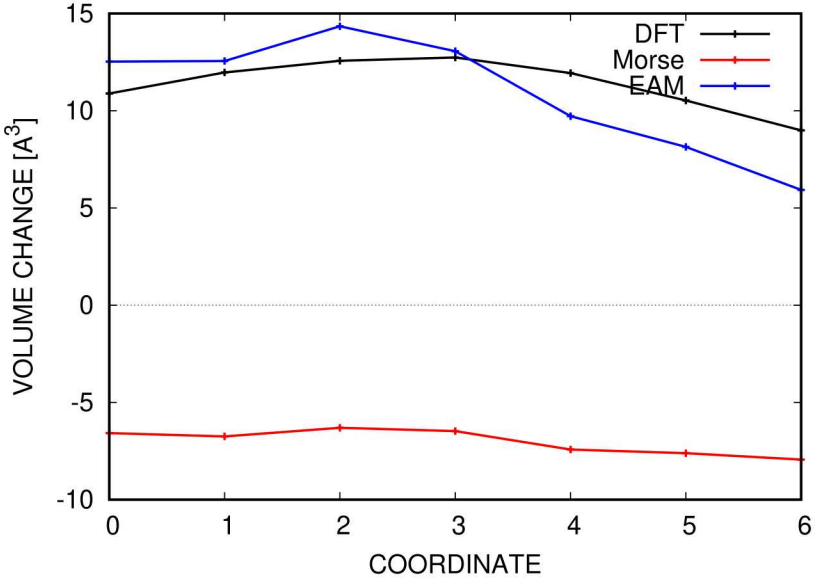
Full EAM NiZrH potential – current state

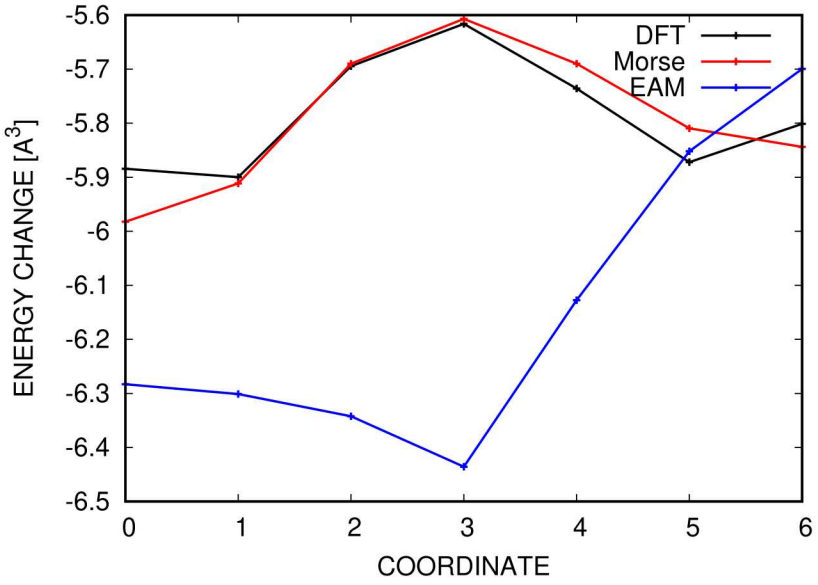
NiH





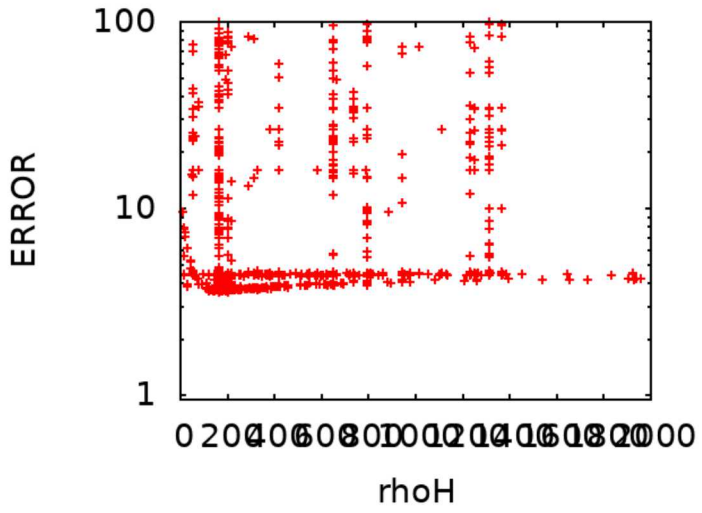
ZrH

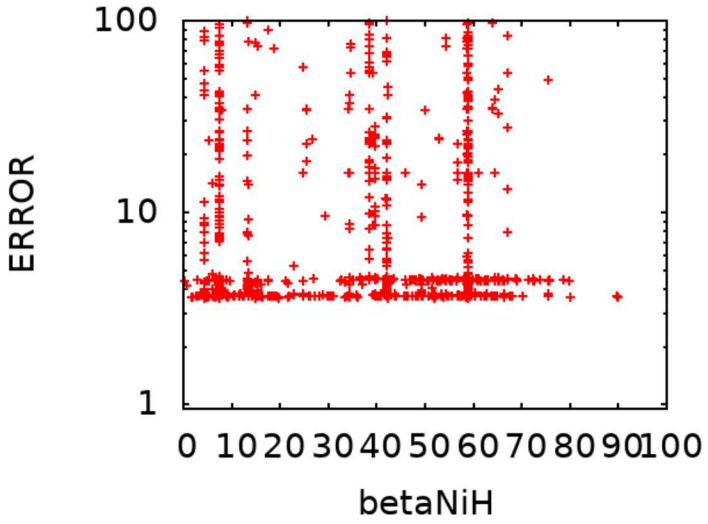


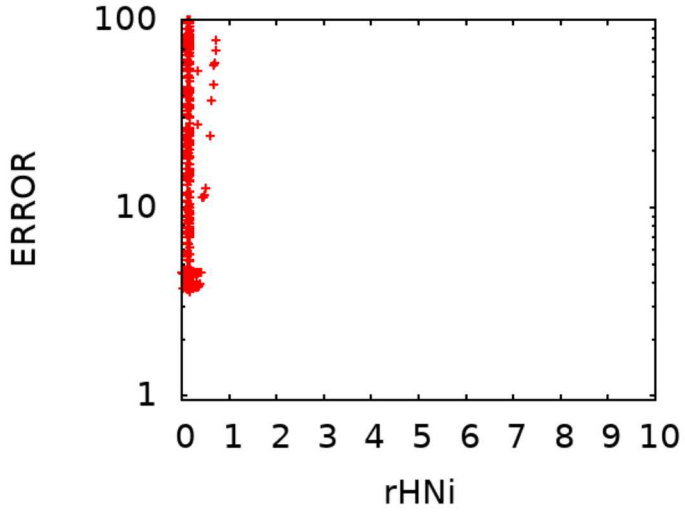


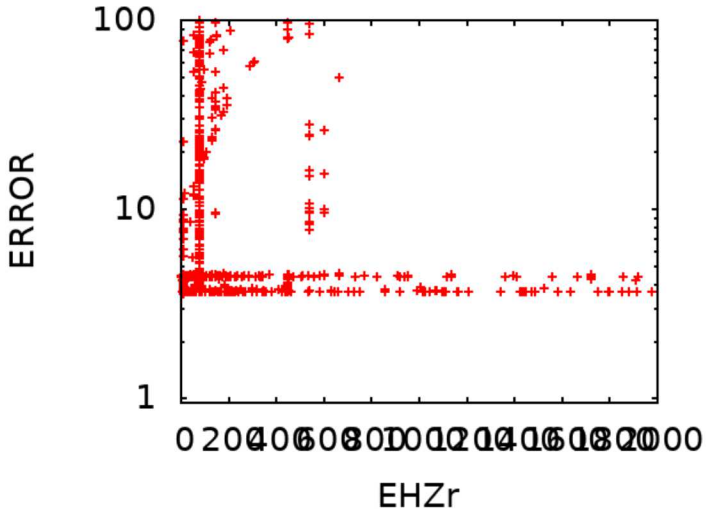
Full EAM NiZrH potential – current state

Issues with parameter identifiability: generating additional H in NiZr alloy training data







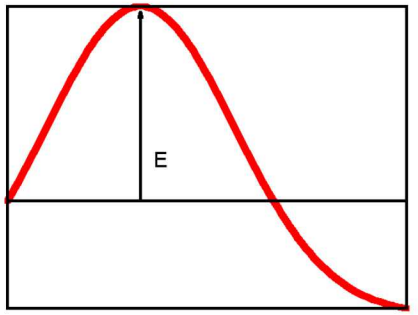


Methodology for diffusion estimates

Diffusion in a slow process for MD, we need to accelerate it with high temperatures

- Assume Arrhenius behavior – single dominant barrier with energy *E*

$$D(T) = D_0 \exp\left(-\frac{E}{k_B T}\right)$$

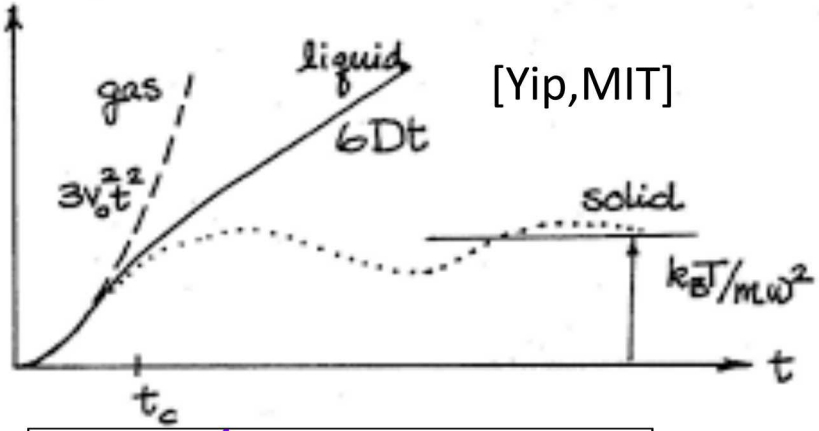


REACTION COORDINATE

- Use the Einstein relationship between mean squared displacement (MSD) and diffusion

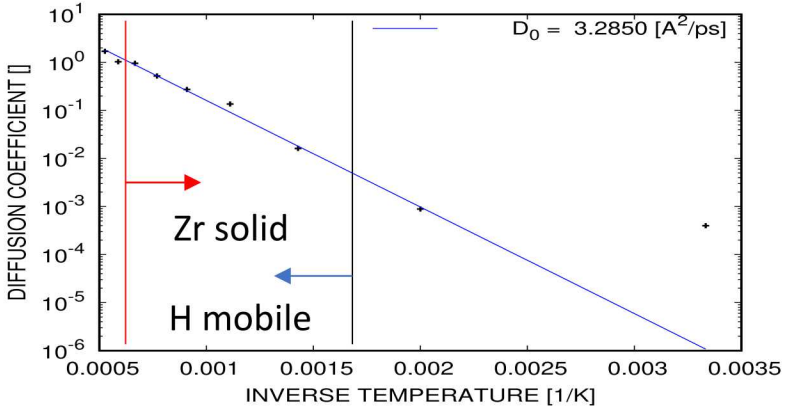
$$D = \frac{1}{6}\lim_{t\to\infty}\frac{\partial}{\partial t}\langle \|\Delta \mathbf{x}(t)\|\rangle$$

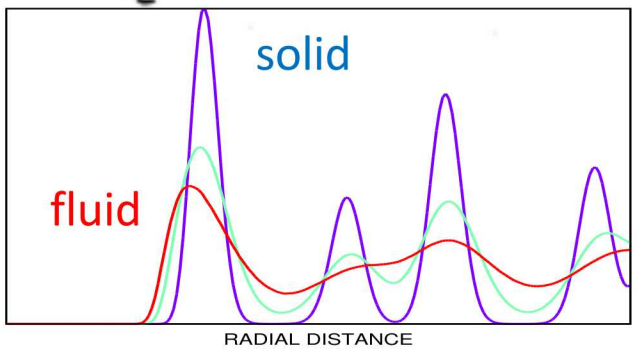
$\langle \Delta^2 r(t) \rangle$



[Yip,MIT]

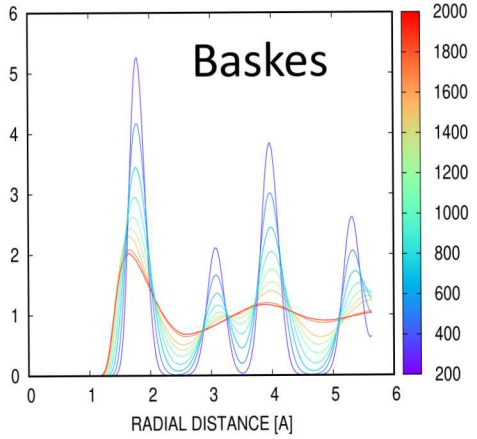
- Check radial distribution functions for H in NiZ and the NiZr solid to assess fluid/solid states

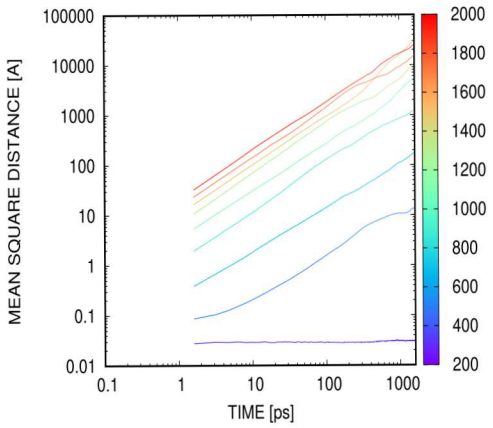


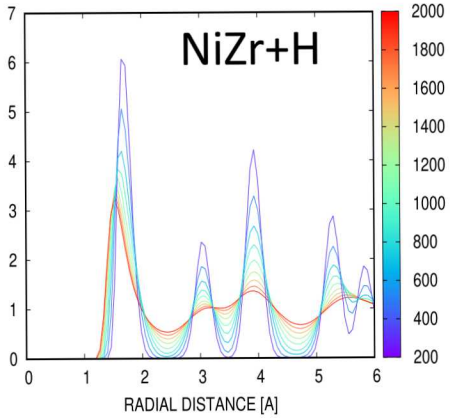


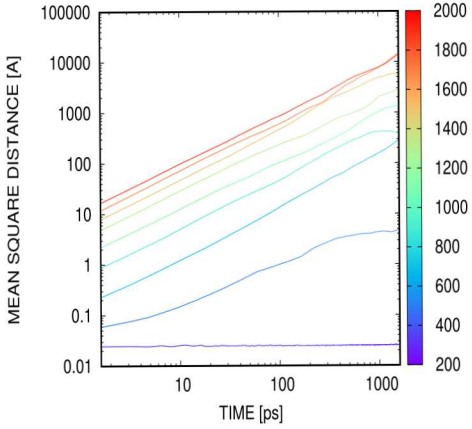
The lattice must be stable a sufficiently high temperature for this to work

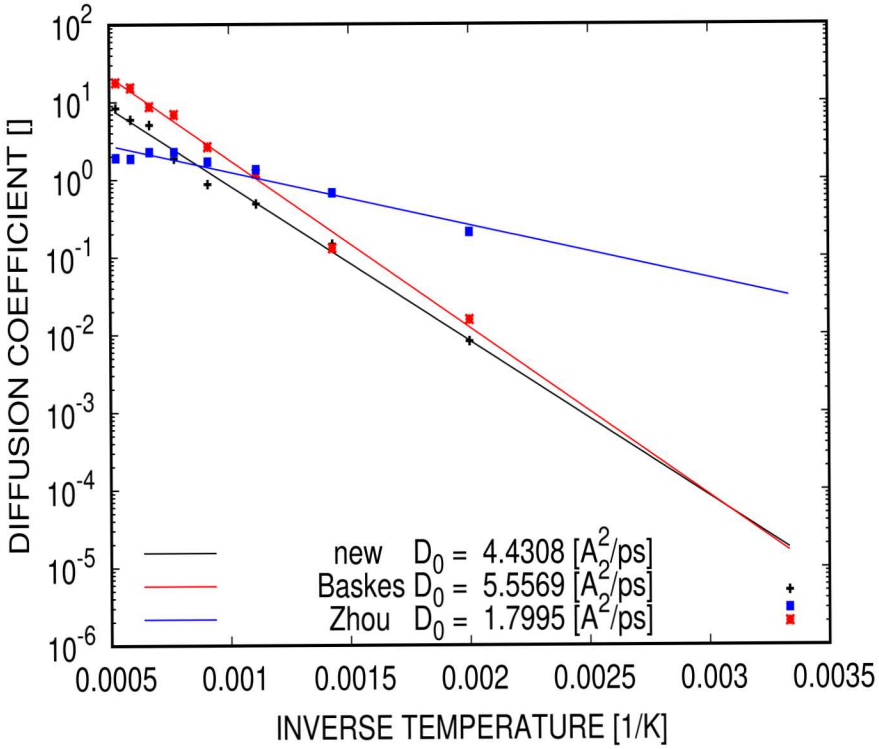
Validation: NiH



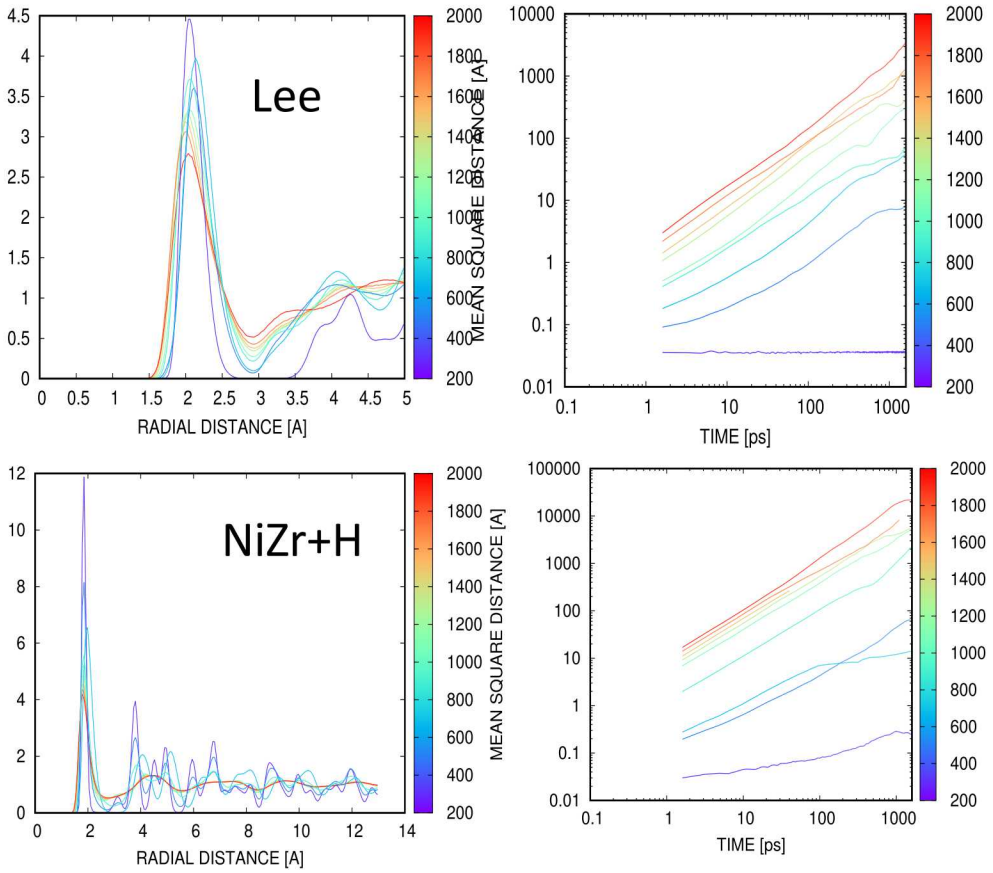


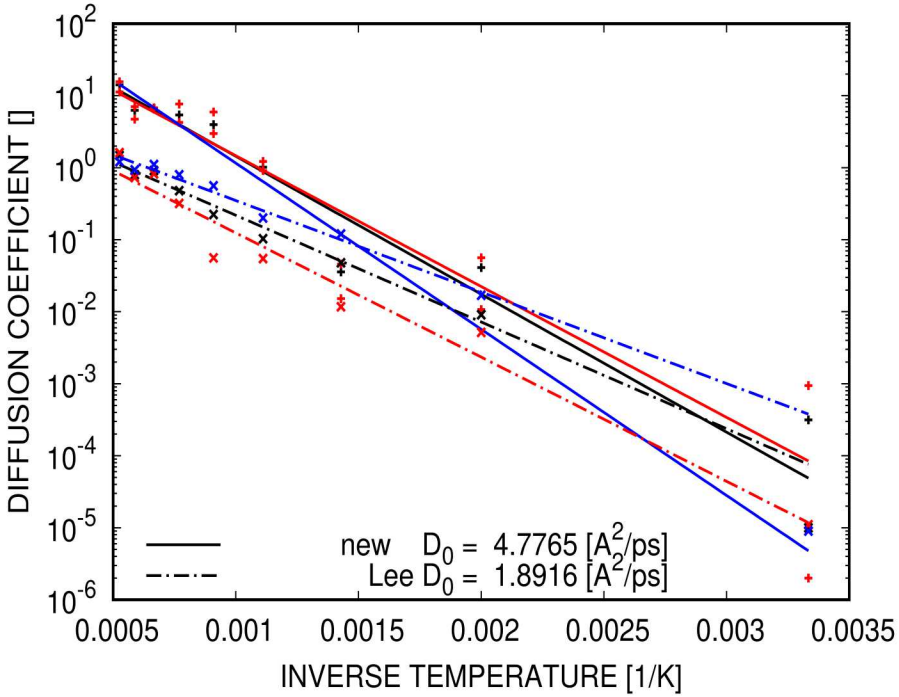






Validation: ZrH



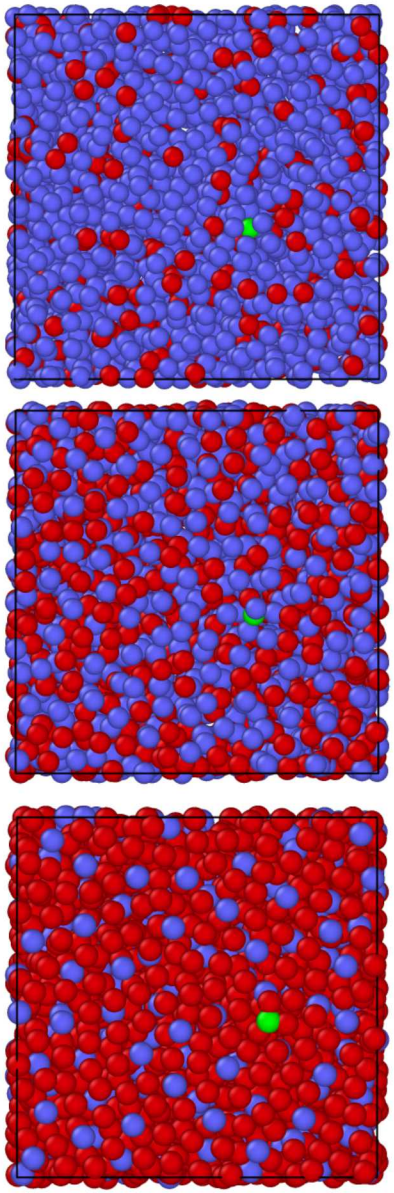


Different predictions for:

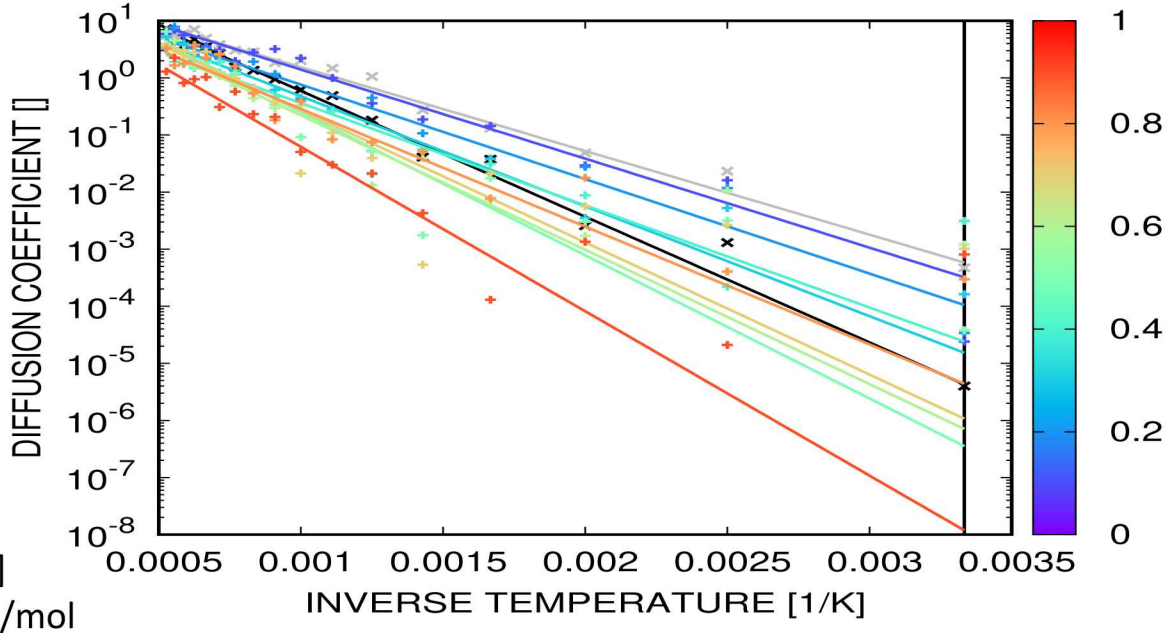
- Average
- a-plane
- c-axis

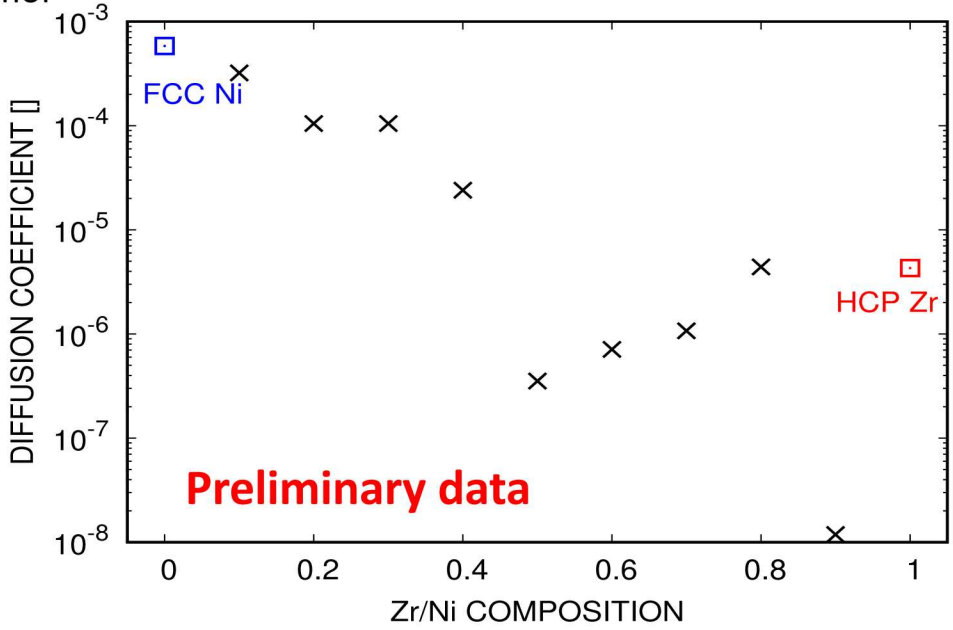
H diffusion in $Ni_x Zr_{1-x}$

Interface is diffuse so we examine independent layers



[Causey 2009]
Ni: E = 39.5 kJ/mol
Zr: E = 45.3 kJ/mol



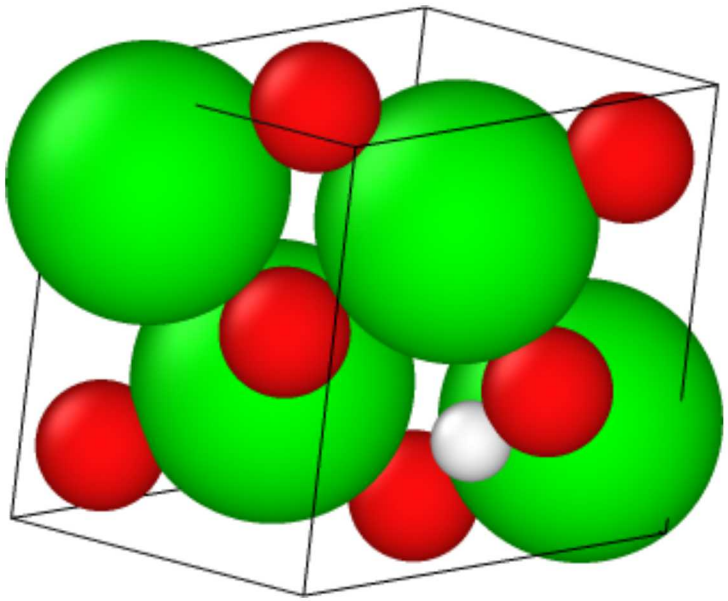


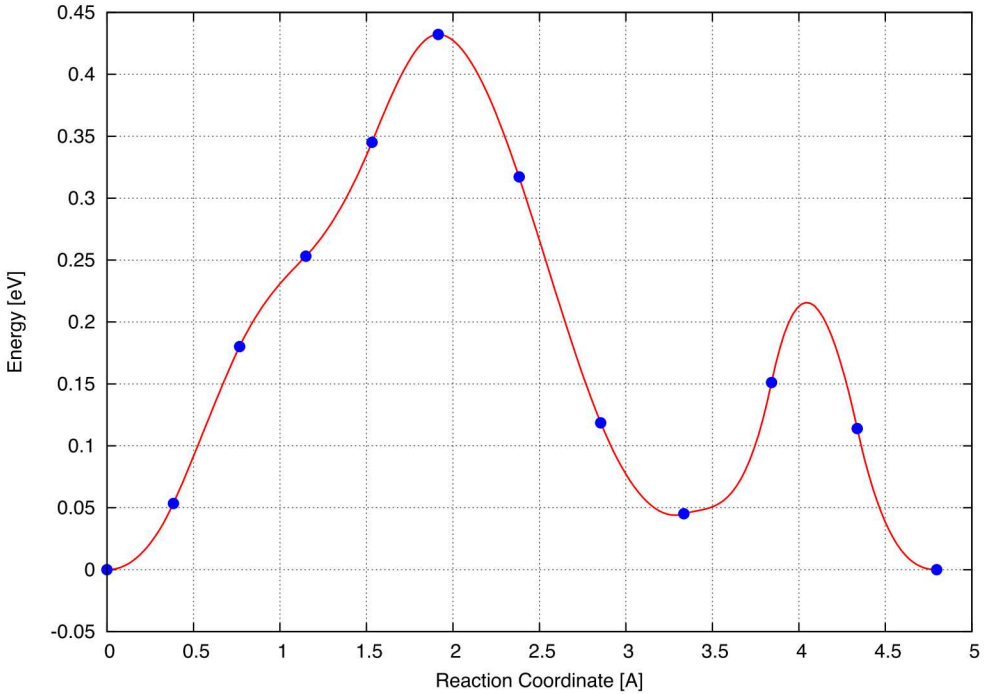
Exponential trend.

Noise due to insufficient sampling of amorphous configurations

H in ZrO_2 training data

ZrO has a multitude of phases/structures, e.g. monoclinic, cubic, and others that are stabilized with dopants

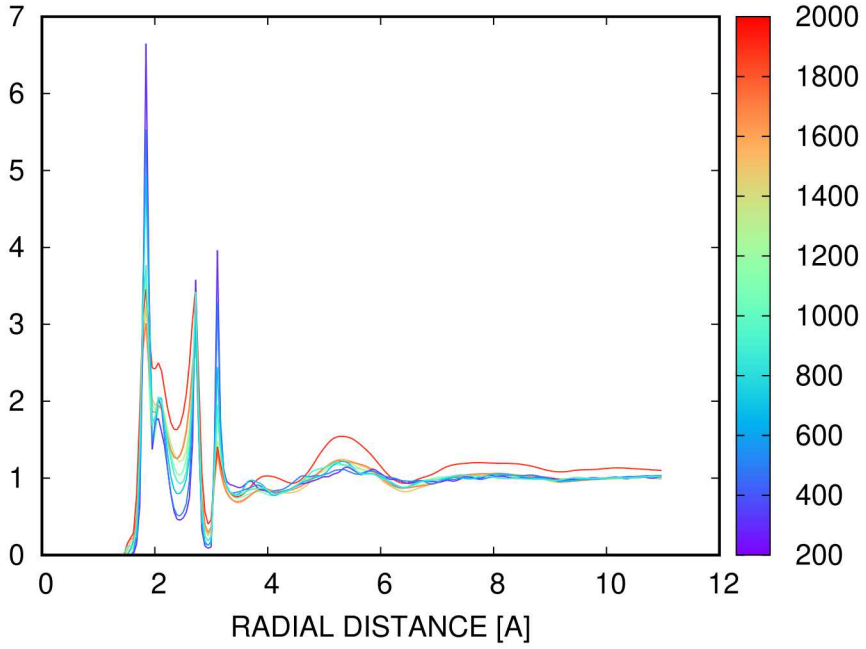


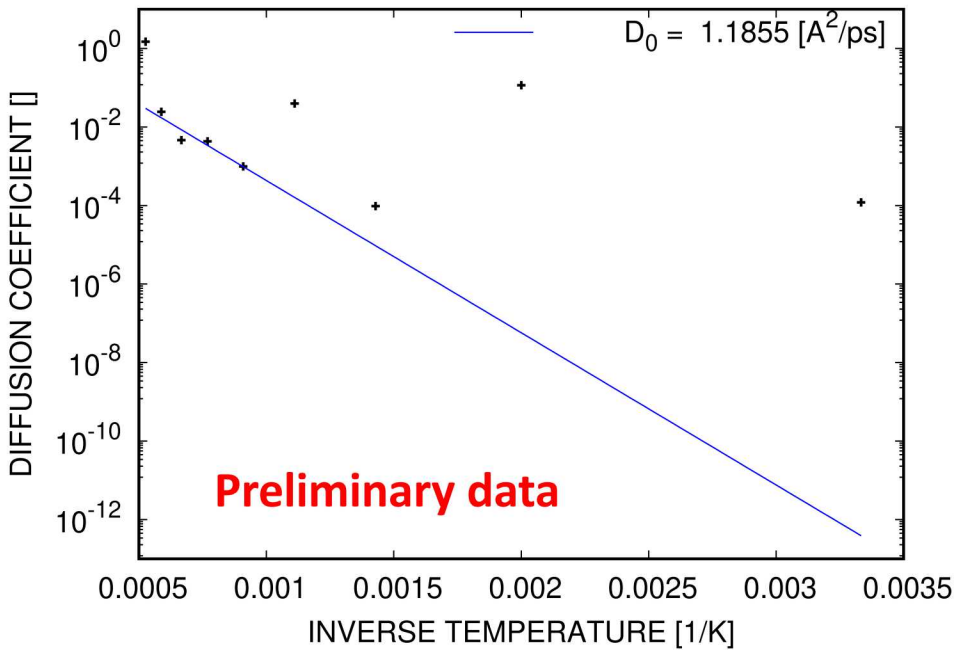


H diffusion in ZrO (monoclinic–low T phase).

H diffusion in ZrO_2

COMB is a variable charge potential where charges are calculated by electronegativity equalization. The short-range repulsion and attraction are Tersoff like. It is very computationally expensive but does represent oxides.

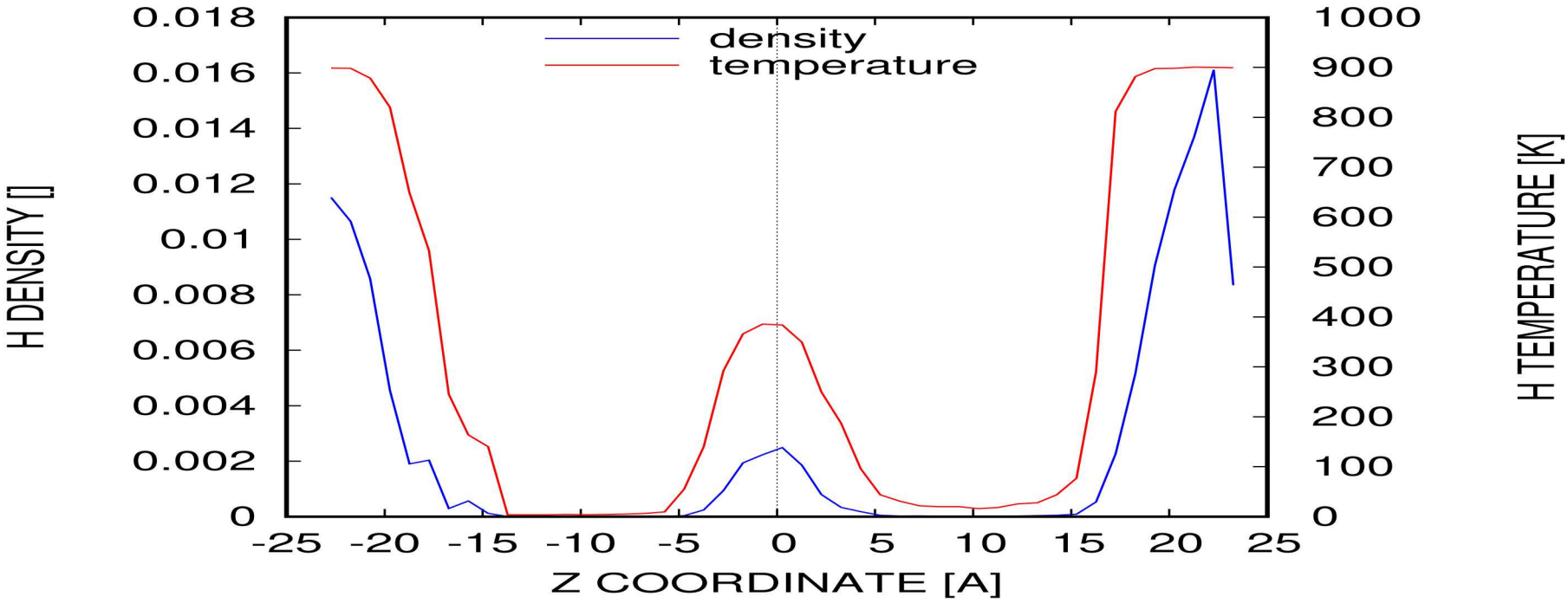




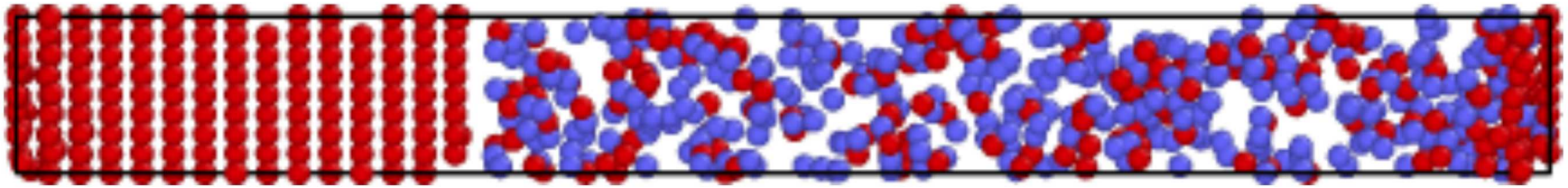
This is data is from an insufficient number of time samples

H diffusion in $Zr|ZrO_2$

Assessing diffusivity of components in a heterogeneous system is difficult

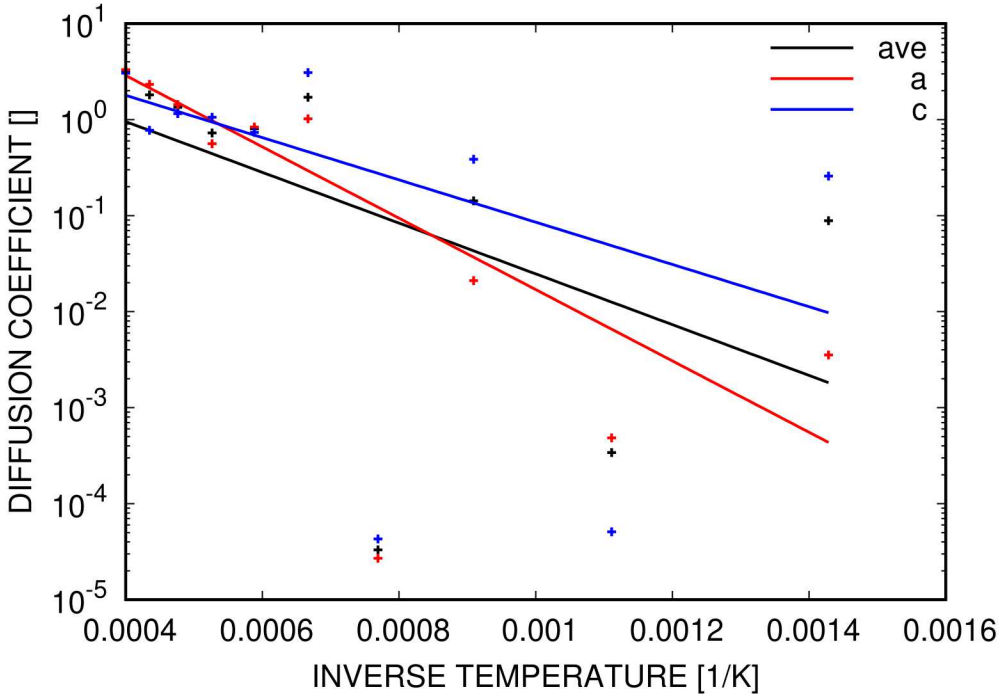


Preliminary data



Ni diffusion in Zr

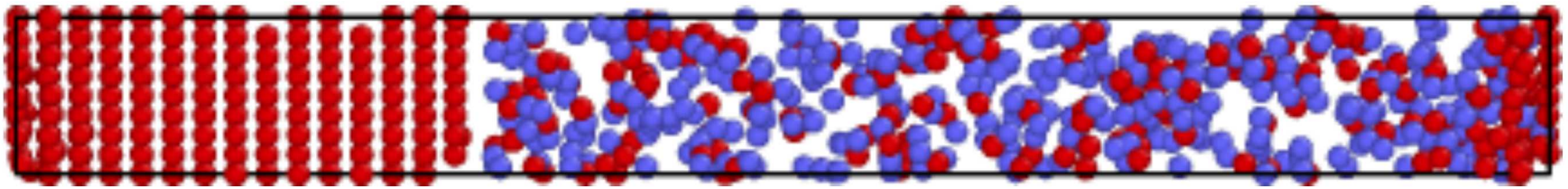
In the literature is is stated that Ni order of magnitude more mobile in Zr than reverse.
We can estimate Ni diffusion with the EAM potential



Preliminary data

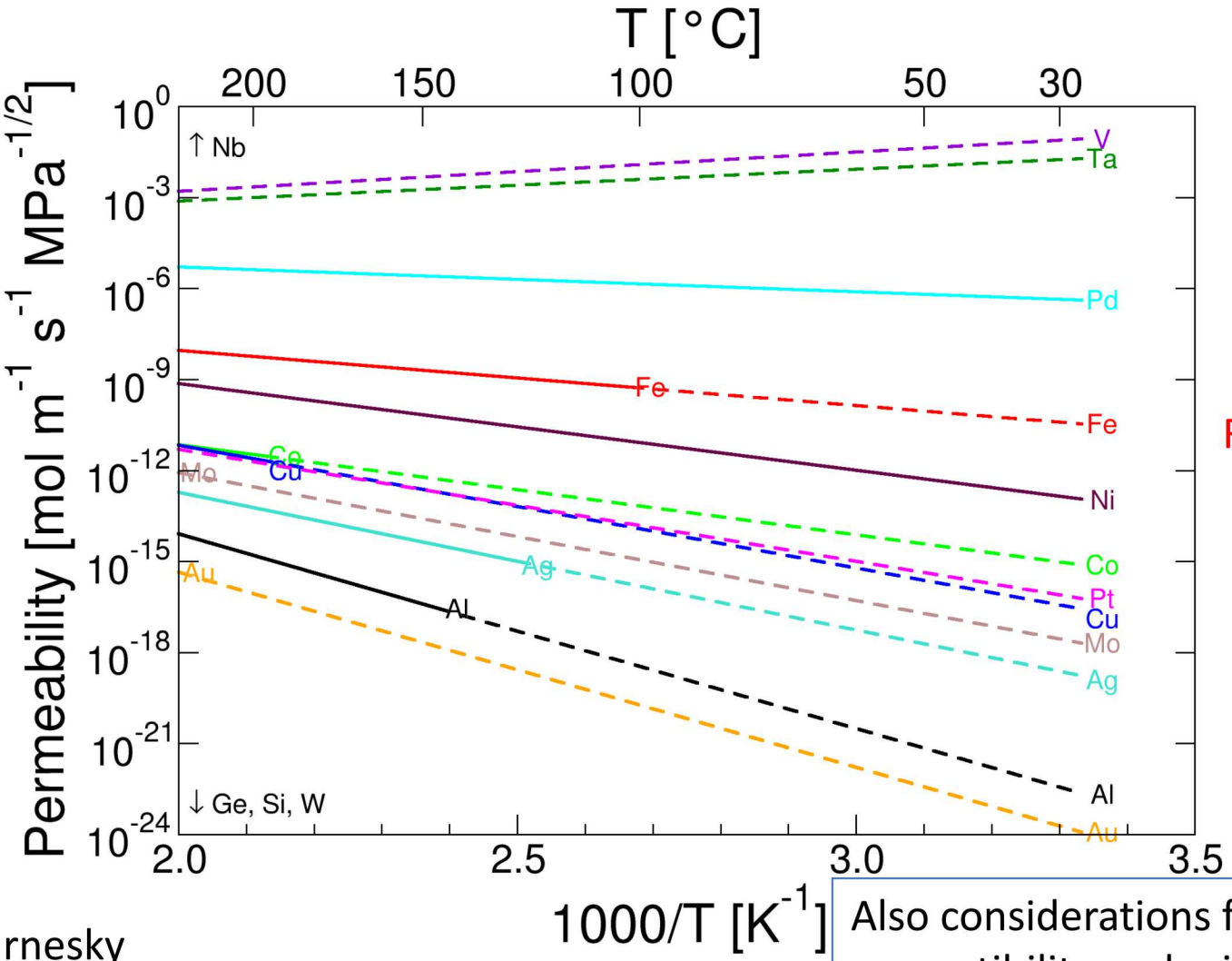
Conclusion

- A diffuse/intermixed NiZr interface between pure Ni and Zr is likely, possibly amorphous but likely crystalline
- This region could have much lower H-diffusivity than HCP Zr, and perhaps FCC Ni
- Best to avoid layers that are disordered and high in Zr
- Work to obtain a full NiZrH EAM potential is ongoing. With it I'll redo the diffusion calculations
- It can also be used to estimate H solubility in NiZr phases
- The effects of interstitials & vacancies can be calculated but for limited crystal structures given the current potential (crystanline Ni_xZr_{1-x} is only stable at low temperatures)
- A NiZrOH potential that could model oxides, coatings and hydride and other phases would be challenging but we have DFT calibration data



Conclusion

- Alternative materials with high permeability (diffusivity x solubility)



Ferritic steel?

Karnesky

http://arc.nucapt.northwestern.edu/refbase/show.php?record=10704

Also considerations for reactor compatibility and miscibility with Zr

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