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Sandia National Laboratories

Machine-learned Interatomic Potential Development for H Trapping in ZrC Strengthened W

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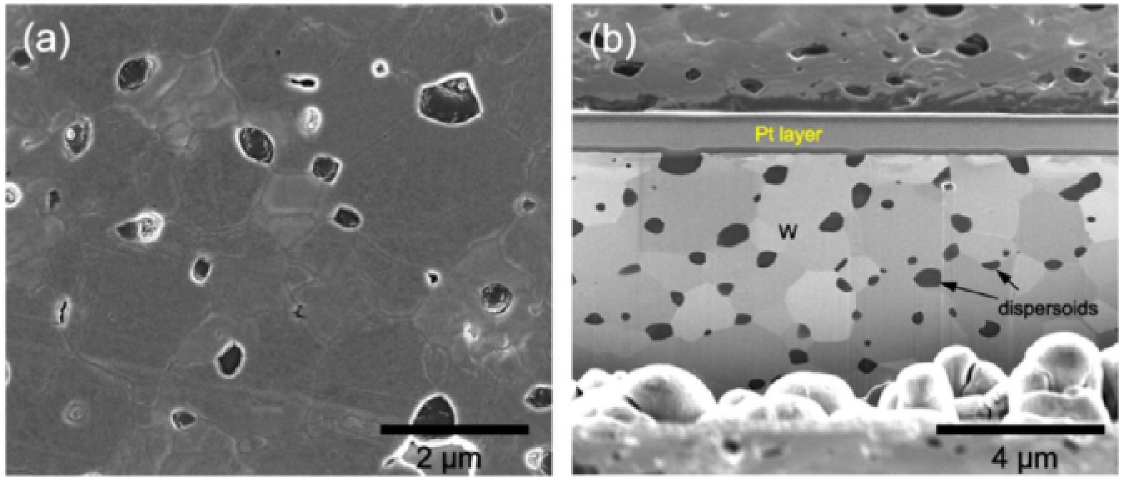
Outline

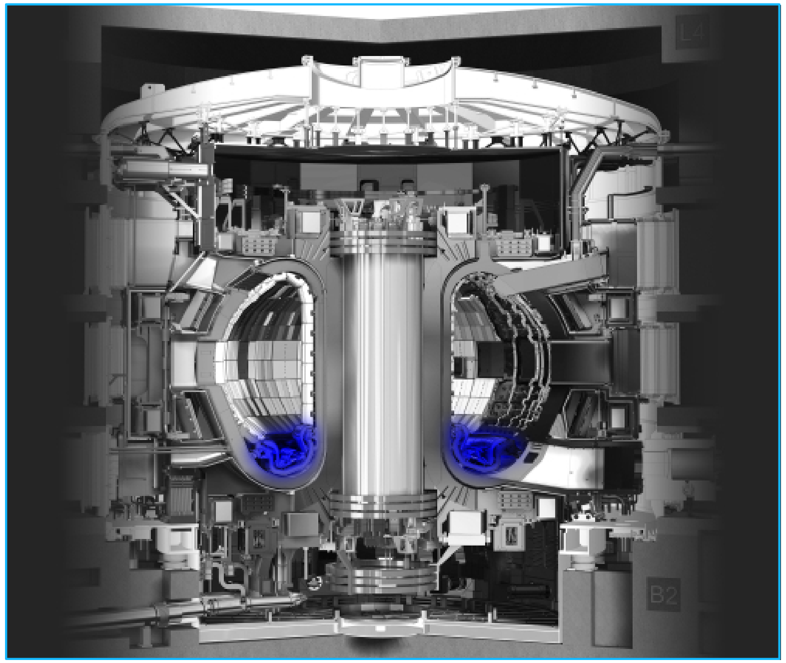
- Application/Needs of the Potential(s)
- Potential Development Workflow (W-ZrC)
    - DFT Training Data
    - Objective functions vs. energy and force errors
- Example of the W-ZrC Potential's Performance: Bicrystal Tensile Tests
- Preliminary W-ZrC-H properties and performance

How can we predict fusion material performance?

- W suffers from a high brittle-to-ductile transition temperature (>473 K)[2] and may undergo recrystallization and grain growth above 1000 K[3].
- Strengthening W with zirconium carbide (ZrC) can improve mechanical properties, but these mechanisms and effects on hydrogen fuel retention are not well understood.



SEM of ZrC dispersoid strengthened W[4]



The divertor in a fusion reactor will control the waste and withstand the highest heat loads of the machine[1].

[1] www.iter.org/mach
[2] Xie, et al. Sci. Rep. 5, 1-11 (2015)
[3] Lang, et al. J. Nucl. Mater. 545, 152613 (2021)
[4] Kolasinski, et al. Int. J. Refract. Met. Hard Mater. 60 (2016).

The divertor in ITER is expected to reach up to 2573 K under normal operation.

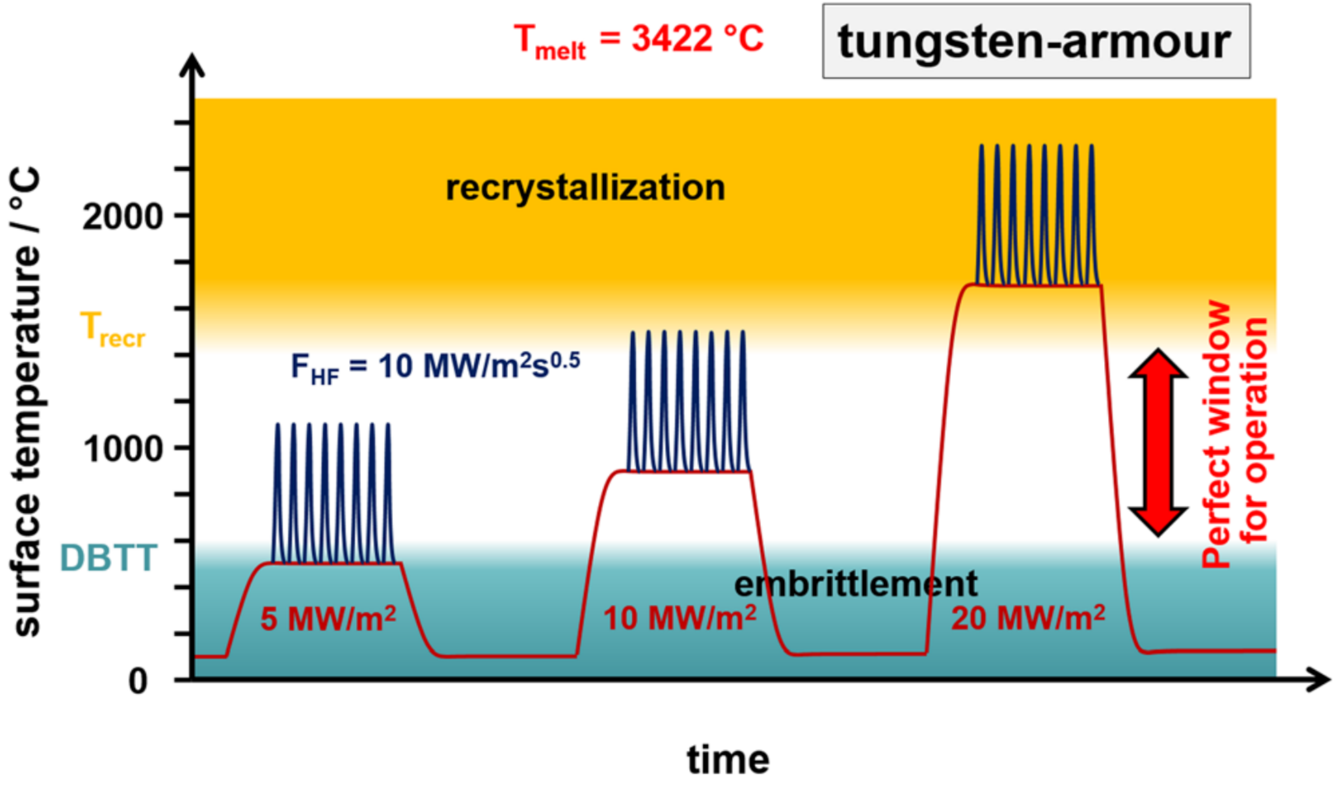


FIG. 3. Schematic presentation of the surface temperature of tungsten-armored divertor targets in ITER at three different power density levels (5, 10, and 20 MW m^{-2}). Thermal spikes caused by mitigated ELMs with an assumed intensity of 10 MW m^{-2} s$^{0.5}$ are shown in dark blue.[16] Reproduced with permission from Rieth *et al.*, J. Nucl. Mater. **519**, 334-368 (2019). Copyright 2019 Elsevier.

Linke, et al. Matter Radiat. Extremes 4, 056201 (2019)

The Spectral Neighbor Analysis Potential (SNAP) can map quantum data to a classical interatomic potential.

Model Form

- Each neighbor position, (r, θ, ϕ), is mapped to a point, (θ_0, ϕ, θ), on the unit 3-sphere.
- The basis can be described with bispectrum components, B_k^i .
- Fitting the linear coefficients, β_k, produces the SNAP potential:

$$E_{SNAP}^i = \beta_0 + \sum_{k=1}^{K} \beta_k (B_k^i - B_k^i 0)$$

Linear Regression

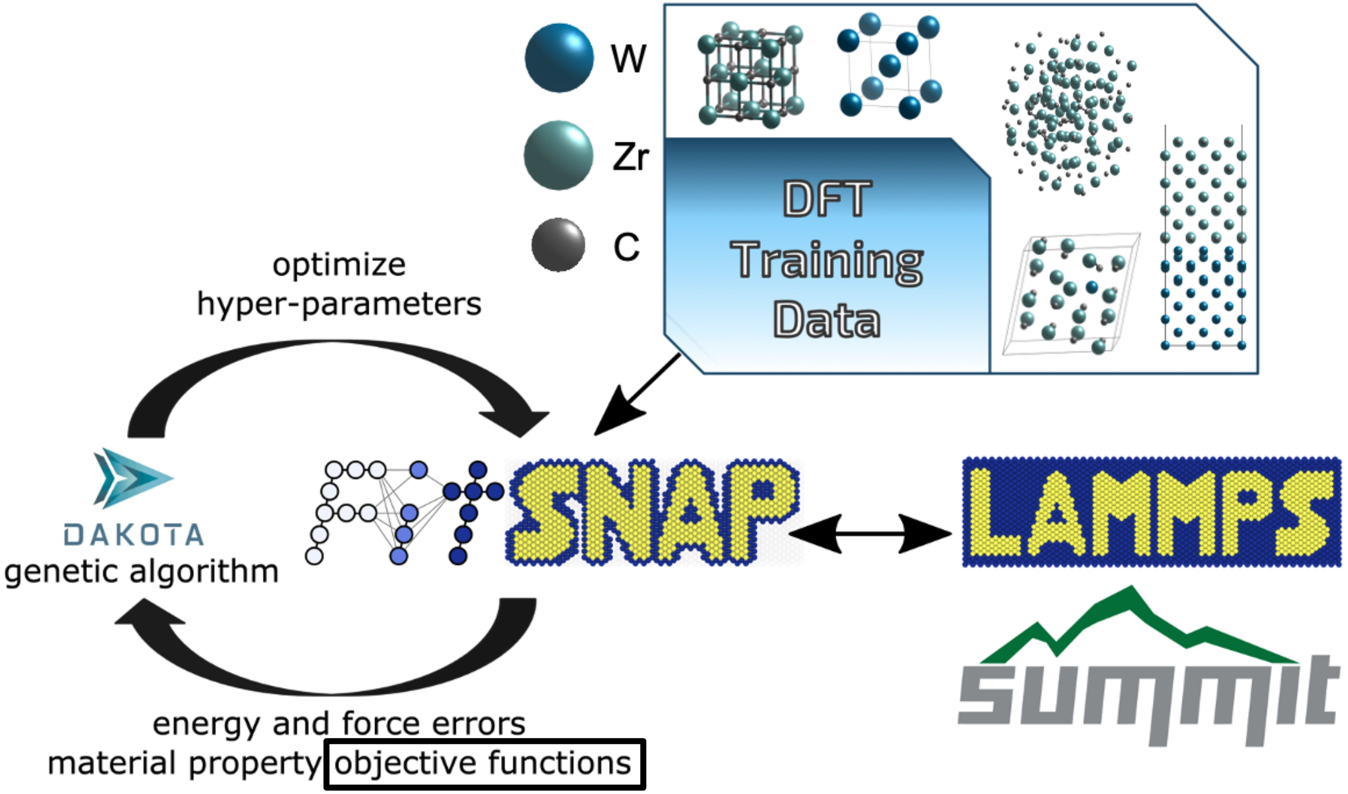
$$\min(||\epsilon \cdot (D\beta - T)||^2)$$

group weight

descriptor prediction

DFT training

Work flow



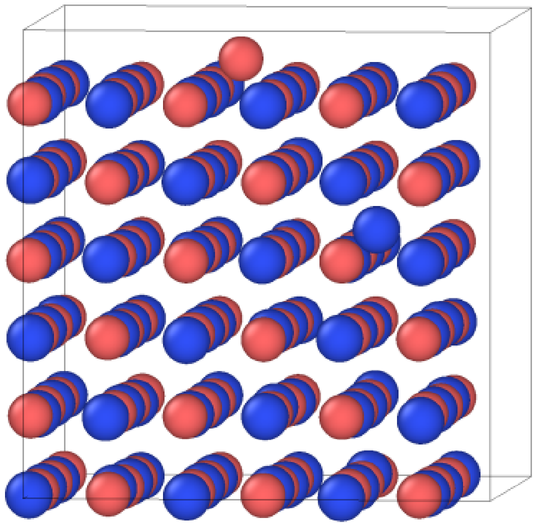
Code available: https://github.com/FitSNAP/FitSNAP

→(Now with docs! https://fitsnap.github.io/)

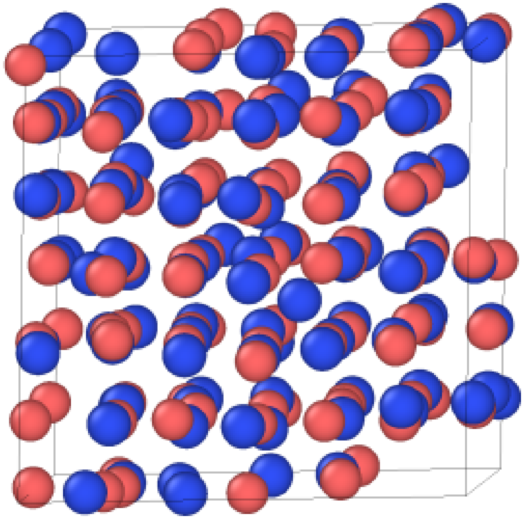
Aidan Thompson et al. / J. Comput. Phys. **285** (2015)
Mitch Wood et al. / Phys. Rev. B **99** 184305 (2019)

What is an objection function? Example using Radial Distribution Function

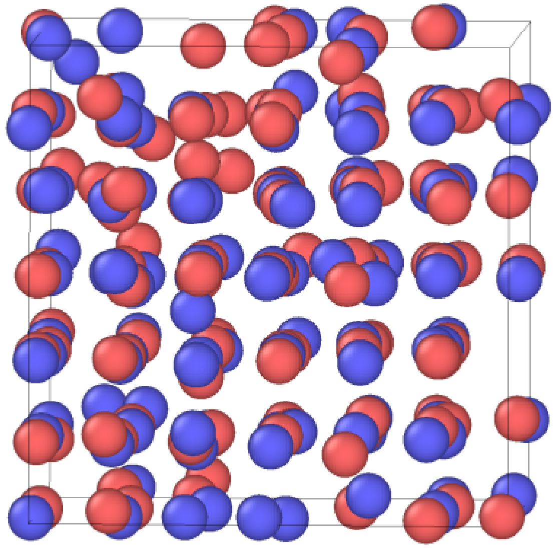
- NPT run @ 2000K
- 3x3x3 supercell with 1U and 1N interstitial
- RDF calculated with 50 bins and 4 Å cutoff
- Objective function takes the absolute value of the difference between SNAP and DFT for each bin
- Throws arbitrary error if atoms cluster



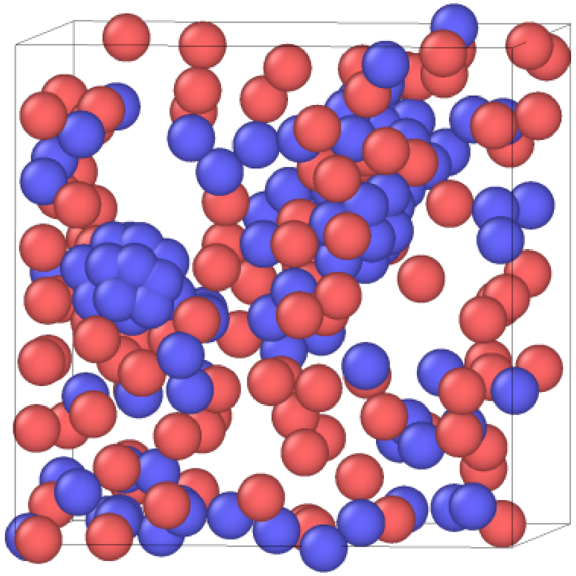
Starting structure



AIMD after 817 fs

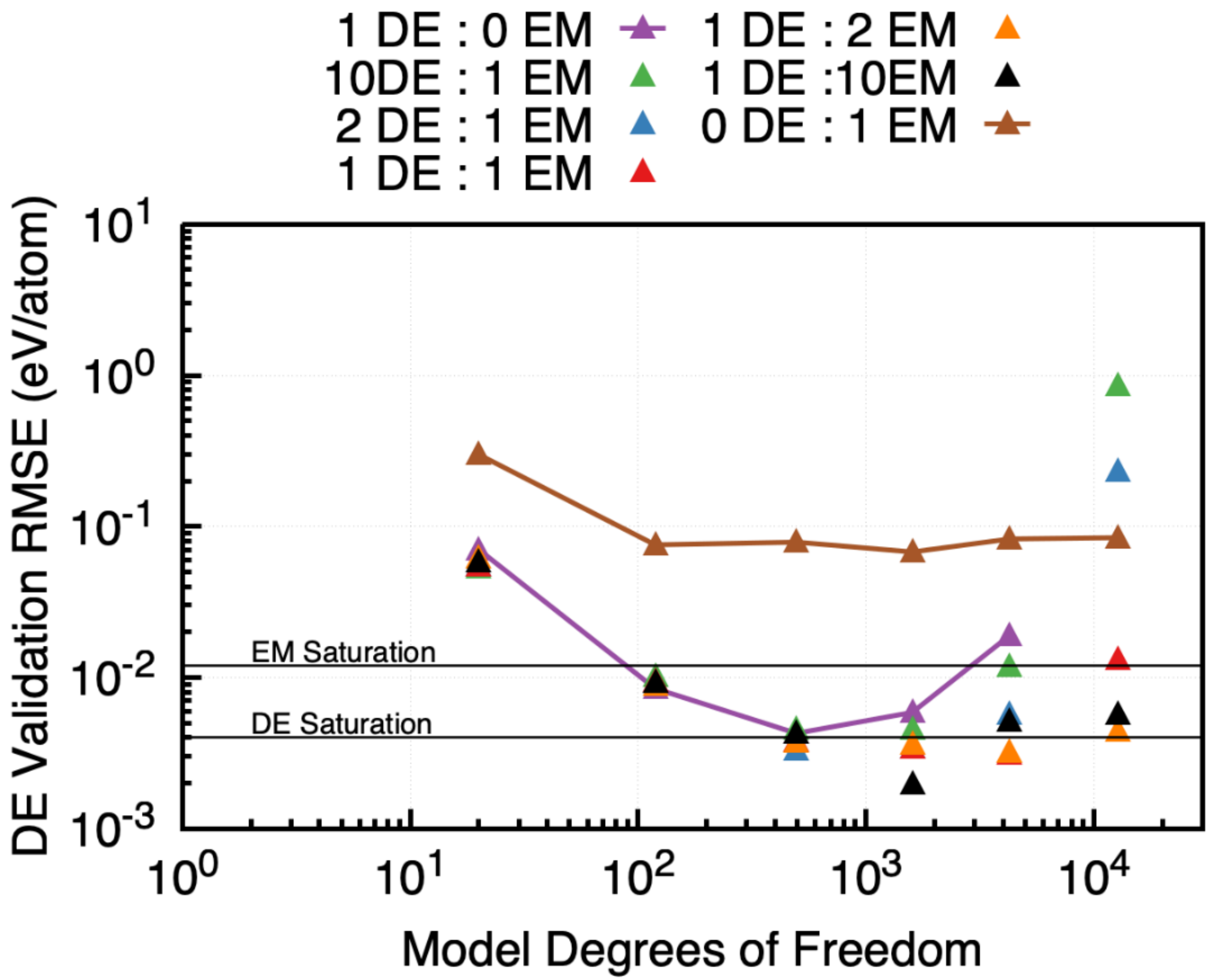


Obj. f. value 5.33



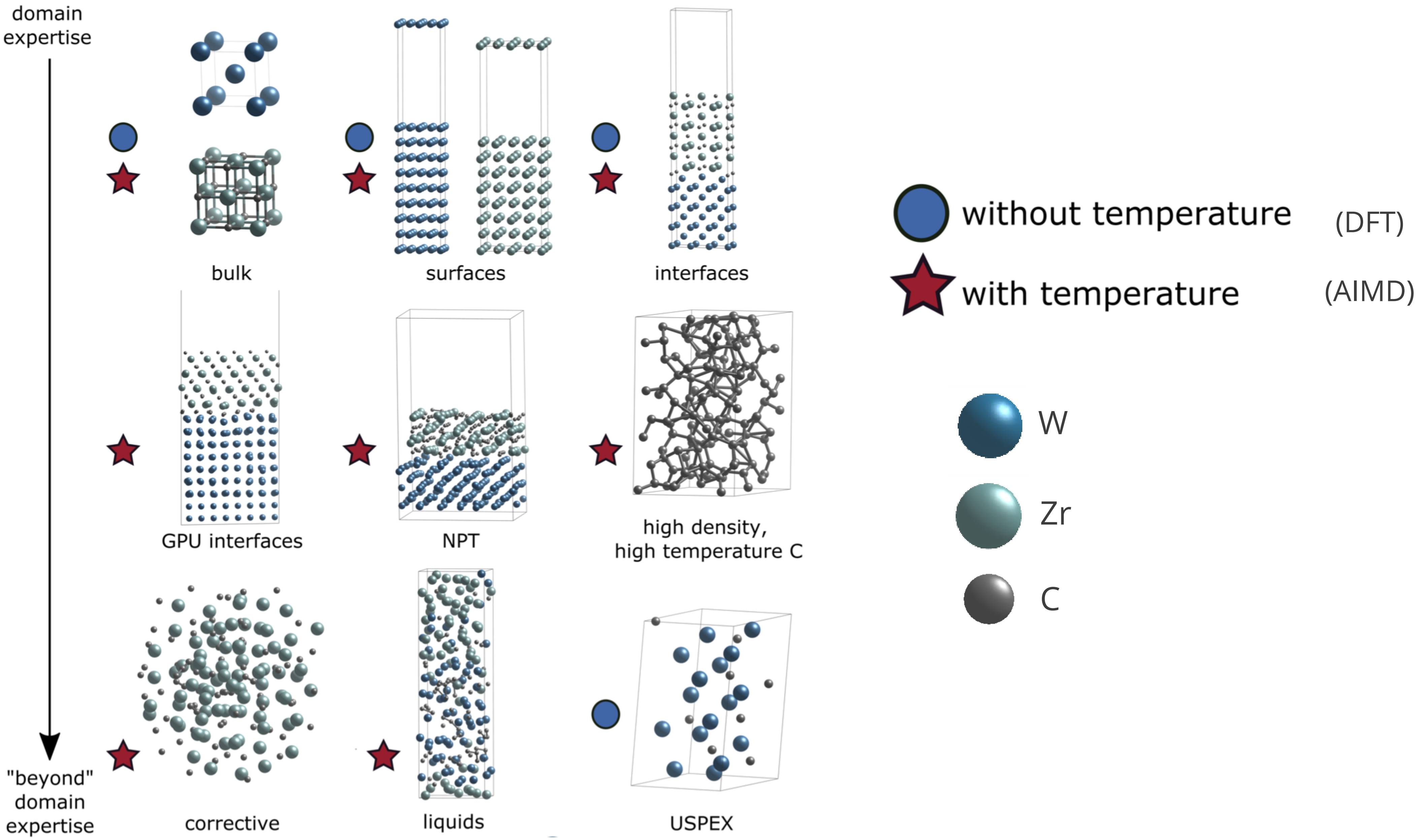
Penalty value 2113

How to make a transferable potential – "domain expertise" (DE) vs. "entropy maximized" (EM)



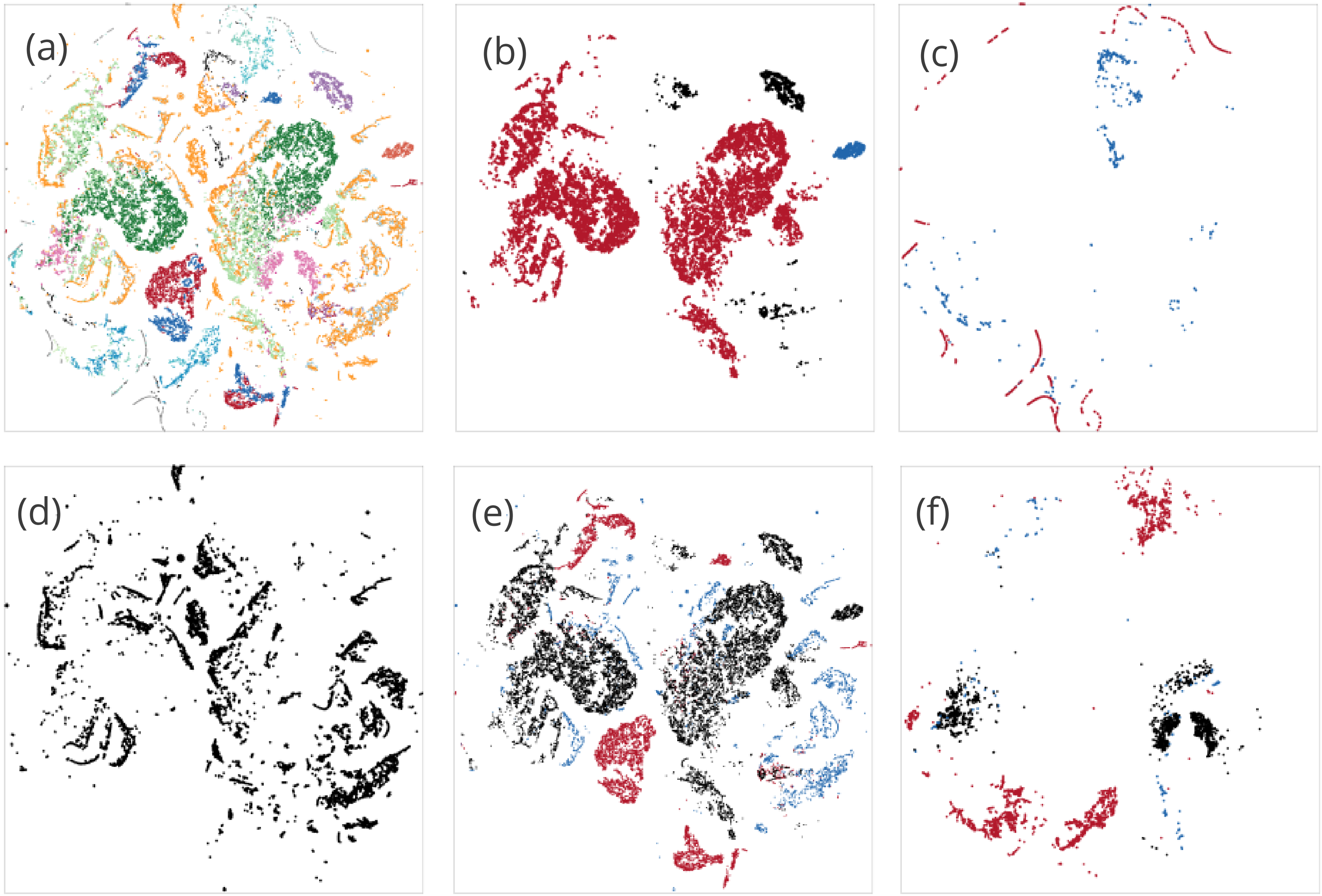
David Montes de Oca Zapiain, et al. npj Comput. Mater. 8 (2022)

What is in the training set? (~9,000 structures)



Ember Sikorski et al. / J. Chem. Phys. **158** (2023)

How does this configuration data look to SNAP?



t-SNE visualization of SNAP descriptors in 2D

(a) All data
*labels omitted

(b) By constituents
W ZrC C

(c) Ground states
EOS unit cells, defects, etc.

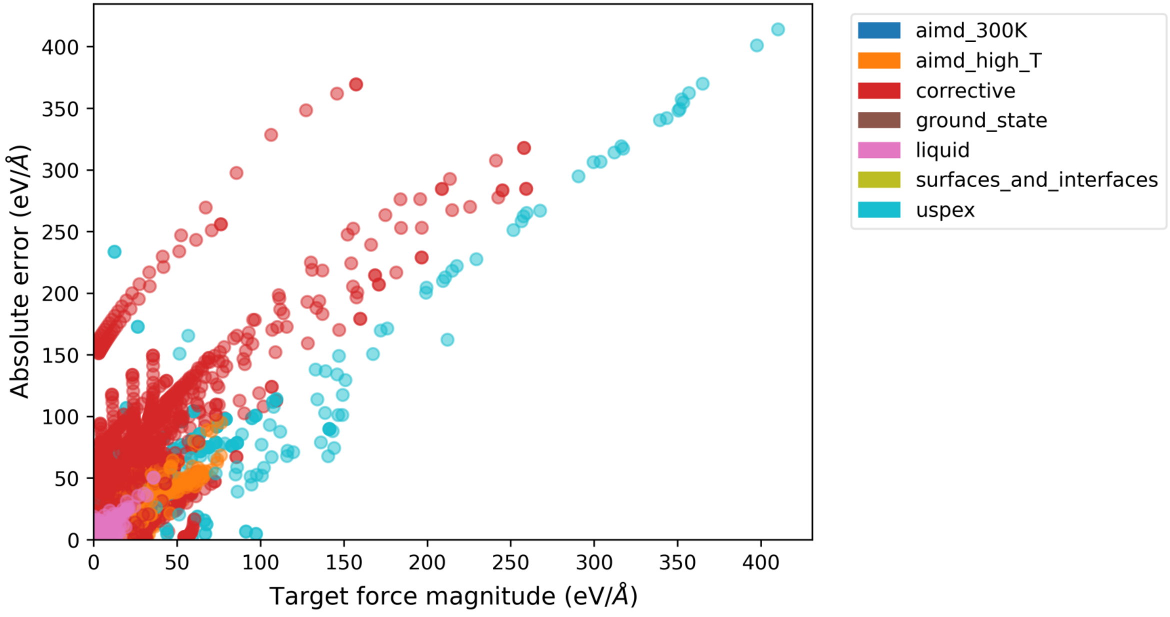
(d) AIMD – 300K

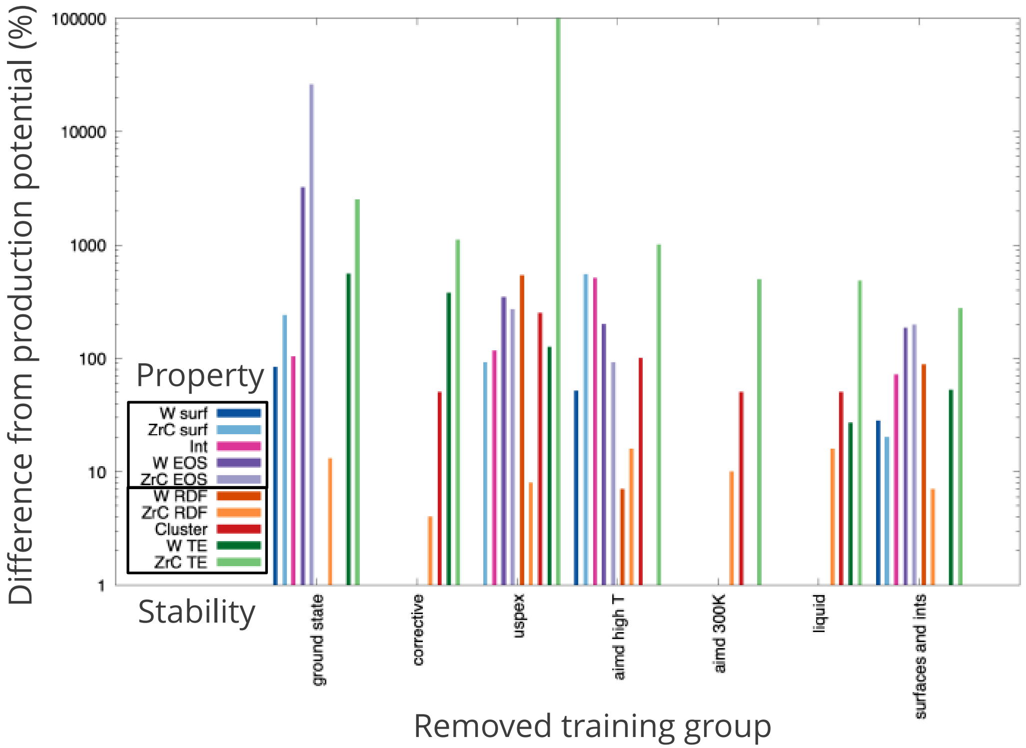
(e) AIMD – 1000 – 5000K
W/ZrC/C >200 atoms interfaces

(f) Beyond domain expertise
liquid USPEX corrective

Ember Sikorski et al. / J. Chem. Phys. **158** (2023)

Do low force and energy errors prove a potential is good?





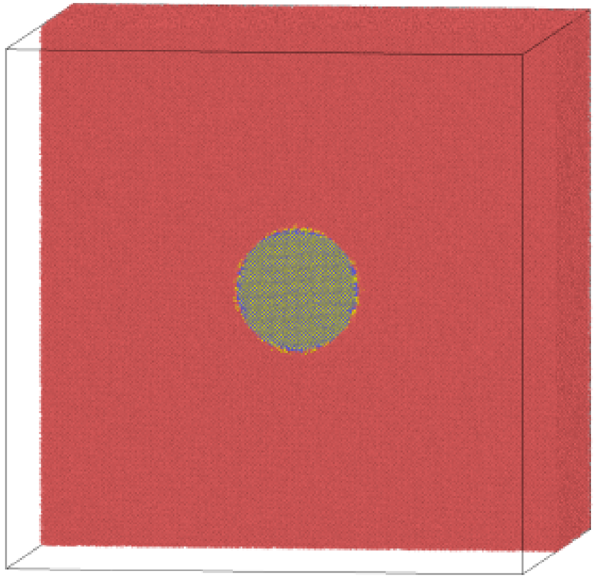
	Production Potential	Drop ground state	Drop corrective	Drop USPEX	Drop AIMD High T	Drop AIMD 300K	Drop Liquid	Drop surfaces and ints
Energy error (eV/atom)	0.43	0.34	0.43	0.25	0.41	0.55	0.44	0.44
Force error (eV/Å)	1.02	0.78	0.95	0.62	1.53	1.22	0.95	1.04

→ No, low force and energy errors may be promising, but provide no information on the accuracy of material properties or dynamics.

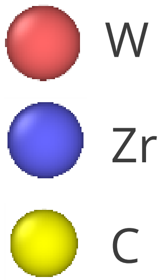
Ember Sikorski et al. / J. Chem. Phys. **158** (2023) (see supplementary material)

The W-ZrC SNAP agrees well with DFT and can now run 10s of nm/10 million atom structures.

- The W-ZrC SNAP potential yields material properties in good agreement with DFT values for lattice parameter, *a* (Å), bulk modulus, *B* (GPa), and surface energies, E_{surf} (eV/Å).
- Using the W-ZrC SNAP potential we can run millions of atom simulations at divertor temperature ranges (~373 – 2573 K[8]).



Spherical ZrC in crystalline W at 1700 K (~10 million atoms, 56 nm per side)

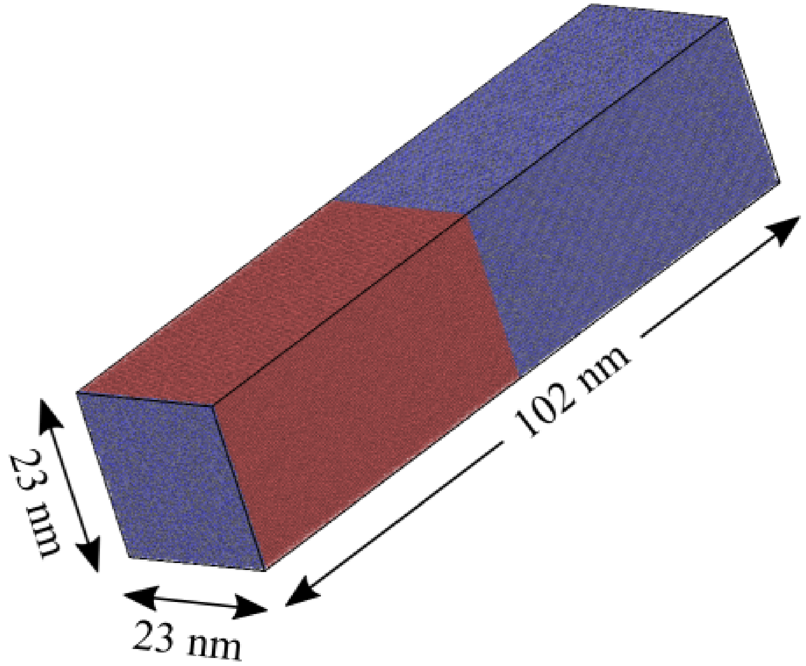


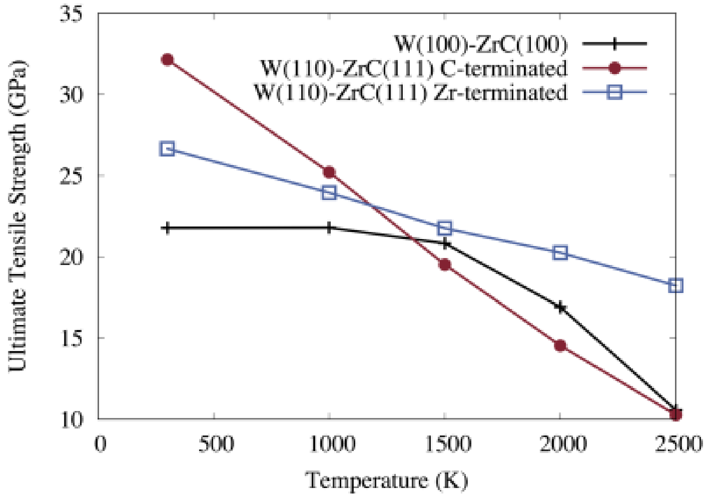
Material properties predicted by DFT vs. SNAP

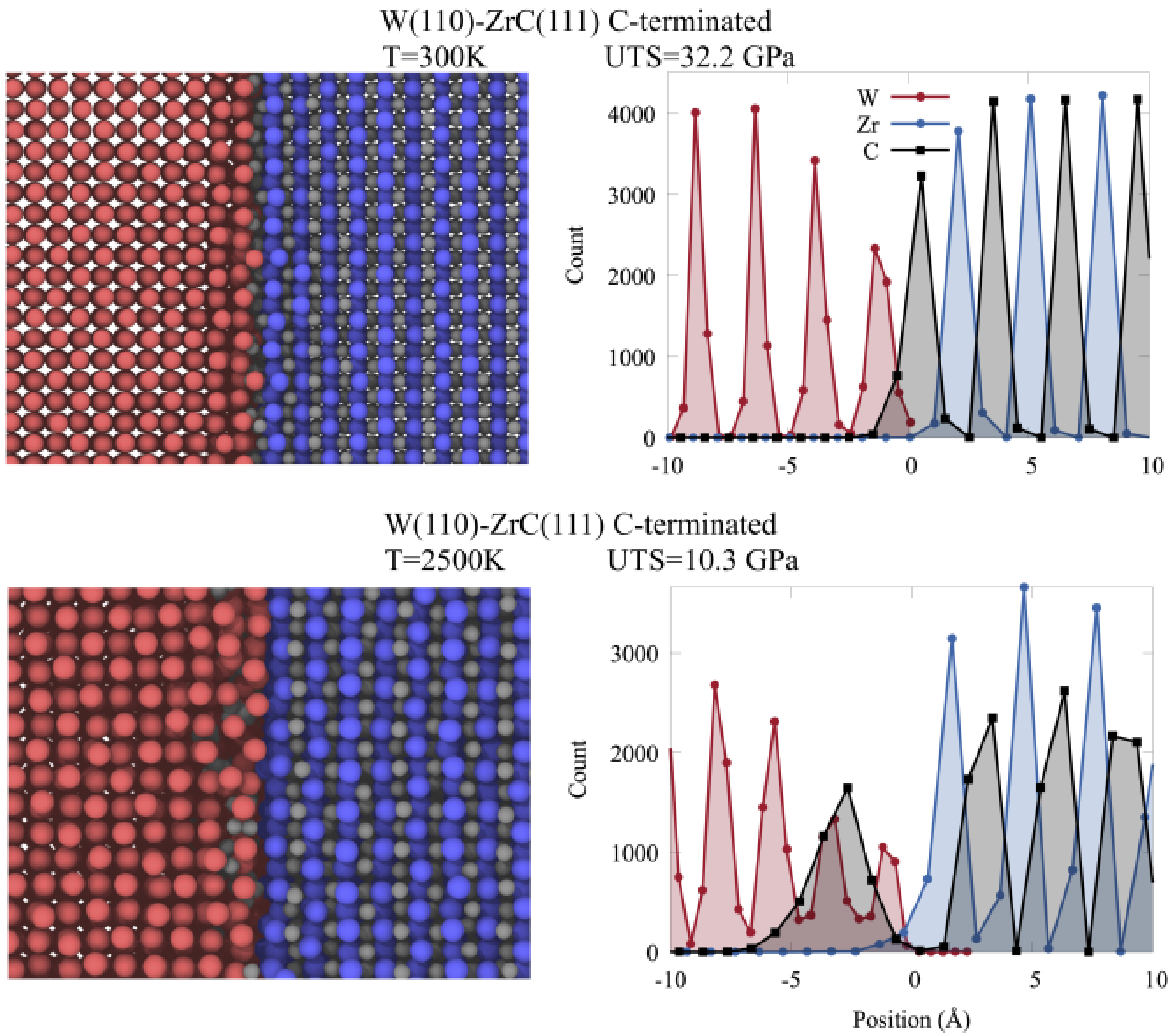
	B_W	B_{ZrC}	E_{surf} W (100)	E_{surf} W (110)	E_{surf} ZrC (100)	E_{surf} ZrC (110)	a_W^{2600K}	a_{ZrC}^{2600K}
DFT/expt.	301.4	216.0	4.13	3.18	1.63	3.31	1.31	1.85
SNAP	303.3	209.0	3.38	3.22	1.40	2.75	1.05	1.50

Ember Sikorski et al. / J. Chem. Phys. **158** (2023)

Example of W-ZrC Performance: bicrystals for tensile testing







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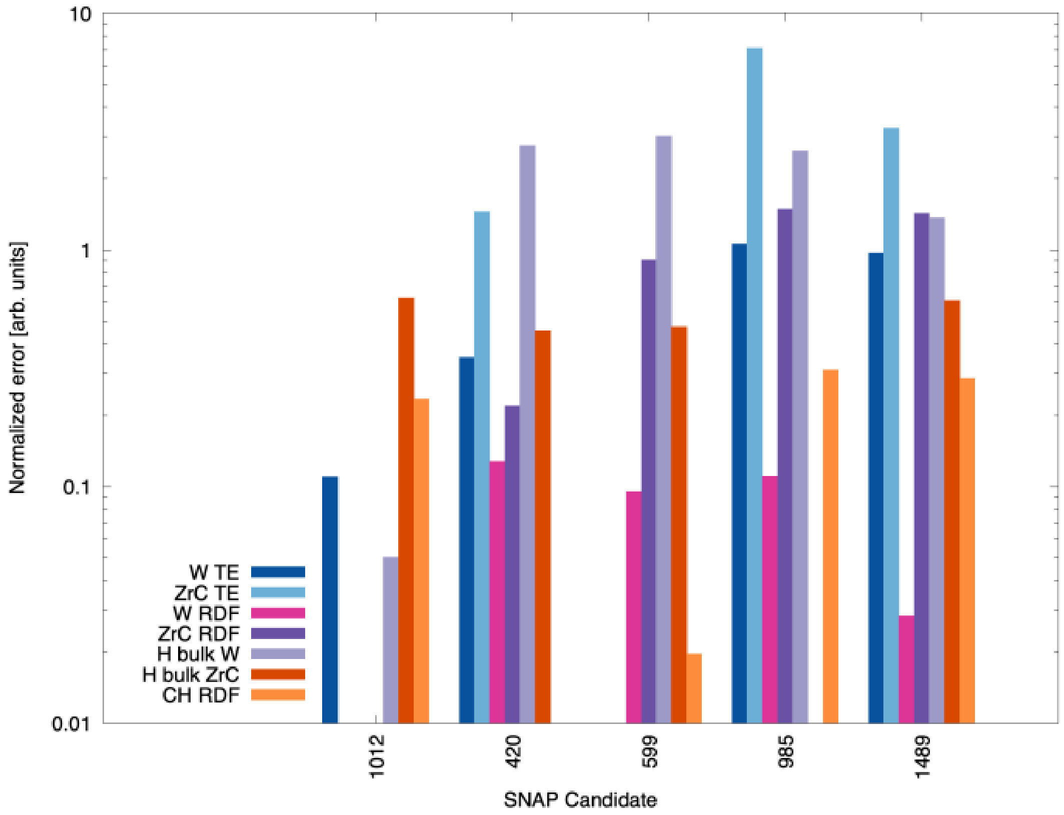
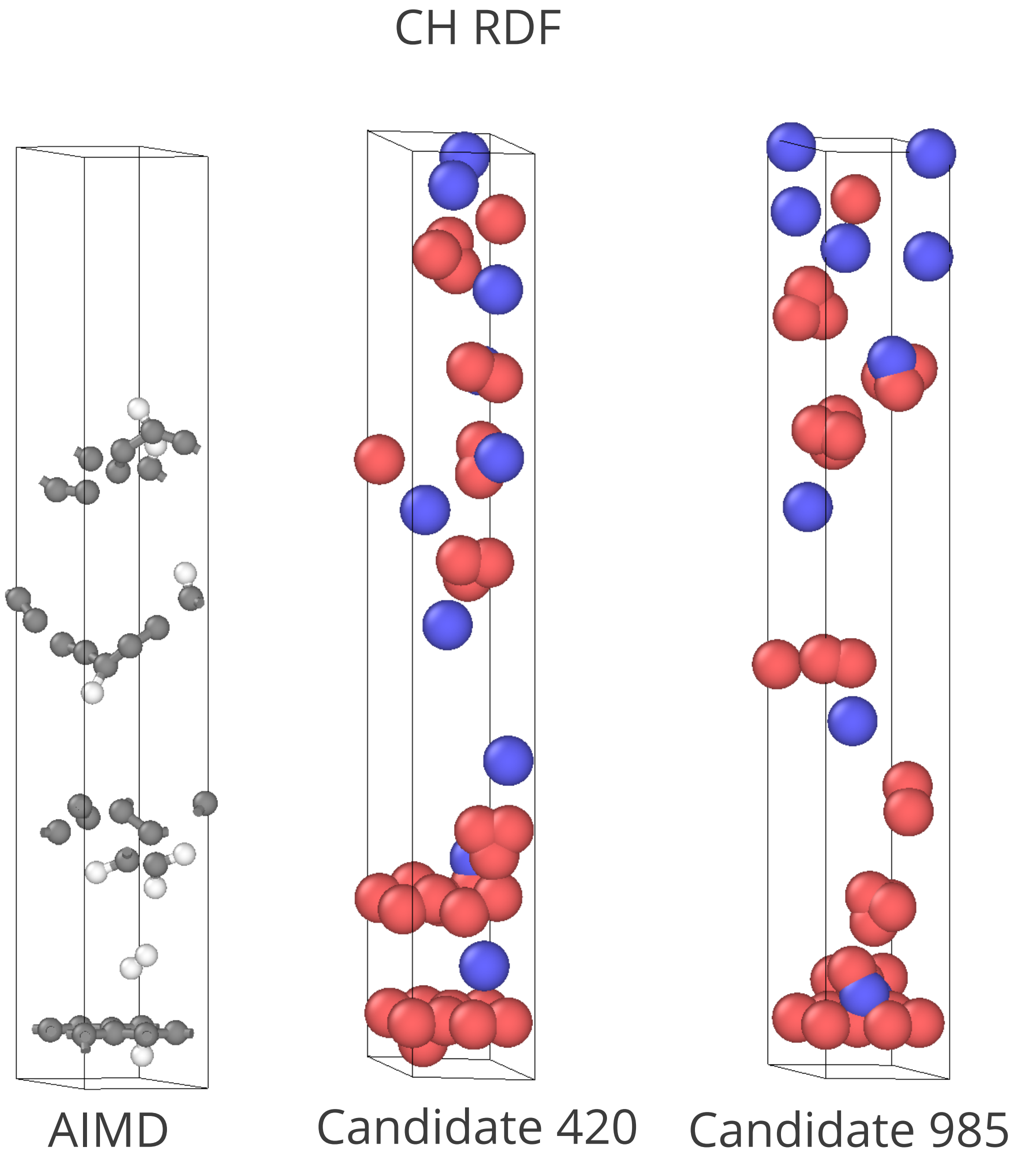
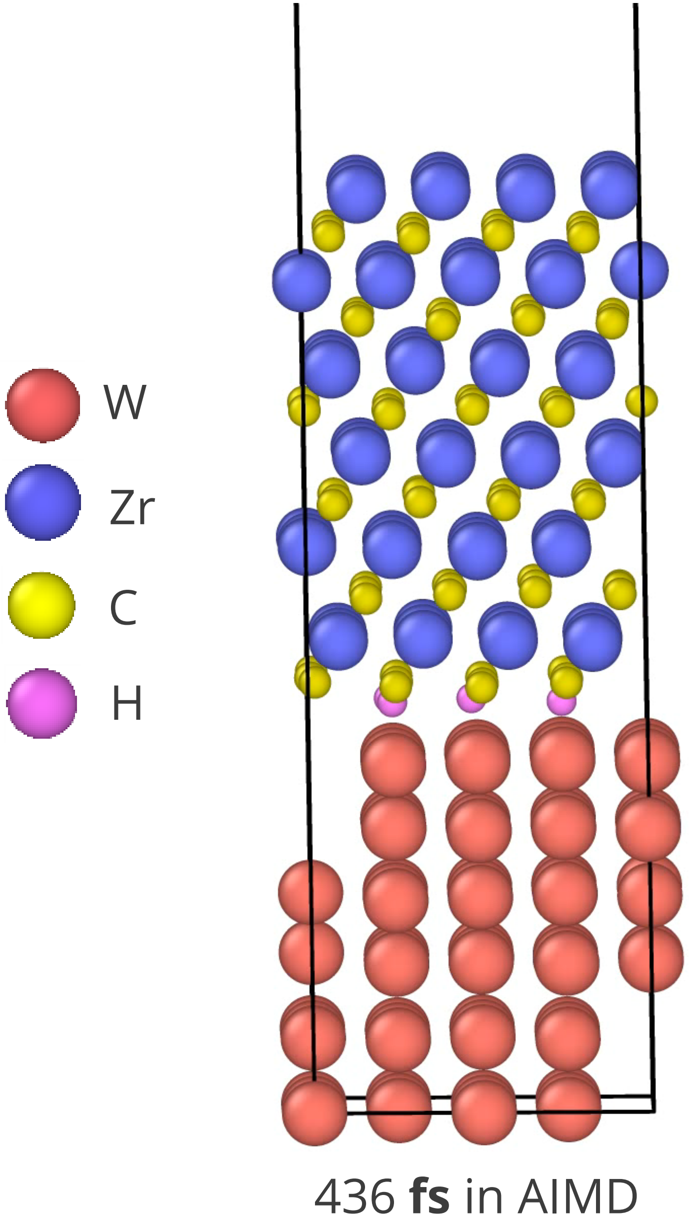
SNAP candidates often exhibit trade-offs.

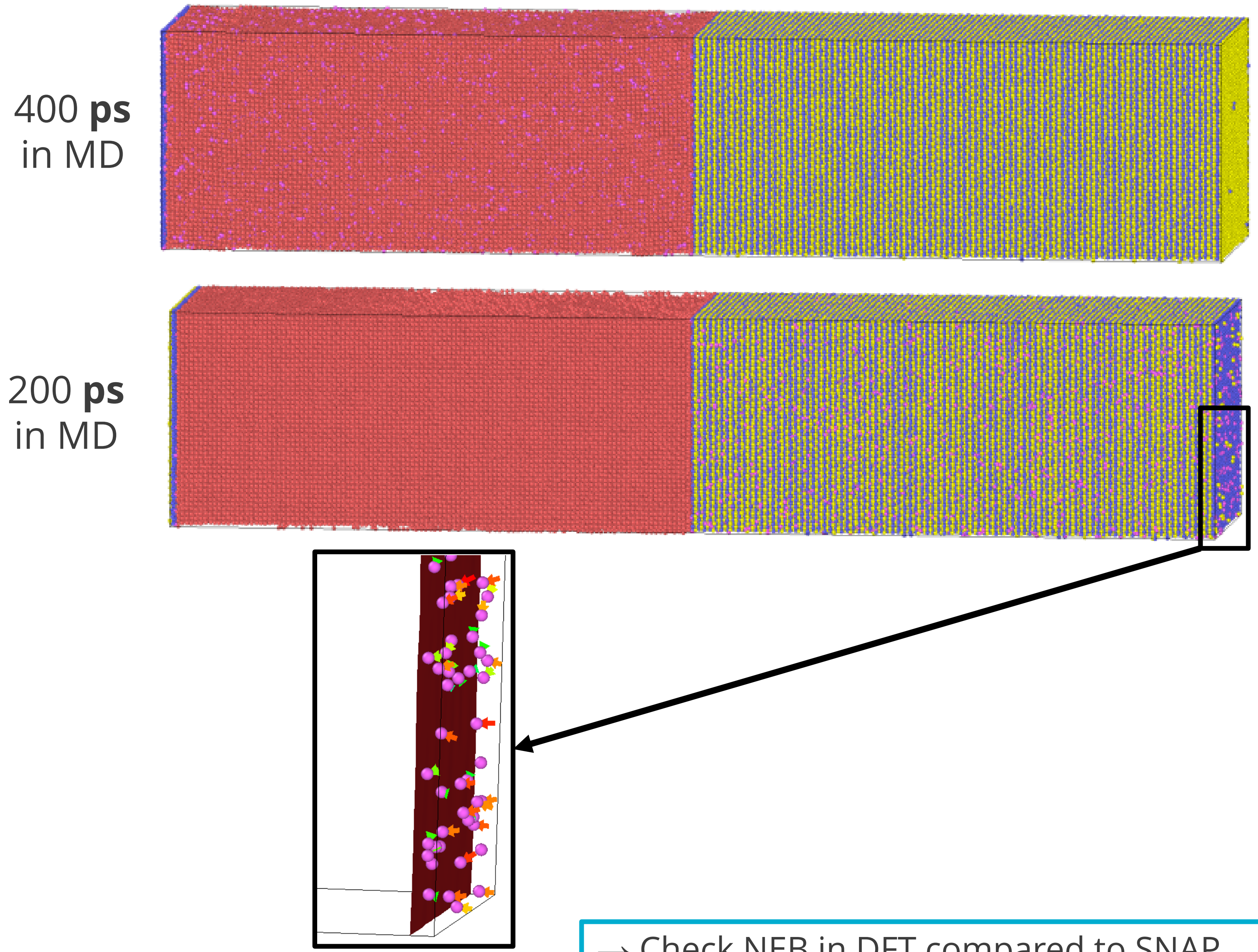


	SNAP (1012)	DFT
H tetrahedral - W	-0.89	0.88
H octahedral – W	-0.71	1.26
H tetrahedral - ZrC	4.53	11.33
H substitional C - ZrC	1.61	-1.44



W-ZrC-H (SNAP candidate 1012) is stable at 1000 K.

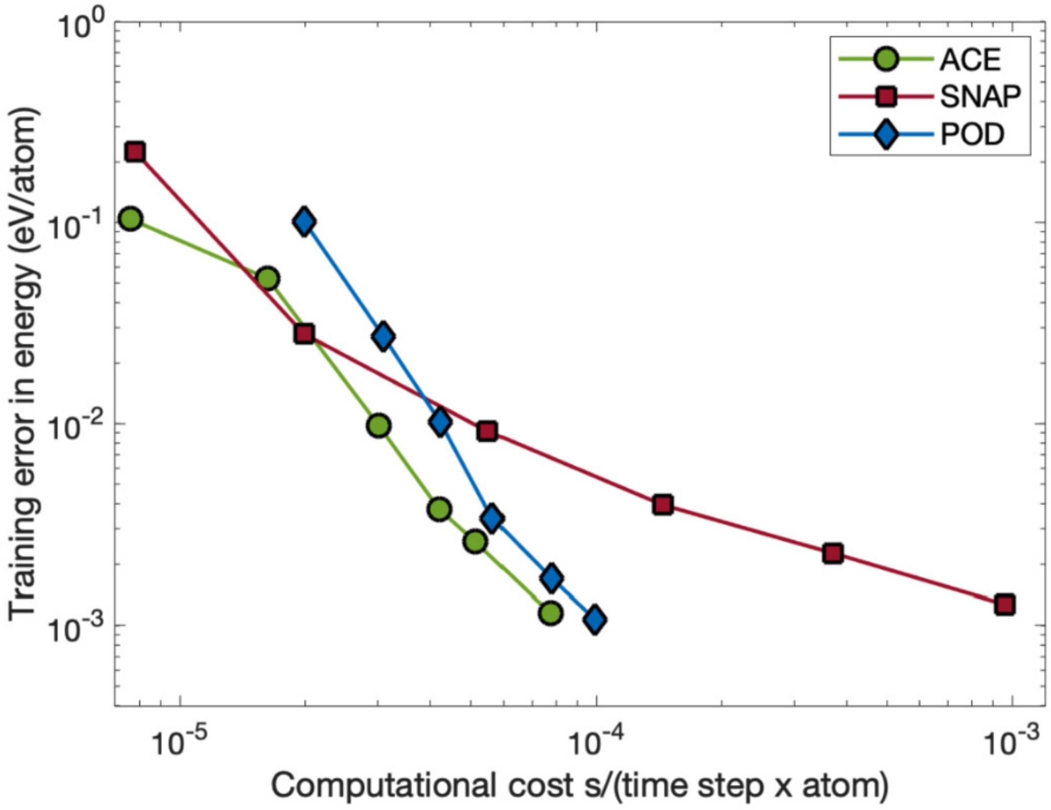




→ Check NEB in DFT compared to SNAP.

Remaining Machine-Learned Potential Questions/Next Steps

- Can we combine objective function optimization with other MLP forms (ACE, Neural Networks, POD, etc.)? Will these yield lower force, energy, and objective function errors?
- Can we use active learning alongside objective functions?
- What is the best way to capture rare events? Can we train on NEB calculations?



Nguyen and Rohskopf / J. Comput. Phys. **480** (2023)

Summary

- We developed a W-ZrC SNAP potential that agrees well with DFT and can run millions of atom MD simulations at high temperature.
- Robust training sets include “domain expertise” and “beyond domain expertise” structures.
  - → cover large regions of descriptor space with USPEX and large AIMD simulations
- Low force and energy errors alone do not ensure good potentials.
- W-C bonds correlate to high tensile strength, though at high temperature much of the C in terminating layers diffuses into W.
- Preliminary W-ZrC-H potentials exhibit objective function tradeoffs but can run stable dynamics at 1000 K.

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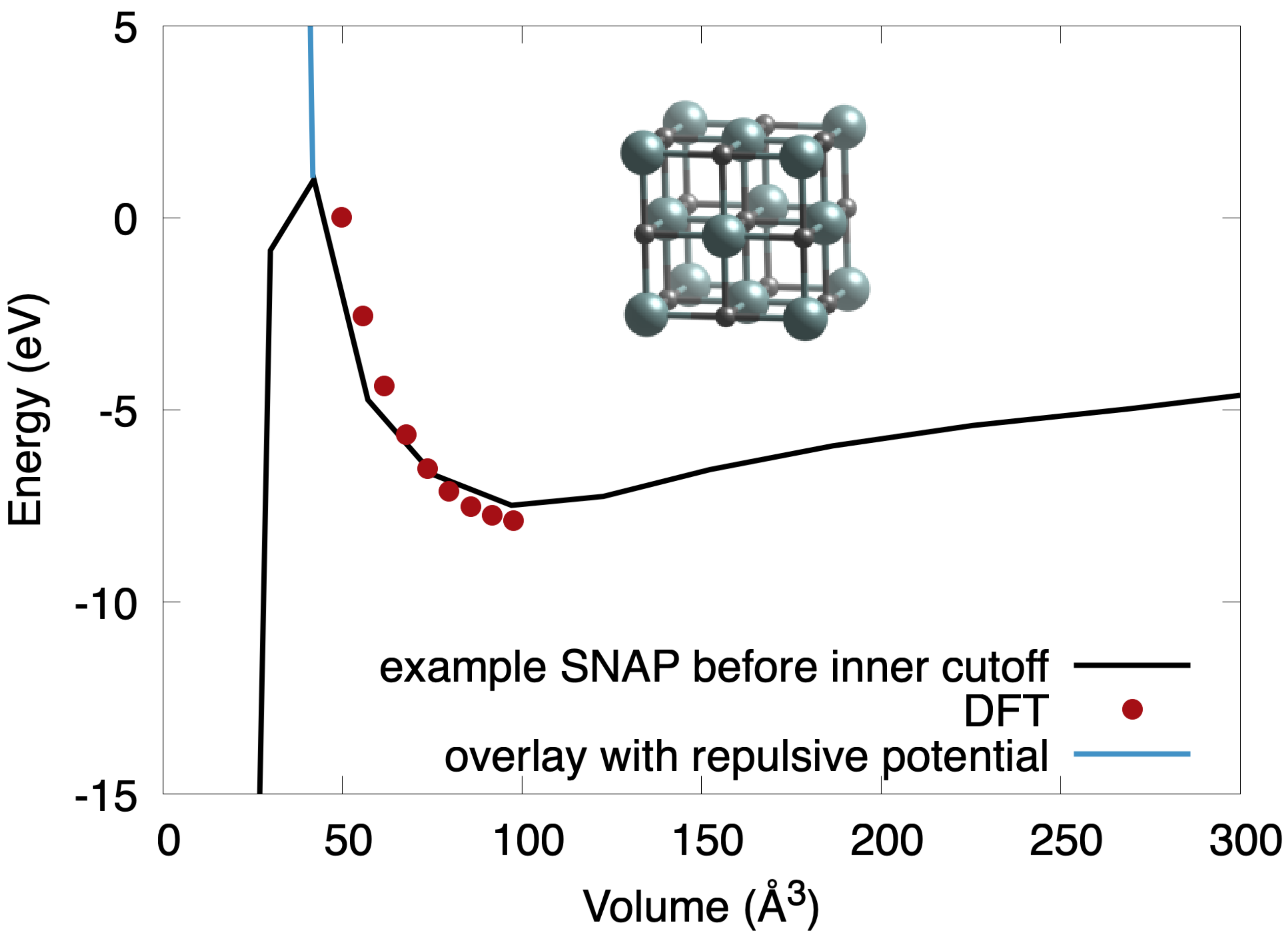
Additional slides

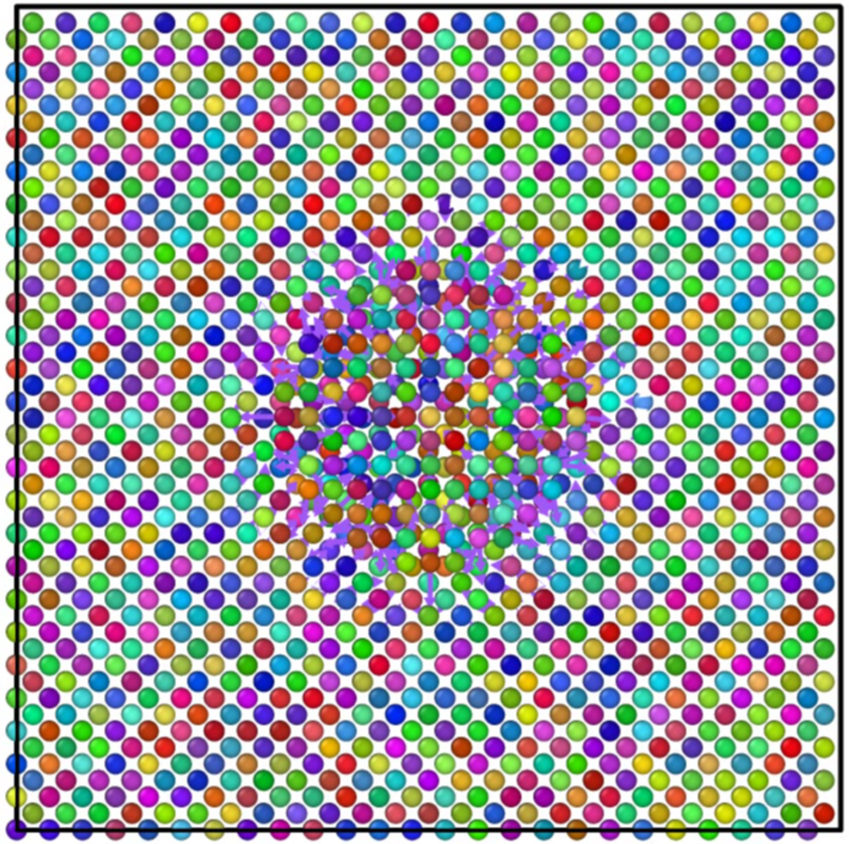
Inner cutoff is now live in FitSNAP and LAMMPS.



Candidate potentials need to be checked in NVT/NPT simulations for stability.

"Laminating"

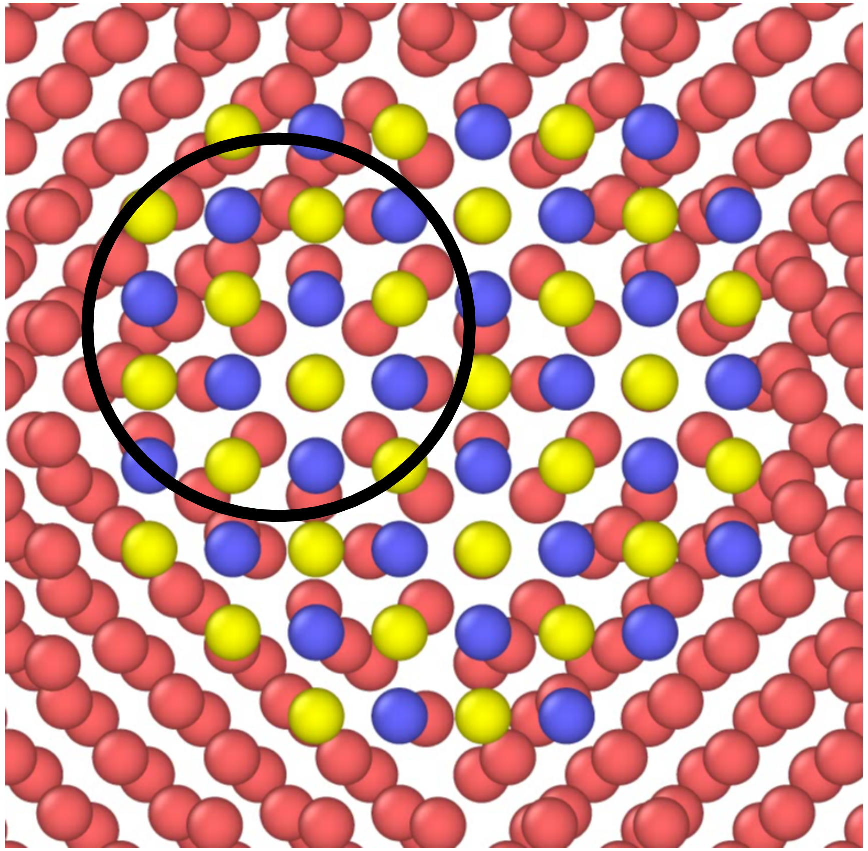
Cluster analysis <2.2 Å



$$\min\left(||\epsilon \cdot (D\beta - T)||^2\right)$$

- Increase group weight for forces on surface training data

"Black holing"



- Add training with low interatomic distances
- Tune zbl overlay
  - Increase repulsive diameter
  - Decrease radial cutoff

Why do we separate DFT training into different groups?

Training groups in the W-ZrC potential:

- ("traditional") DFT
- Surfaces and Interfaces
- AIMD – 300K
- AIMD 1000 - 5000K
- MACtive
- Liquid
- USPEX

"Traditional" DFT

- Unit cells – small supercells (3-64 atoms)
- High cutoff energy (~500 eV)
- Really high k-points (~8x8x8)

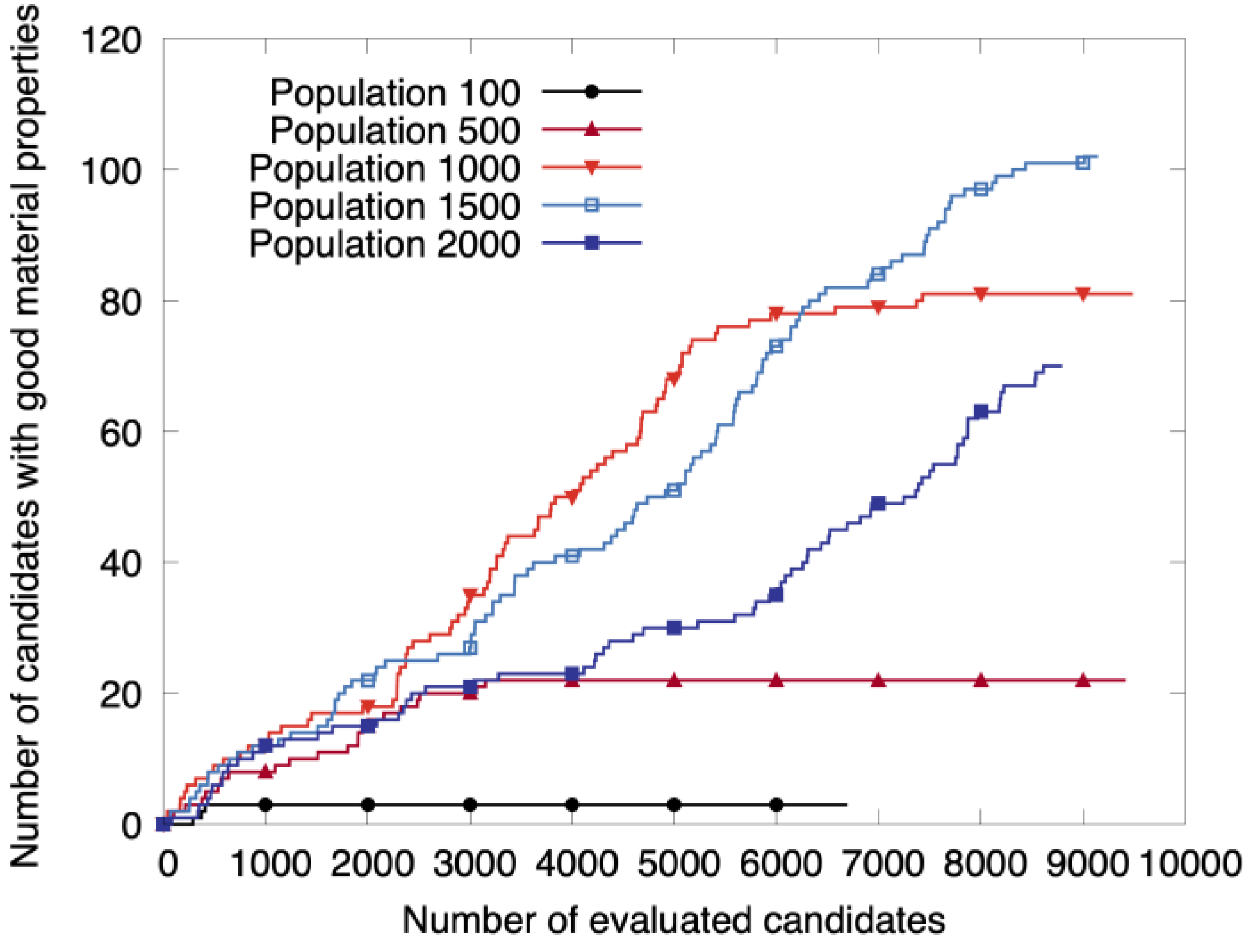
Ab initio Molecular Dynamics (AIMD)

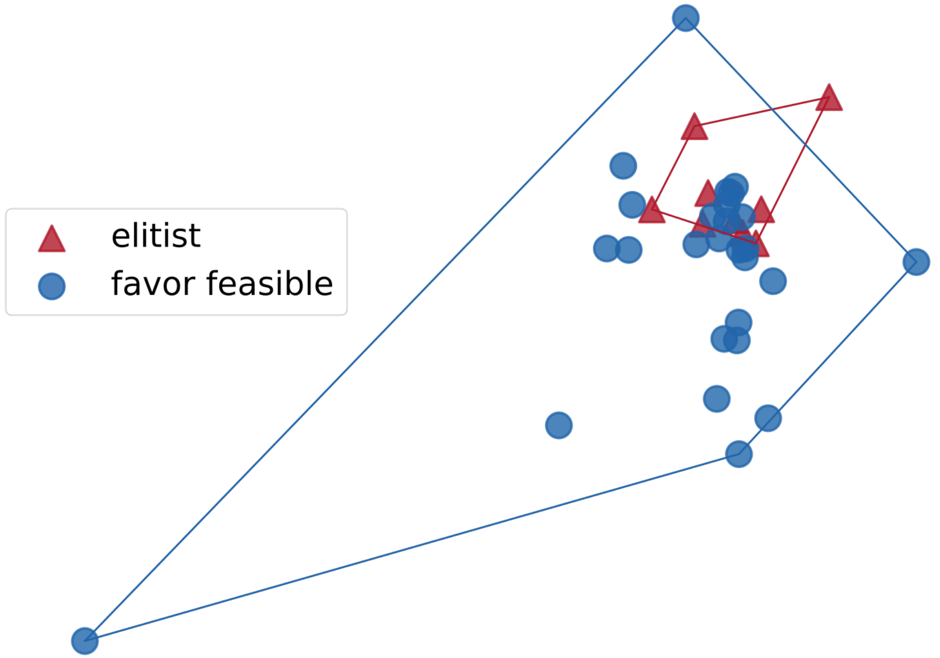
- Supercells (100-700 atoms)
- Really high cutoff energy (~500 - 750 eV)
- Sampled at gamma point (1x1x1)

Group weights of final potential:

dft E	dft F	uspex E	uspex F	surf E	surf F	aimd1 E	aimd1 F	aimd2 E	aimd2 F	mactive E	mactive F	liquid E	liquid F
7.32	3.94	6.41	3.94	3.36	6.52	3.5	3.74	4.1	7.94	3.3	3.62	3.68	5.36

Highlights of Dakota optimization





t-SNE of quality candidates
produced by two replacement types

USPEX – genetic algorithm structure predictor

"USPEX allows to predict crystal structure with arbitrary P-T conditions by knowing only chemical composition of the material."

- USPEX generates trial structures which are then relaxed and evaluated by an external code interfaced with USPEX
- Based on the ranking of the relaxed structures, USPEX generates new structures, which are again relaxed and ranked

1. Create training data (bypass intended use; generate 1 giant first generation)
2. Test SNAP candidates/produce active training (follows intended use)

Code

- VASP
- LAMMPS

→

Choose structure constraints

- Element types
- Number of atoms
- Composition

→

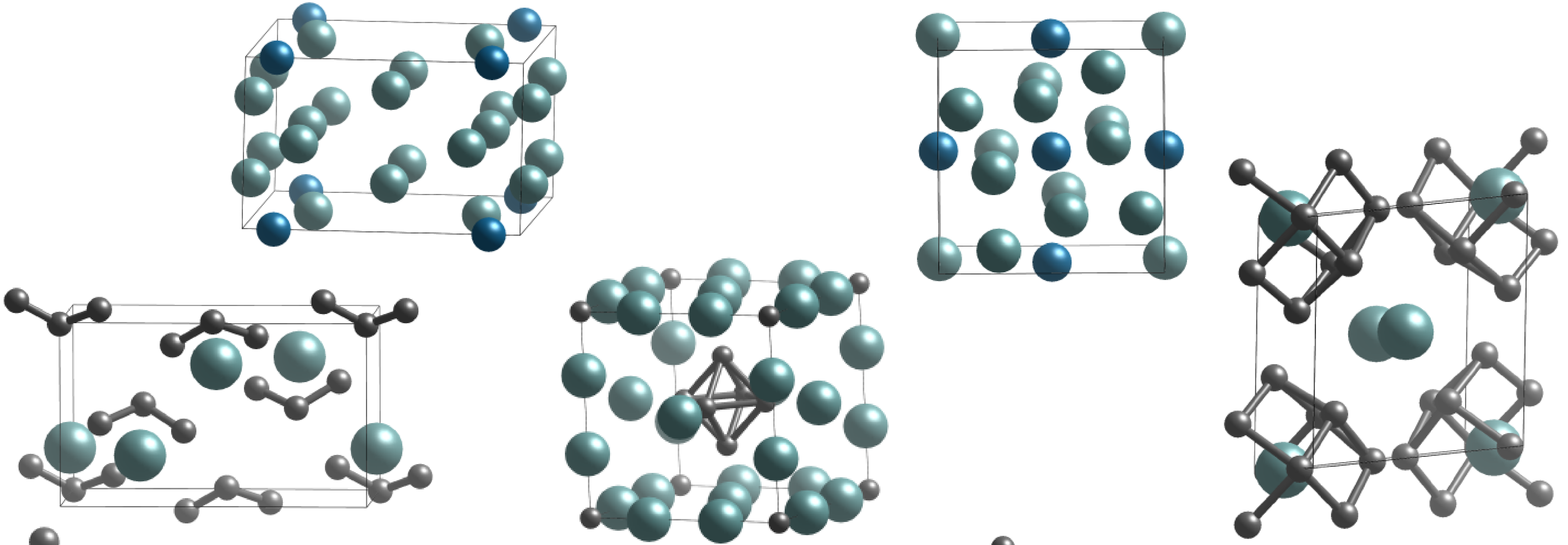
Genetic Algorithm

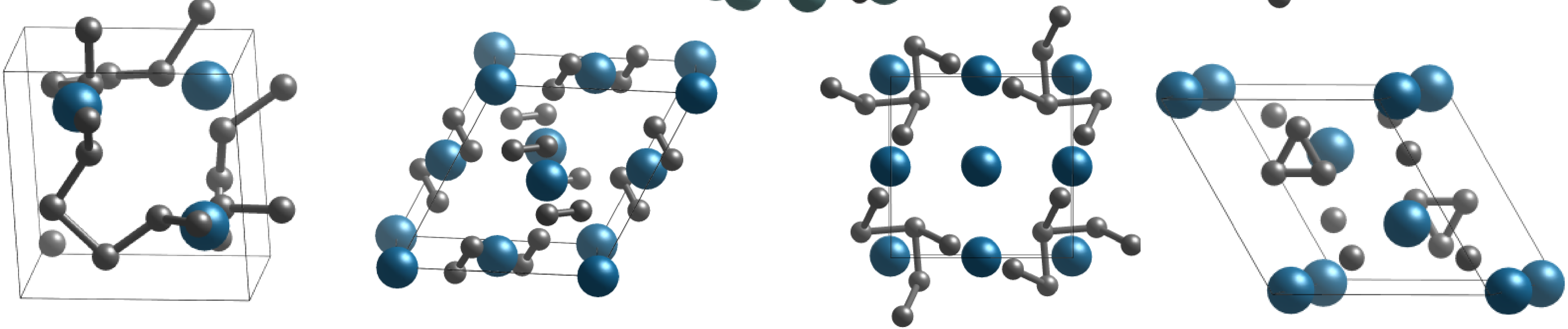
- 200 population size/1 generation
- 50 population size/~20 generations

https://uspex-team.org/en

USPEX (method 1) - composition sweeps for training (about 200 structures each)

- WZr
- ZrC
- WC

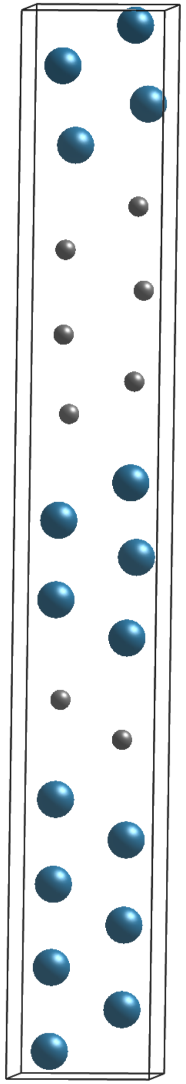




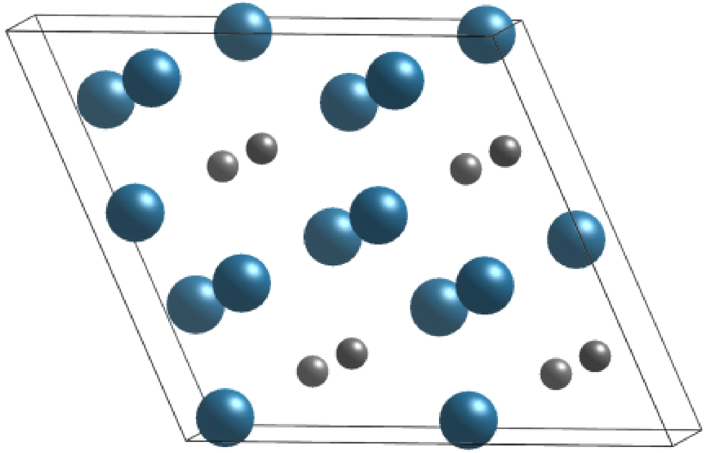
USPEX (method 2)– Testing SNAP candidates on W_2C

Most stable structure

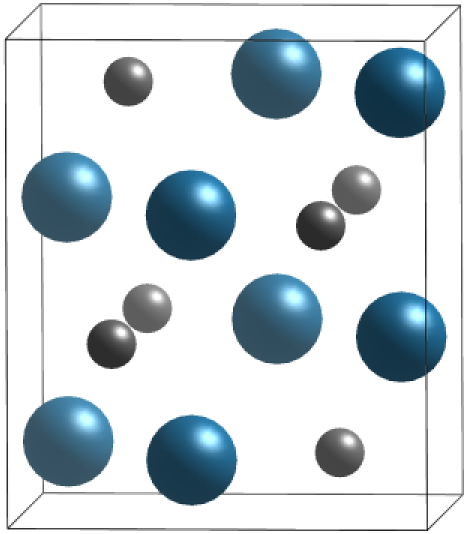
SNAP - **without** USPEX training



SNAP - **with** USPEX training



Materials Project



“Active” USPEX (method 2) – Using a SNAP candidate to produce training