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First-Principles and Classical Computational Study of W/ZrC Interfaces

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Abstract

Dispersion strengthened tungsten alloys (W/ZrC, W/TiC, etc.) are sought after as plasma facing materials (PFMs) due to their excellent mechanical properties, high thermal shock resistance, good high-temperature stability and excellent resistance to low-energy high-flux plasma irradiation [1,2].
The objective of our work is to develop and test a new machine learned interatomic potential for W-ZrC to use in molecular dynamics (MD) simulations. As a first step, first-principles calculations aiming at investigating the energetics and stability of the W/ZrC interfaces were performed. The collected data is used to both:

- create a database of atomic configurations on which the machine-learning interatomic potential is trained,
- probe the predictions of the obtained potential to evaluate its accuracy and range of validity.

Once developed and tested, this potential can be straightforwardly leveraged to perform accurate and large-scale MD simulations of the interplay between W and ZrC dispersoids (for example temperature and pressure influence on interface coherence and stability, or on mechanical properties) and eventually can be extended to include plasma species such as hydrogen.

[1] Xie, Z. M., *et al.* Scientific reports 5 (2015): 16014.

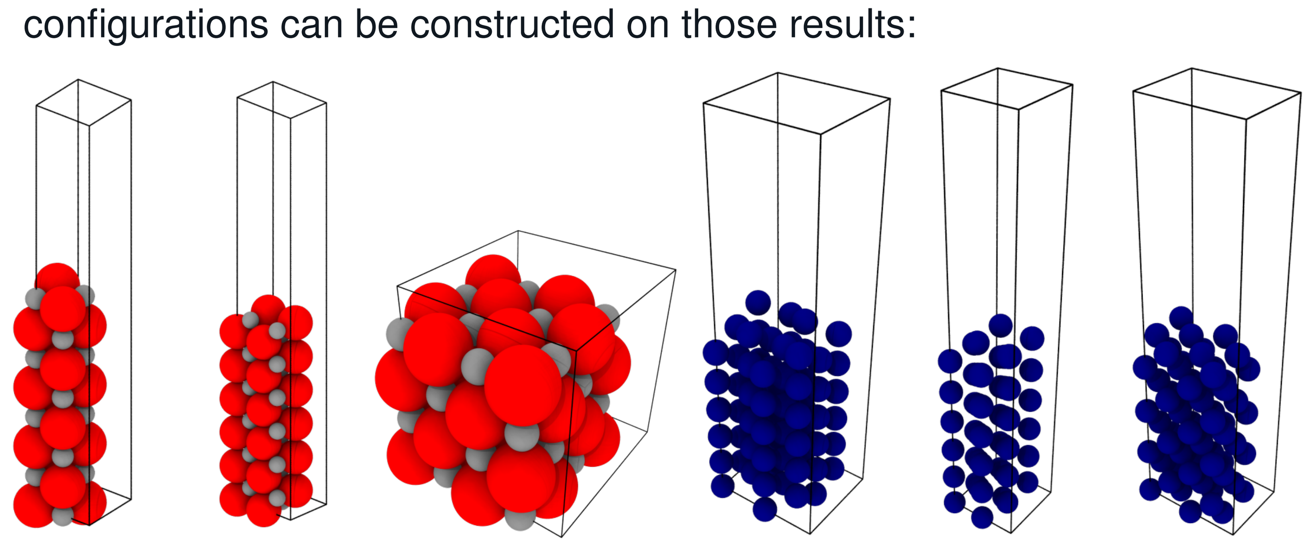
[2] Liu, R., *et al.* (2018). Nuclear Materials and Energy, 16, 191-206.

First-Principles Calculations

- We use Density-Functional Theory (DFT) as implemented in VASP [1].
- In all calculations the PBE [2] exchange-correlation functional was employed. The plane wave cutoff was set to 320 eV and the convergence in each self-consistency cycle was set to 10^{-8}.
- Bulk and surface calculations for W and ZrC are performed, we obtain the following results:

Property	Former work [5]	Our DFT
a_W (Å)	3.176	3.18
B_W (GPa)	310	322.2
Surf. W(100) (J/m^2)	3.93	4.0
Surf. W(110) (J/m^2)	3.19	3.26
Surf. W(111) (J/m^2)	4.23	4.32
a_{ZrC} (Å)	4.726	4.71
B_{ZrC} (GPa)	220	223.06
Surf. ZrC(200) (J/m^2)	1.65	1.67
Surf. ZrC(110) (J/m^2)	3.23	3.29

- Very good agreement with former studies is recovered. A database of atomic configurations can be constructed on those results:



ZrC - surf. (200) ZrC - surf. (110) ZrC - bulk W - surf. (100) W - surf. (110) W - surf. (111)

[3] G. Kresse and J. Hafner, Phys. Rev. B 47 , 558 (1993); ibid. 49 , 14 251 (1994).

[4] Perdew, John P., Kieron Burke, and Matthias Ernzerhof. Phys. Rev. Lett. 77.18 (1996); 3865.

[5] Zhang, Xu, *et al.* Applied Surface Science 499 (2020): 143995.

SNAP methodology [6]

- The approach first expresses the density of neighbor atoms surrounding a central atom i as a sum of δ-functions:

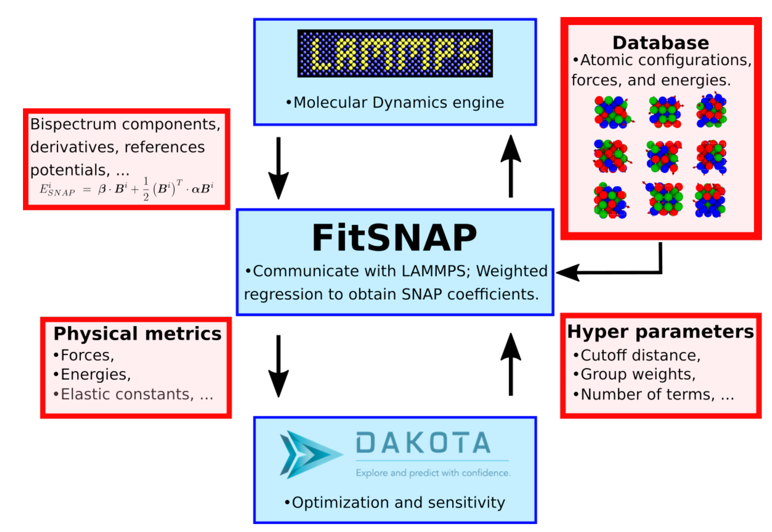
$$\rho(\boldsymbol{r}) = \delta(\boldsymbol{r}) + \sum_{r_{ij}<R_{cut}} f_c(r_{ij})\,\omega_i\,\delta(\boldsymbol{r}-\boldsymbol{r}_{ij}) \quad (1)$$

with ω_i weigths for the different atomic species, and $f_c(r_{ij})$ radial switching functions.

- This density can be expressed in terms of bispectrum components, from which the energy and forces on a given atom are expressed:

$$E^i_{SNAP} = \beta^i_0 + \boldsymbol{\beta}^i \cdot \boldsymbol{B}^i \quad (2)$$

$$\boldsymbol{F}^i_{SNAP} = -\boldsymbol{\beta}^i \cdot \sum_{i=1}^{N} \frac{\partial \boldsymbol{B}^i}{\partial \boldsymbol{r}_j} \quad (3)$$



- Out training procedure leverages the genetic algorithm of the Dakota package [7].

[6] Thompson, Aidan P., *et al.* Journal of Computational Physics 285 (2015): 316-330.

[7] Adams, Brian M., *et al.* Sandia National Laboratories, Tech. Rep. SAND2010-2183 (2009).

Training Results

- Two SNAP potentials are trained on the following groups of atomic configurations.
- Each potential corresponds to a different number of bispectrum coefficients: 31 coefficients / element (2jm6) and 56 coefficients / element (2jm8).

	# of Config.	# of Forces	Target Property	Energy MAE (eV/atom) 2jm6	Energy MAE (eV/atom) 2jm8
ZrC - EOS	40	960	Vol. Deform	0.0633	0.056
ZrC - DFT-MD	60	11520	Bulk phonons	0.041	0.014
ZrC - Defects	24	3840	Struct. Disorder	0.330	0.481
ZrC - Surfaces	8	576	Surf. Energetics	0.085	0.151
Zr - DFT-MD	100	5400	Pure Zr	0.0211	0.0237
C - DFT-MD	100	5400	Pure C	0.0036	0.0037
W - EOS	42	2016	Vol. Deform	0.0583	0.068
W - MD	60	21600	Bulk phonons	0.0162	0.052
W - defects	420	144000	Struct. Disorder	0.0268	0.0328
W - surface	180	6480	Surf. Energetics	0.0201	0.055

- Those potentials can be used to compute static properties for W and ZrC:

Property	DFT	SNAP (2jm6)	SNAP (2jm8)
a_W (Å)	3.18	3.18	3.18
B_W (GPa)	322.2	319.0	328.3
Surf. W(100) (J/m^2)	4.0	3.84	4.16
Surf. W(110) (J/m^2)	3.26	3.084	4.017
Surf. W(111) (J/m^2)	4.32	3.802	3.43
a_{ZrC} (Å)	4.71	4.69	4.68
B_{ZrC} (GPa)	223.06	221.4	222.6
Surf. ZrC(200) (J/m^2)	1.67	0.92	1.338
Surf. ZrC(110) (J/m^2)	3.29	0.7629	2.361

- Good agreement between the SNAP and DFT results is recovered for the bulk properties and the W surfaces, but ZrC surface agreements have to be improved.
- The potentials can be straightforwardly used to run large-scale MD calculations of W and ZrC surface and bulk systems.

Scaling Results on Summit

- Our objective is to perform very large scale calculations of W/ZrC interfaces.
- We leverage an ECP allocation on Summit to probe the performance of SNAP potentials leveraging LAMMPs and its KOKKOS implementation on the Summit supercomputer (GPU nodes).

EXASCALE COMPUTING PROJECT

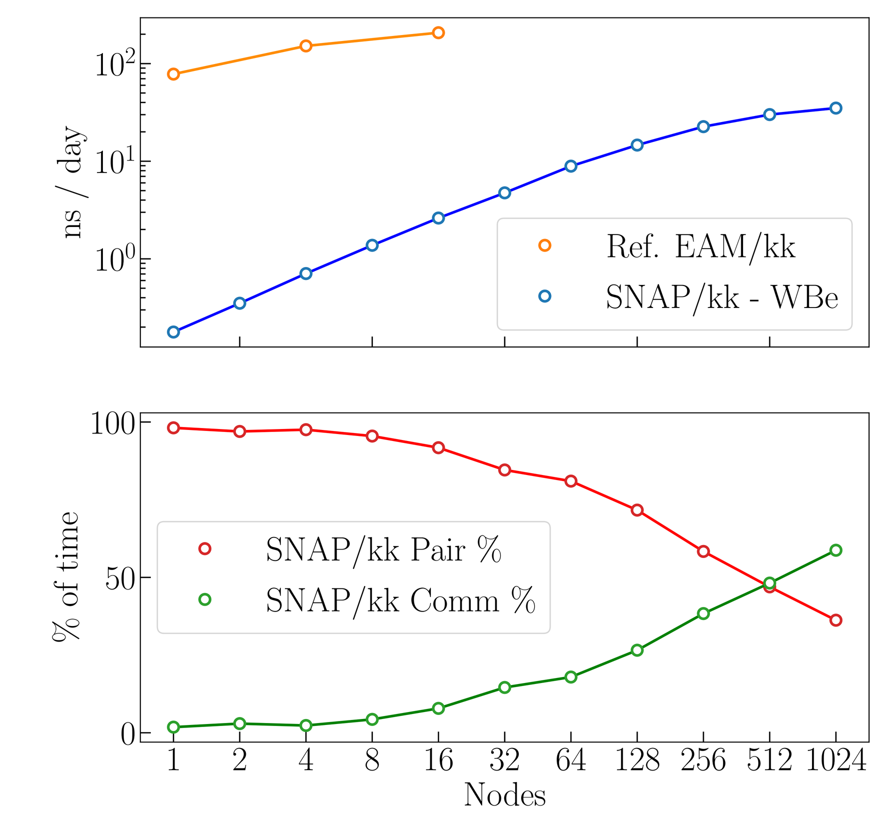
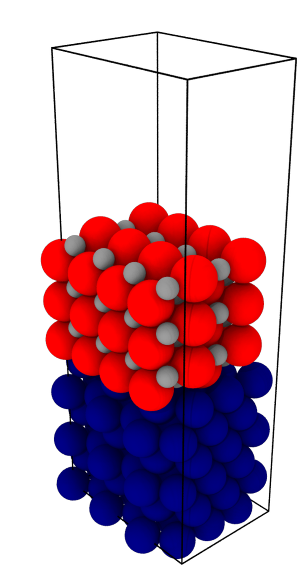


Fig. 4: Strong scaling results on Summit. On each node, we use 6 CPUs and 6 GPUs.

- We achieve excellent strong scaling up to 512 nodes, with a computing rate of 31 ns / day for a system of 4 million atoms.

Next step: Interface calculations

- W / ZrC interface cells have been generated.
- Our next step is to run the corresponding DFT calculations, and to compute the interface energies.
- Once correct surface energies are recovered, the corresponding configurations will be added to the training set, and new SNAP potentials will be trained.



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