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Development of Deep Learning Potentials for Ultra-high Temperature High-Entropy Driven Diborides

Gregory Vatrano
Missouri State University

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Gregory Vatrano
Dept. of Physics, Astronomy and Materials Science
Missouri State University
Advisor: Ridwan Sakidja

Abstract

In this study, we developed deep learning potentials for ultra-high temperature high entropy driven diborides of MB_2 where M is the transition metal made of Ti, Zr, Hf, Nb, Ta. The materials are being considered as the heat shield for hypersonic vehicles due to their high thermal conductivity and thermal stability. We used quantum mechanical data as our input for training and validation. Specifically, we use the energy, force and stress data from molecular dynamics simulations at elevated temperatures using quantum mechanics calculations. We used the DeePMD neural network code and varied the hyper-parameters such as cut-off radius, batch size, the number of hidden layers, maximum number of neighbors per atom site, etc. We showed the effect of these parameters on the accuracy of the potential models. Similarly, we showed the effect on the simulated stability of the crystal structure at 2000K. Overall, we have been able to develop a new potential capable of modeling the thermal stability of the diboride compounds.

Introduction

The implementation of artificial intelligence to model potentials is imperative due to the scaling of computational time for increasingly larger molecules based on the quantum mechanical calculations following the DFT (Density Functional Theory) approximations. This is captured in Figure (1), with AI (Artificial Intelligence) scaling linearly as opposed to DFT which is on a cubic scale. Modeling these larger molecules could aid in finding better compounds for heat-shielding in hypersonic vehicles. In this paper, we investigated the interatomic potentials for high-temperature, high-entropy-driven diborides of MB_2. M is the transition metal made of Titanium, Tantalum, Zirconium, Hafnium and Niobium in random concentrations intercalated by Boron-only layers. MB_2 was modeled with the software DeePMD [1] and by varying the hyperparameters, a new potential was developed.

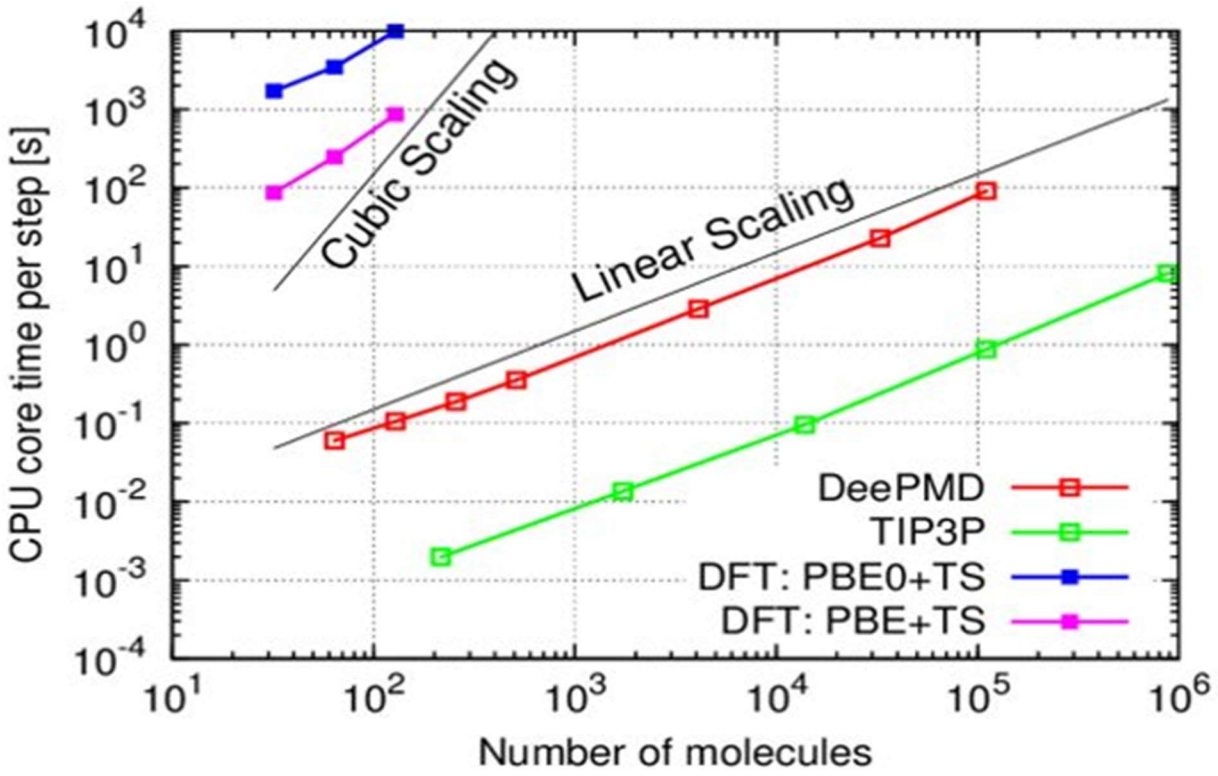

Figure 1 Computational time based on number of molecules showing linear scaling for DeePMD and cubic scaling for DFT [2].

Methods

We modeled MB_2 using three different models, model A, B, and C, varying the hyperparameters for each model. Model A has 72 neighbors for each atom except Boron with 92 neighbors. This assumption is because Boron is a smaller-sized atom. A cut-off radius of 6 Angstrom and a smooth cut-off radius of 5.8 Angstrom. Three hidden layers with 240 neurons each. A batch size of 1 was used. Model B also has 72 neighbors for each atom except Boron with 92 neighbors, a cut-off radius of 6 Angstrom and a smooth cut-off radius of 5.8 Angstrom. The hidden layers were increased to five layers with 240 neurons each. The batch size was increased to 50. Model C has 12 neighbors for each atom including Boron. A smaller cut-off radius of 2.2 Angstrom and a smooth cut-off radius of 1.5 Angstrom. The hidden layers were reduced to just one layer with 32 neurons. The batch size was reduced to 1. This is shown in Table (1) The learning rate decay stays the same for all three models; starting with 0.001 then ending up to 3.51e-8 with a decaying of 5000.

	Model A	Model B	Model C
Batch Size	1	50	1
Cut-Off Radius	6 (Angstrom)	6 (Angstrom)	2.2(Angstrom)
Smooth Cut-Off Radius	5.8 (Angstrom)	5.8 (Angstrom)	1.5 (Angstrom)
Hidden Layers	3	5	1
Neurons Per Layer	240	240	32
Neighbors	[72,72,72,72,72,92]	[72,72,72,72,72,92]	[12,12,12,12,12,12]

Table 1 Showing differences between hyperparameters of the three models.

The sample combines three types of information energy, force and stress, with the weight emphasized on the force at first and gradually changed to an almost equal weight for each. We used the quantum mechanical software VASP (Vienna Ab initio Software Package) [3,4,5] to perform the electronic structure calculations to get this data. The energy convergence is less than 10^-5 eV. The code was run at two GPU workstations at Missouri State; one with NVIDIA's RTX-6000 and another with NVIDIA V100. Each model ran for one million epochs. The DFT calculations were performed at Perlmutter Supercomputer at NERSC in the Lawrence Berkeley National Laboratory (LBNL).

Results

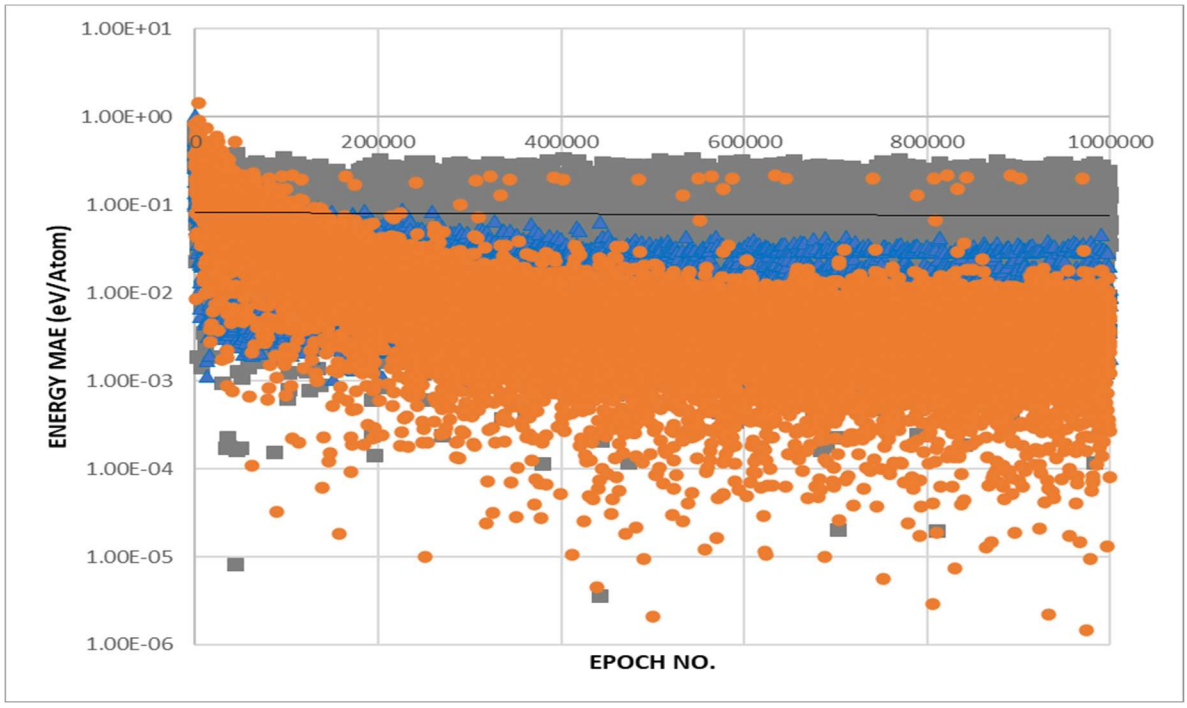

Figure 2 Comparison between the MAE for energy (in eV/Angstrom) for three models: Model A in blue, Model B in orange, Model C in grey.

Figure (2) above shows the comparison in terms of the relative error in energy between the results from quantum mechanical calculations as the ground truth versus those from DeePMD Learning Potentials. Model C gives the worst results, while Model B gives the best results, Model A is in between. This is quite consistent with our expectation from the hyperparameters listed in Table (1). Model C is the worst because it has the smallest cut-off radius, which may have resulted in parts of the nearest neighbors not being included within some of the clusters considered for the neural network model, the shallow hidden layer with just 32 neurons would also influence the results. Model B has the least average error because of several reasons; 1) it has a larger cut-off radius, 2) it has a larger batch size and 3) it has more hidden layers, each with more neurons. This combination resulted in the best model with MAE for energy reaching below 1x10-4.

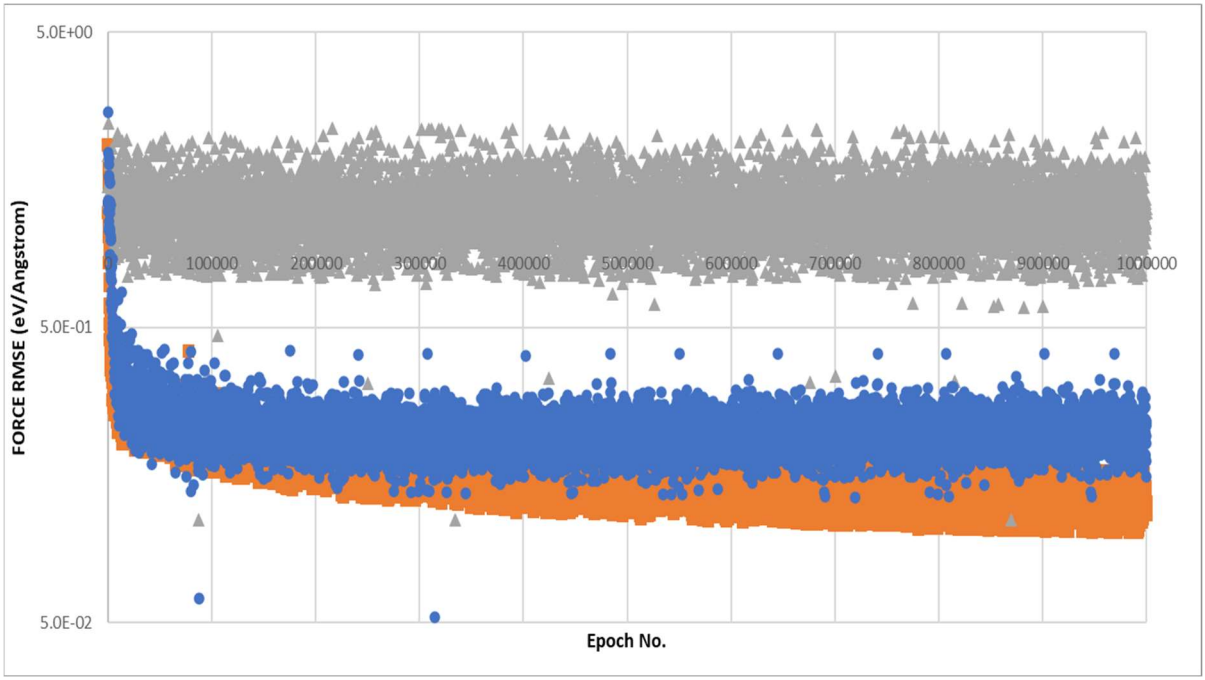

Figure 3 Comparison between the RMSE for force (in eV/Angstrom) for three models: Model A in blue, Model B in orange, Model C in grey.

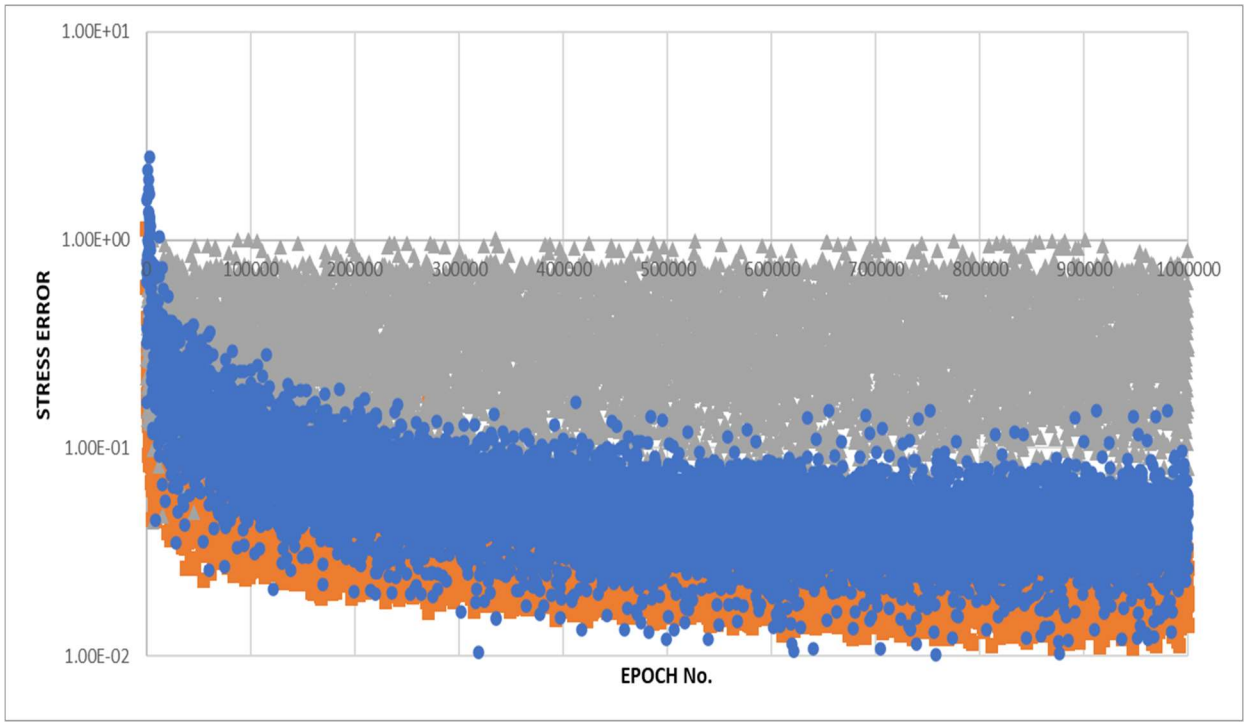

Figure 4 Comparison between the RMSE for stress for three models: Model A in blue, Model B in orange, Model C in grey.

Figure (3) and Figure (4) show the error results from force and stress respectively. They both also confirm the same trend that we saw in the error of the energy, namely Model C gave the worst results while Model B supplied the best results. This is not surprising. The more correct the energy predictions are, the better the predictions for force or stress are.

Analysis

We tested the AI models with Molecular Dynamics simulations for a hypothetical MB_2 crystal structure made of a randomly distributed transition metals intercalated by Boron-only layers as shown by Figure (5) below:

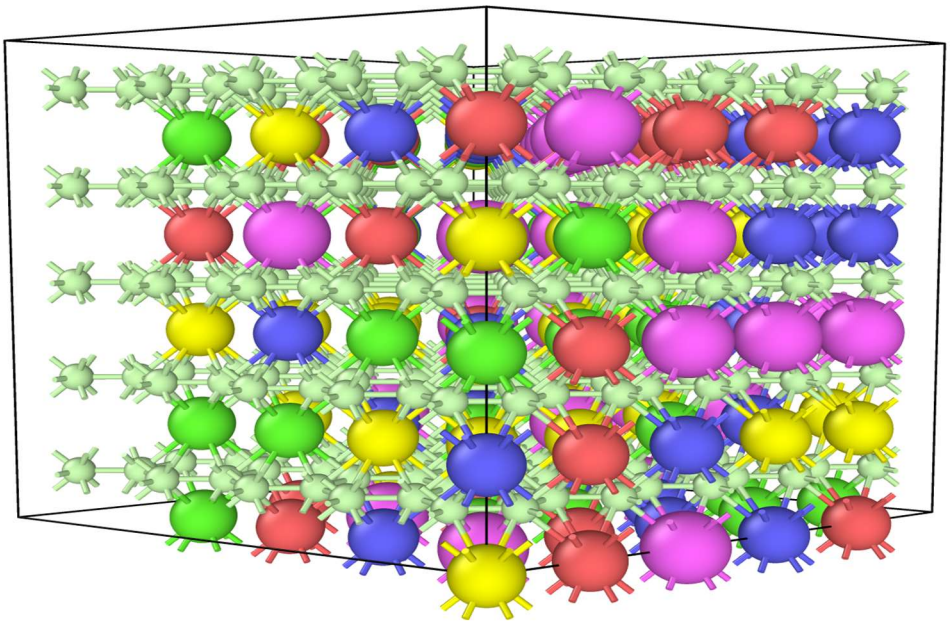

Figure 5 Crystal structure of MB_2 (M= Hf, Ti, Zr, Ta, Nb) B_2 at its ideal atomic position.

As shown in Figure (6) below, the crystal structure stays relatively stable when we tested at 2000K under atmospheric pressure using Model B. Certainly, because of the elevated temperature, the atomic positions are no longer exactly at their original positions, rather they move around the original positions due to the thermal fluctuations. The high thermal stability of this compound is consistent with the experimental results of the research group from UC San Diego [6]. In their experiments, the MB_2 with the same composition, is stable at least up to 2000°C (2273K). We only tested ours up to 2000 K, but this does prove the high thermal stability of the diboride phase using our Model B. Model A also showed similar thermal stability.

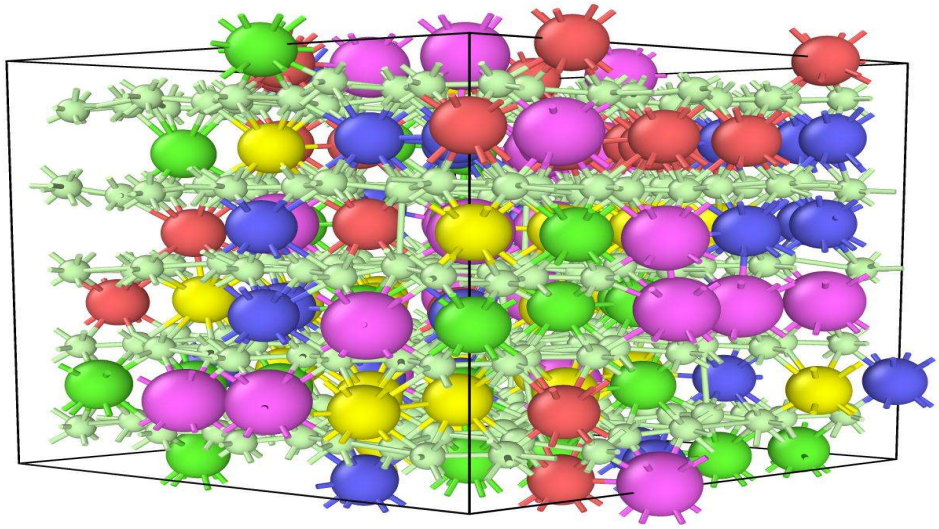

Figure 6 The same MB_2 crystal structure heated to 2000K under atmospheric conditions, simulated using Model B.

In contrast, when we tested the potential Model C, the structure became unstable as shown by Figure (7) below. This was because the forcefield and thus the force balance between atoms within this model are not correct and the overshot of the force balance causes the structure to become disordered and catastrophic results occurred.

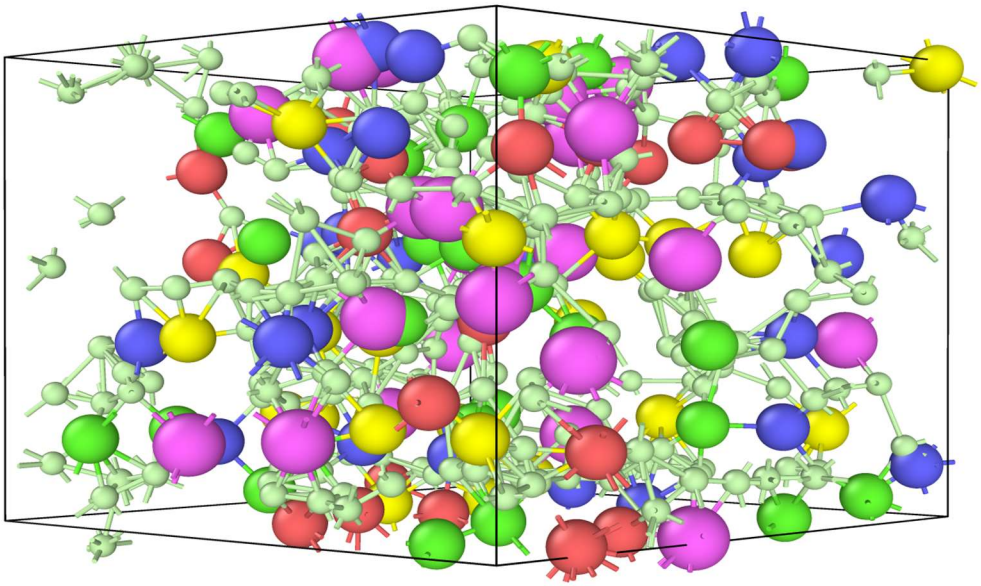

Figure 7 The same MB_2 crystal structure heated to 2000K under atmospheric conditions, simulated using Model C.

Conclusion

In this study, we have developed the Deep Learning potentials that can be used to simulate relatively complex diboride compounds. By using the correct hyperparameters, we can construct the correct model that would produce a relatively low error level in terms of energy, force and stress. We have proven the thermal stability of the diboride phase consistent with the experimental results from the literature.

Acknowledgements

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Biography

My name is Gregory Vatrano, I am from Las Vegas, Nevada. I am a senior at Missouri State University in the Physics, Astronomy and Materials Science Department. I am a physics major interested in artificial intelligence and nuclear physics.

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