# Machine-learning force fields for hydrogen and vacancy complex in tungsten

Yuki NOGUCHI[1] (noguchi.yuki@nifs.ac.jp), Daiji KATO[2,1]
1. Kyushu Univ. 2. National Institute for Fusion Science

## Abstract

- To investigate hydrogen-vacancy (H-V) interactions in tungsten (W), we constructed W-H-V Machine Learning Force Fields (**MLFF**) based on SOAP descriptors and Bayesian linear regression (on-the-fly ML method).
- The present MLFF can predict formation energies and hydrogen binding energies calculated by DFT with fair accuracy.
- The present studies showed that hydrogen trapping stabilizes vacancy clustering.

## Background

- Tungsten is a primary candidate as a plasma-facing material in the divertor (Fig. 1).
- However, the neutron irradiation experiment showed that the formation of defect structures in tungsten increased deuterium (D) retention (Fig. 2).
- H–V interactions in tungsten have been investigated through molecular simulations, including density functional theory (DFT) and molecular dynamics (MD).

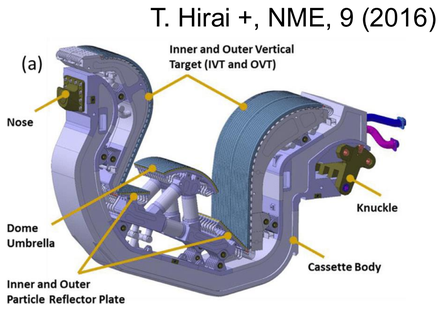


Fig. 1: ITER divertor

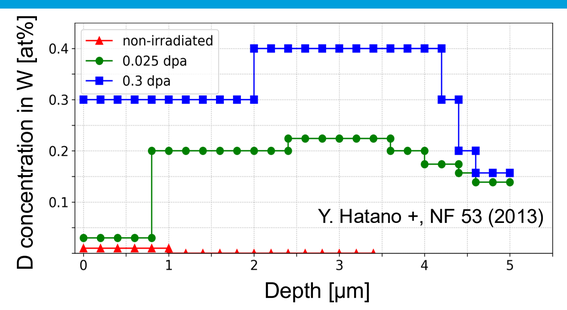


Fig. 2: Depth profile of D in non-irradiated and n-irradiated W specimens after exposure to D plasma at 773K

## Machine Learning Methods – on-the-fly machine learning *R. Jinnouchi +, PRB (2019)

- **MLFF**, a non-parametric approach, has recently emerged as a promising alternative to empirical potentials.
- On-the-fly machine learning* efficiently samples DFT data of various atomic configurations and modifies MLFFs sequentially using **Bayesian linear regression with kernel functions represented by SOAP descriptors**. The DFT calculation is performed only for atomic configurations with large uncertainty in the Bayesian inference.
- The on-the-fly MLFF is implemented in the first-principles code VASP. We have constructed an MLFF from 1291 W-H-V structures.

## Result

- The present DFT-PBE calculations are consistent with previously reported DFT results (Table 1).
- The present MLFF can predict DFT formation energies of $V_n$ and $V_nH$ complexes within an average accuracy of 0.5 eV (Fig. 3).
- Both MLFF and DFT calculations indicate that trapping a single hydrogen atom at a vacancy reduces its formation energy (Fig. 3).
- In both di-vacancy and tri-vacancy structures, **the O-site at the center** was found to be more favorable for hydrogen trapping than those at the periphery.

Table 1: Vacancy formation energies and binding energies of a single H atom to vacancies by DFT-PBE

| configuration | Formation energy | | H binding energy | |
|---|---|---|---|---|
| $V_1$ | **3.166** | 3.17[1] 3.168[2] | **1.231** | 1.225[2] |
| $V_2$ 1NN | **6.470** | 6.50[1] 6.451[2] | **1.448** | 1.80[1] 1.441[2] |
| 2NN | **6.778** | 6.81[1] 6.816[2] | **1.862** | 2.15[1] |
| 3NN | **6.458** | 6.47[1] 6.445[2] | **1.309** | |
| $V_3$ 1NN+1NN-s | **9.719** | 10.71[3] | **1.450** | |
| 1NN+1NN-$b_1$ | **9.638** | 10.58[3] | **1.534** | 1.44[3] |
| 1NN+1NN-$b_2$ | **9.816** | | **1.725** | |
| 1NN+2NN | **9.979** | 10.96[3] | **1.836** | 1.83[3] |
| 2NN+2NN-s | **10.407** | 11.36[3] | **1.834** | |
| 2NN+2NN-b | **10.205** | 11.16[3] | **1.862** | |

[1]D. Kato +, JNM, 417 (2011) [2]K. Ohsawa +, JNM, 527 (2019) [3]D. Kato (**AM05**), MoD-PMI 2017

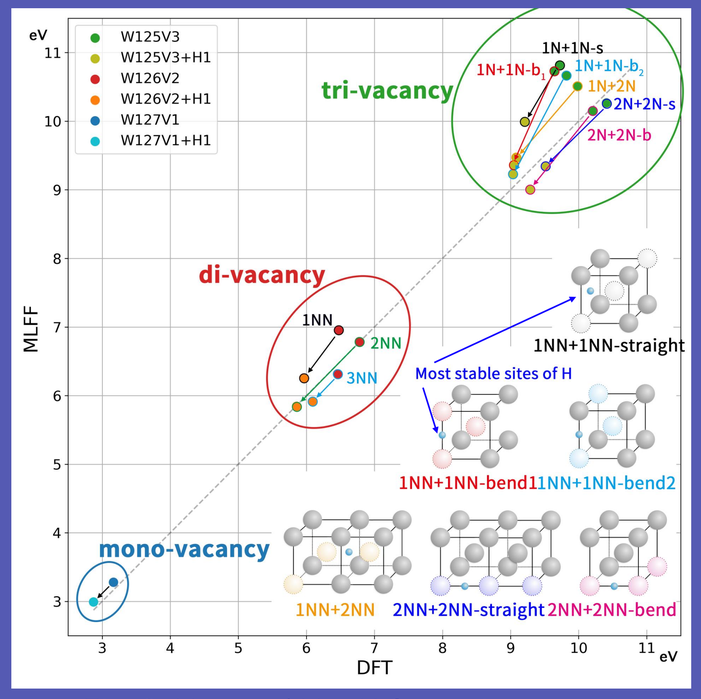


Fig. 3: $V_{1\sim3}$ and $V_{1\sim3}H$ formation energies

- DFT calculations revealed that the trapping of a single hydrogen atom altered the most stable configuration of vacancy clusters. This has an important implication in <u>modeling vacancy agglomeration interacting with ambient hydrogen atoms</u>.
- The present MLFF provides more accurate predictions for the binding energies of multiple hydrogen atoms at mono-vacancies than available empirical potentials (Fig. 4 (a)).
- The 1NN di-vacancy case also showed similar consistency with DFT (Fig. 4(b)).

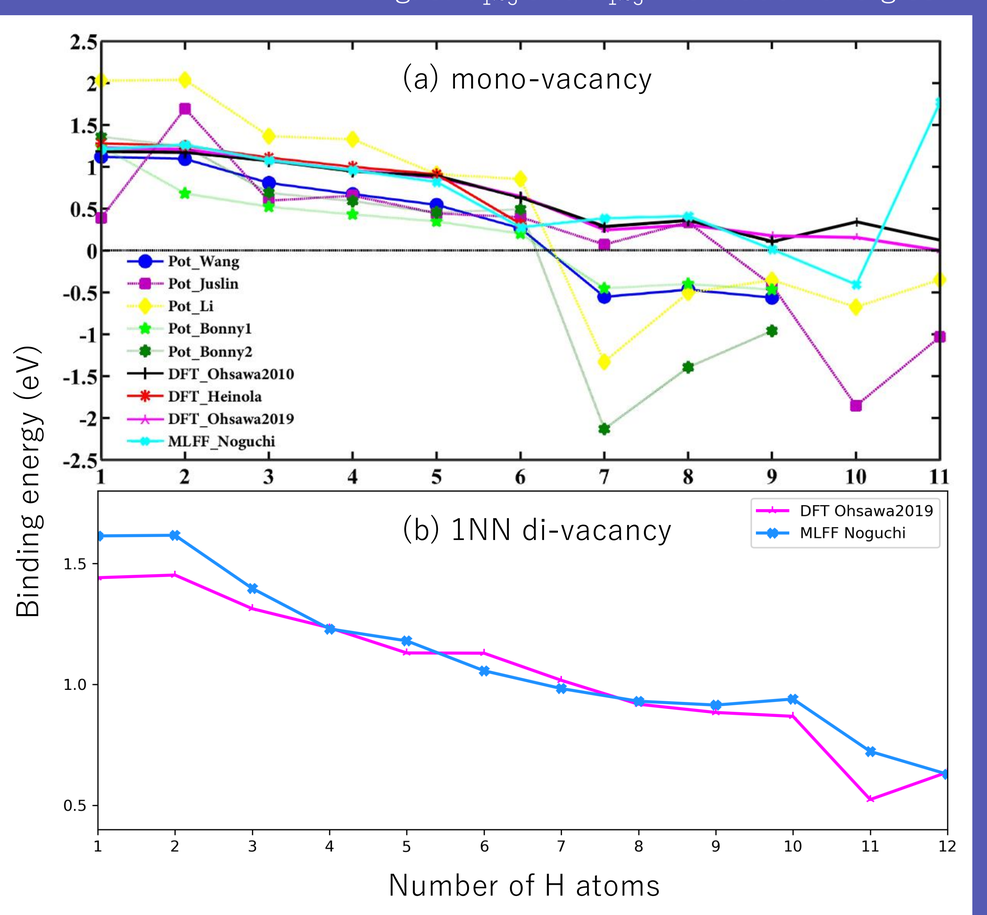


Fig. 4: Binding energies of H atoms to vacancies

## Conclusion & Future work

- For tungsten containing atomic vacancies and hydrogen atoms, we constructed the MLFF that can reproduce DFT results fairly accurately.
- The present studies showed that hydrogen trapping stabilizes vacancies. Further insights into the effects of hydrogen in vacancies can be gained through density of states (DOS) analysis.
- The MLFF will be further optimized to search for stable structures of larger clusters and to perform molecular dynamics simulations at finite temperatures.

## Acknowledgements

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