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# Influence of helium diffusion and aggregation on the variety of metal nanostructures under helium plasma irradiation

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## Background

- The formation of fiber-like nanostructures on tungsten(W) and molybdenum(Mo) are observed after helium(He) plasma exposure.
- Their formation mechanism has not been understood well.
- The He bubble seems to have a influence on these nanostructures.

## Purpose

1. To investigate microscopic relationship between metal and He
   - ➢ Calculate variations of He binding energies using first principle calculation based on density functional theory (DFT)
2. To expand macroscopic simulation from DFT calculation result
   - ➢ Developed He diffusion / aggregation simulation code based on kinetic Monte Carlo method (KMC)
   - ➢ Investigate differences in formation of bubbles for each metal by KMC simulation

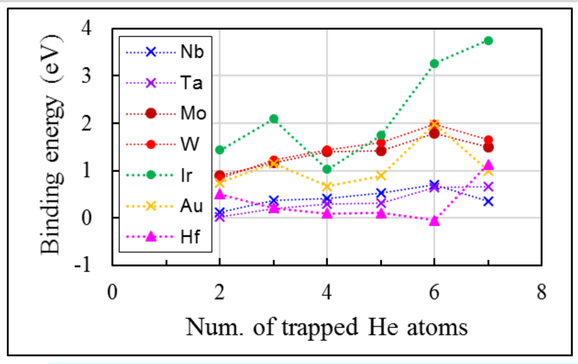
※In this study, KMC simulation is performed on interstitial sites of bcc metal, which does not consider vacancy.

## Results of DFT calculation

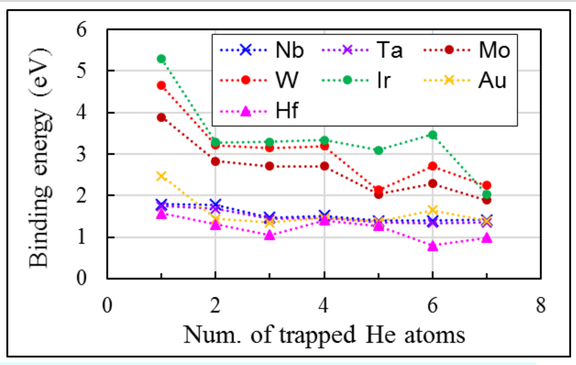
### Definition of He bind energy

$$E_{int}^{bind}(n) = -E_{\mathrm{tot}}(\mathrm{Me}_N\mathrm{He}_n) - E_{\mathrm{tot}}(\mathrm{Me}_N) + E_{\mathrm{tot}}(\mathrm{Me}_N\mathrm{He}_{n-1}) + E_{\mathrm{tot}}(\mathrm{Me}_N\mathrm{He}_1)$$
$$E_{vac}^{bind}(n) = -E_{\mathrm{tot}}(\mathrm{Me}_{N-1}\mathrm{He}_n) - E_{\mathrm{tot}}(\mathrm{Me}_N) + E_{\mathrm{tot}}(\mathrm{Me}_{N-1}\mathrm{He}_{n-1}) + E_{\mathrm{tot}}(\mathrm{Me}_N\mathrm{He}_1)$$

**The binding energies at interstitial sites**




**The binding energies at a monovacancy**



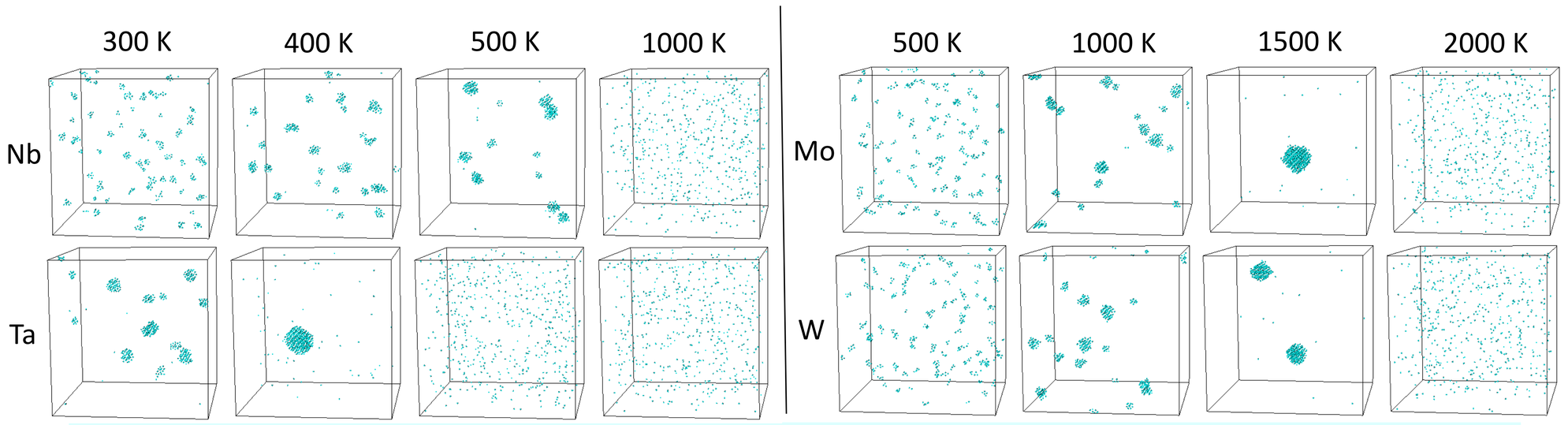

- A metal with monovacancies has a higher binding energy
- A fiber-like nanostructure formable metal has a higher binding energy
- The peak position of the graph depends on the crystal structure

## He bubble formation at each temperature in BCC metal's calculated by KMC simulation

Simulation condition

- Box size: $30 \times 30 \times 30$ unit cells
  - ➢ Periodic boundary
- He/W : 0.01
  - ➢ The number of helium does not change over time
- Elapsed time : 1μs
- The initial position of helium is randomly arranged at the T-site of each metal
- Diffusion coefficient:
  - ➢ $3.36 \times 10^{-8}\exp(\Delta E_0/k_BT)$

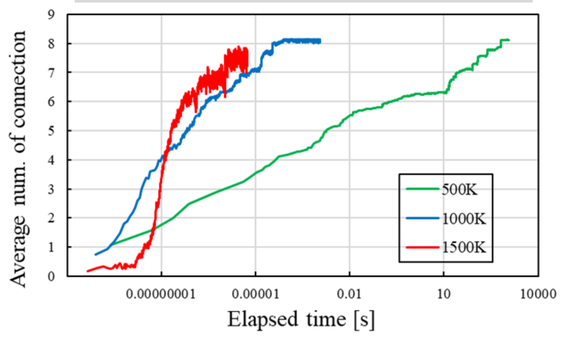
| Metal | $\Delta E_0$ [eV] |
|---|---|
| W | 0.0623 |
| Mo | 0.0572 |
| Ta | 0.0847 |
| Nb | 0.0996 |



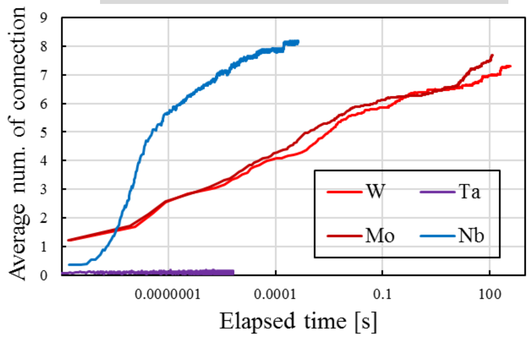

- Appropriate temperature, at which bubbles tend to be large, varies from metal to metal (Agrees with experimental study)
- At low temperatures, since helium bonds are hard to break, it takes time to grow into large bubbles if small clusters are massive.
- At high temperature, the diffusion of helium will be fast and the bond will tear off easily, so the bubble will not grow
- It seems that the difference of bubble formation for each metal is greatly affected by the DFT calculation result

## Average number of connection per elapsed time

**Temperature dependence (W)**



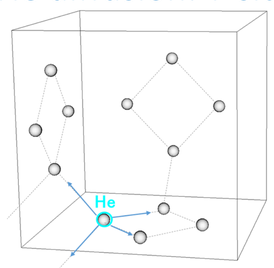

**Metal dependence (500 K)**




## He connection model in KMC

- Single He atom in BCC metal passes through the migration path on the right figure and is trapped at the T-site
- In DFT calculation, He tends to be trapped more easily at the T-site than the O-site when aggregated between the lattices and He is likely to be trapped in the vicinity of the second nearest T-site when viewed from the other He

He diffusion: T-site

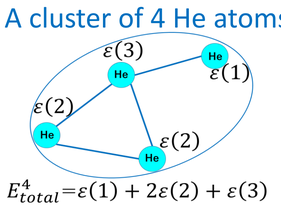



- ➢ In this KMC model, He jumps only between T-sites and is regarded as connecting when He is adjacent at the second nearest T-site.

### How to obtain $\Delta E_{connect}^{k}$ from DFT calculation result

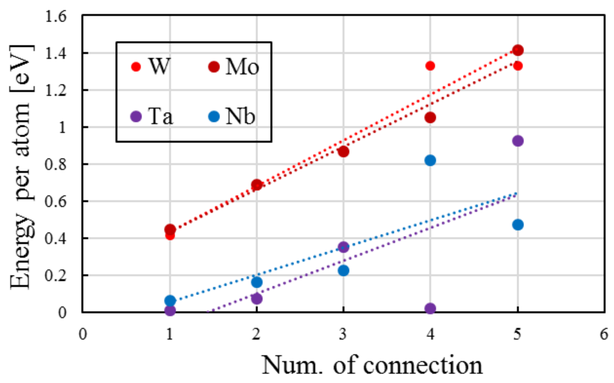
- We want to express $\Delta E_{connect}^{k}$ as a function of a certain number $k$ of connections with another He.
- If there is cluster of $n$ helium, the total energy that the cluster has $E_{total}^{n}$ is
  $E_{total}^{n} = \sum_{i=1}^{n}\varepsilon(k_i)$
  $E_{int}^{bind}(n) = E_{total}^{n} - E_{total}^{n-1}$
- Fitting $\varepsilon(k_i)$ in the form of $ak + b$

A cluster of 4 He atoms




$E_{total}^{4} = \varepsilon(1) + 2\varepsilon(2) + \varepsilon(3)$

| Metal | $a$ [eV/atom] | $b$ [eV] |
|---|---|---|
| W | 0.2471 | 0.1853 |
| Mo | 0.2289 | 0.2067 |
| Ta | 0.0687 | -0.2532 |
| Nb | 0.1217 | -0.0936 |

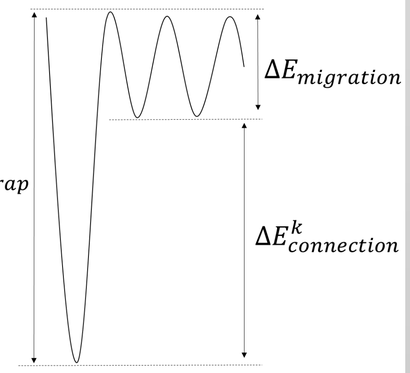



## KMC calculation method

- What is KMC:
  - ➢ A method to make statistical behavior by repeating the particle jumping randomly between lattices with certain event
- What is KMC's event in this research:
  - ➢ One helium at a certain T-site moves to some neighboring T-site
- The probability $P_i$ that an event $i$ occurs
  $P_i = P_0\exp(-\frac{\Delta E}{k_BT})\ [\mathrm{s}^{-1}]$
  ※$P_0 = 6D(\mathrm{T})/nl^2\exp(-\Delta E_0/k_BT)\ [\mathrm{s}^{-1}]$

| | |
|---|---|
| $\Delta E$ | Barrier energy [eV] |
| $P_0$ | Prefactor |
| $D(\mathrm{T})$ | Diffusion coefficient |
| $n$ | Number of migration path for each T-site |
| $l$ | Length of the migration path [m] |
| $\Delta E_0$ | Activation energy [eV] |

- A migration barrier energy $\Delta E_{migration}$ that a certain He does not connect to other He
  $\Delta E_{migration} = \Delta E_0$
- A detrap barrier energy $\Delta E_{detrap}$ that a certain He is connected to $k$ other He
  $\Delta E_{detrap} = \Delta E_{migration} + \Delta E_{connect}^{k}$
  - ➢ $\Delta E_0$ and $\Delta E_{connect}^{k}$ are obtained by DFT




## Summary

- By comparison between DFT calculation and experimental results, fiber-like nanostructures tend to be formed with metals with a higher binding energy.
- In order to investigate the relationship between the DFT calculation results and the He bubble formation tendency, we developed He aggregation / diffusion simulation code based on KMC.
- Temperature and material dependence of He bubble formation was observed by KMC simulation and there is an appropriate temperature range for the bubble grow.
- Large bubbles are formed at temperatures as high as 1000 K or more for W and Mo with higher binding energy obtained by DFT