Evaluation of Tritium Inventory in Irradiated Tungsten by Atomic-Scale Modeling

Takuji Oda, Hyung Jin Shim, Gon-Ho Kim

Department of Nuclear Engineering,
Seoul National University

IACA-CRM (F43021): Plasma-Wall Interaction with Irradiated Tungsten and Tungsten Alloys in Fusion Devices

About our team

<Implementing Institution>
Department of Nuclear Engineering, Seoul National University (SNU)

<Chief Scientific Investigator (CSI) > Takuji Oda
- ✓ Computer simulations (first-principles with DFT, MD, kMC)
- ✓ Experiments with vibrational spectroscopy(FT-IR, Raman), TDS, XPS/UPS

<Secondary CSI> Hyung Jin Shim
- ✓ Development of a Monte Carlo particle transport analysis code (McCARD)
- ✓ Neutronics design and analysis

<Main additional Scientific Staff> Gon-Ho Kim
- ✓ Plasma processing
- ✓ Material morphology change induced by thermal and particle energy

+ 2~3 graduate students and 1 postdoc as of now.

Available facility

(1) IFERC-CSC supercomputer:
we are involved in a project related to this CRP proposal in the current cycle (2013.11-2014.11)
[PI: Dr. Watanabe at JAEA].

(2) Proton accelerator (Kyungju):
100MeV (average beam current 1.6mA, beam repetition 60Hz, and max beam power 160kW) and 20MeV (average beam current 4.8mA, beam repetition 120Hz, and max beam power 96 kW) proton accelerators.

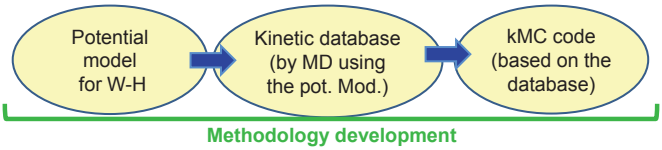
(3) SNU-Edge Plasma-Wall Interaction Simulator
H and D irradiation with ~10^{21} m^{-2} s^{-1} at 100 eV; TDS; sample heating up to 2000 K.

(4) Ion accelerator at Univ. Tokyo
up to 1.7 MV as acceleration voltage of Tandetron; H, He, Fe, W,… are available with nA~μA flux

Research objectives

Our research objective is
(i) to establish a computational methodology to simulate the tritium behavior in irradiated tungsten which contains radiation defects, then
(ii) to acquire estimates of the tritium inventory buildup in plasma-facing tungsten under some proposed fusion reactor conditions.

To achieve this overall goal, we work on 3 subjects for 3 outcomes:
(1) a high-quality potential model for W-H system to simulate correlated behaviors of tritium and defects in W, which is attained in the 1st year;
(2) a kinetics database relevant to the tritium behavior in irradiated tungsten, in the 2nd year;
(3) estimates of the tritium retention (code development) in plasma-facing W under some proposed fusion reactor conditions, in the 3rd year.



Expected outcome for 3 years

To achieve this overall goal, we work on 3 subjects for 3 outcomes:
(1) a high-quality potential model for W-H system to simulate correlated behaviors of tritium and defects in W, which is attained in the 1st year;
(2) a kinetics database relevant to the tritium behavior in irradiated tungsten, in the 2nd year;
(3) estimates of the tritium retention (and possibly a code to evaluate it) in plasma-facing tungsten under some proposed fusion reactor conditions, in the 3rd year.

<1st year> Construction of a quality potential model
- ✓ Potential model
- ✓ DFT data (energies/ force/ stresses)

<2nd year> Construction of a kinetic database
- ✓ Kinetic database (of MD/MS simulations)
  - ➢ V_n-H_m interaction
  - ➢ I_n-H_m interaction
  - ➢ H migration
  - ➢ I migration
  - ➢ V migration

<3nd year> Tritium retention estimates using kMC
- ✓ A (kMC) code for tritium retention evaluation
- ✓ Retention estimates for some fusion reactor conditions

Research objectives

At this moment, our model may ignore
- ✓ He
- ✓ grain boundaries
- ✓ dislocations
- ✓ impurities
- ✓ other elements (by transmutation, as alloy element, etc)

We do not have (have not had) strong existing research projects in this field. So, we are happy to work on something to fill uncovered gaps among what other CRP participants cover.

<What I would like to have in this meeting>
(i) To obtain comments to make our plan better
(ii) To clarify the fields/plans where we can collaborate with other CRP participants, and
(iii) To find some "gap" which should be studied soon and we may make contribution to.

Contents

(1) Construction of a high-quality potential model for W-H system

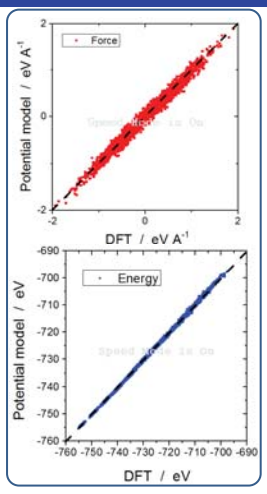
(2) Construction of a kinetics database relevant to the tritium behavior in irradiated tungsten

(3) Construction of a kMC code to estimate the tritium in plasma-facing tungsten

(1) Potential model development

- ✓ We developed a method to effectively construct a potential model to reproduce energies/ forces/ stresses determined by *ab-initio* MD.
  - ➢ The right figure is an example with MgO (Coulomb + 2 body). The *ab-initio* MD data were accumulated in 300-4500 K: 150000 (N) trajectories for energies, M×6 for stresses, and N×128 ×3 for forces.
  - ➢ We are now extending a method to an embedded atom model.

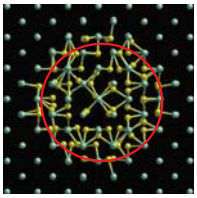
<Fitting target for W-H system>
- ✓ Perfect crystal: W_{128} over 300-5700 K
- ✓ Surfaces: <100>, <110>, <111>, <211> over 300-1500 K
- ✓ Vacancies: V_1 (W_{127}), V_2 (W_{126}), V_3 (W_{125}), V_5 (W_{123}), V_9 (W_{119}) , over 300-4500 K
- ✓ Interstitials: *TBD*
- ✓ H atoms in W: *TBD*
- ✓ H_2: *TBD*

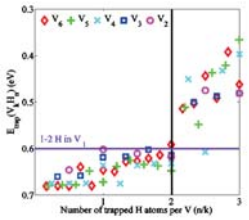


(2) Kinetics database

- ✓ MD using LAMMPS code with the potential model to be developed
- ✓ We evaluate the following quantities of of V_n-T_m (and I_n-T_m) cluster, in which n vacancies and m tritium atoms are involved as a function of n, m and defect configurations.
  - ➢ binding energy
  - ➢ decomposition energy
  - ➢ migration energy
- ✓ We will also estimate the degree of possible errors/uncertainties by comparison with interaction energies obtained using first-principles calculation for some typical V_n-T_m and I_n-T_m clusters.

V51-H80



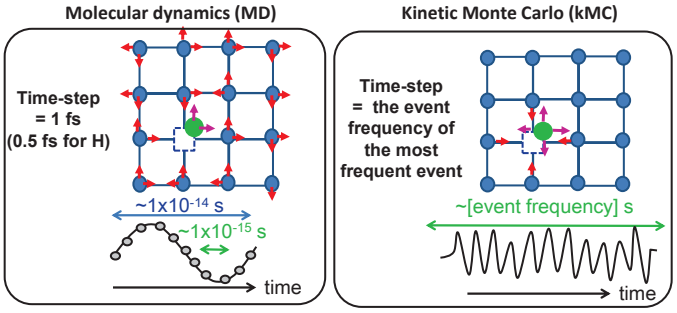


(3) kMC code development

To perform a kMC simulation, we need to prepare the following data:

- ✓ The list of all key processes/events (migration, trapping, detrapping, recombination, desorption, etc) involved in a concerned phenomenon
- ✓ The attempt frequency for each event (ν)
- ✓ The activation barrier for each event (E_a)

$$[mean\ event\ frequenc] = \nu \times exp(-E_a/RT)$$



(3) kMC code development

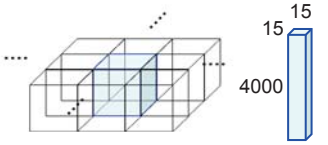


Figure: 2D periodic boundary conditions (width of a layer: 0.317 nm)

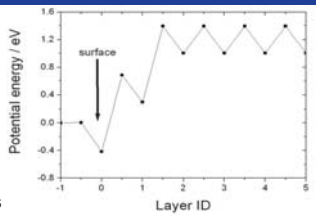


Figure: Potential curve near the surface

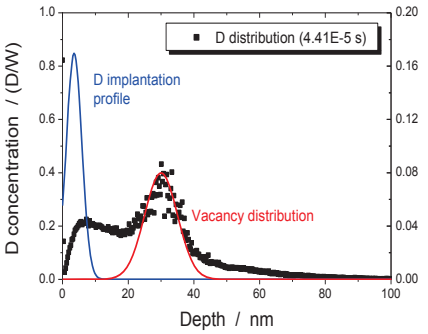
(1) D implantation event
(2) Diffusion events: migration as D atom in bulk & on surfaces
- ✓ including trapping/detrapping by vacancies
- ✓ diffusivity is adjusted and is equal with the value obtained by *R. Frauenfelder, J. Vac. Sci. Technol.* ***6****, 388 (1969).*

(3) Desorption events: recombination desorption as D_2

- ➢Crystalline structure modeled was simple cubic lattice (not bcc) for simplicity.
- ➢Deuterium flux:1×10^{26} D m^{-2} s^{-1} for feasibility; energy: 100 eV.
- ➢Temperature: 473 K

*T. Oda et al. (TITAN), Fusion Sci. Technol. 60 (2011) 1455.

(3) kMC code development



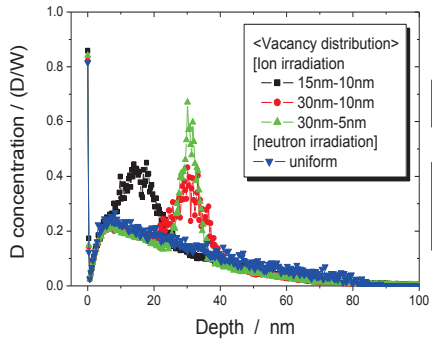
<Defect loading>
➢Ion irradiation

<Deuterium loading>
➢Fulx:$1x10^{26}$ m^{-2} s^{-1}
➢Energy: 200 eV
➢Fluence:$4.4x10^{21}$ m^{-2}

Figure: Correlation between defect and H depth profiles

*T. Oda et al. (TITAN), Fusion Sci. Technol. 60 (2011) 1455.

(3) kMC code development



<Defect loading>
➢Ion or neutron irrad.

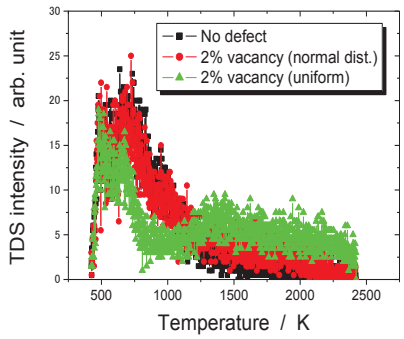
<Deuterium loading>
➢Fulx:$1x10^{26}$ m^{-2} s^{-1}
➢Energy: 200 eV
➢Fluence:$4.4x10^{21}$ m^{-2}

Figure: Comparison in H depth profile

*"15nm-10nm" means, vacancy distribution is centered at 15 nm and its FWHM is 10 nm (normal distribution is assumed)
*Peak trap concentration is 4%.

*T. Oda et al. (TITAN), Fusion Sci. Technol. 60 (2011) 1455.

(3) kMC code development



<Defect loading>
➢Ion or neutron irrad.

<Deuterium loading>
➢Fulx:$1x10^{26}$ m^{-2} s^{-1}
➢Energy: 200 eV
➢Fluence:$4.4x10^{21}$ m^{-2}

<Heating>
➢Rate:10^8 K s^{-1}

Figure: Comparison in TDS spectra

*T. Oda et al. (TITAN), Fusion Sci. Technol. 60 (2011) 1455.

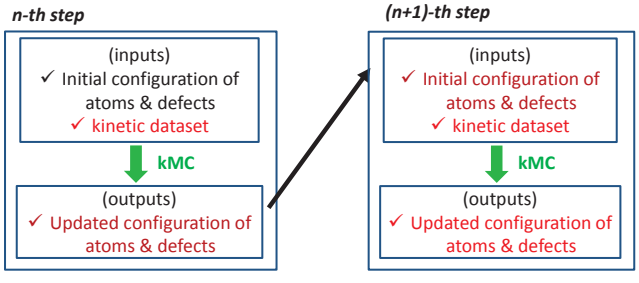
(3) kMC code development

- ✓ The difficulty of "H in W" is the large difference in the event of H and W.
  - ➢ Vacancy-related evens usually hold the activation energies of 1 eV.
  - ➢ H-related events usually hold the activation energies of 0.1 eV.
- ✓ If we consider "H jump" (for migration) as "the most frequent event", the time step of kMC needs to be around $1x10^{-13}$ s (0.1 ps) at 800 K, for example. So, we cannot do simulation comparable with the time scale of experiments.
- ✓ To create a code to evaluate the tritium retention, we may mainly have 3 ways:
  - ✓ (1) Directly perform kMC
  - ✓ (2) Modify rate-theoretical model in TMAP-wise codes based on atomic-scale knowledge.
  - ✓ (3) Come up with a further coarse-graining methods. (for example, treating hydrogen in a continuum model, while treating W in an atomistic model)

>> (1) may not be so promising, but we work on it for this CRP.

(3) kMC code development

- ✓ Even we elaborate a very accurate potential model, some errors are still involved in the database constructed by classical MD (even in a database by DFT, strictly speaking).
- ✓ So, propagation of errors in the inputs (kinetic database) to outputs (configuration of atoms and defects, such as tritium inventory, release temperature, defect structures, etc) needs to be assessed.



(3) kMC code development

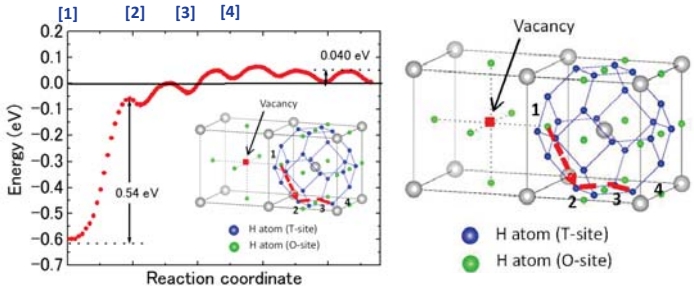
- ✓ There are mainly two ways to know the degree of error propagation:
  - ✓ (1) Direct stochastic sampling: conduct many simulations with changing the value of a certain parameter (e.g. activation energy) to check the significance of the parameters. (sensitivity analysis)
    - ✓ [Pros] easy to implement
    - ✓ [Cons] large calculation cost; correlation of parameters may not be so well understood.
  - ✓ (2) Using the error propagation formulations.
    - ✓ [Pros] correlation of parameters may be clearly given; less cost
    - ✓ [Cons] need some approximations; may not be doable for complicated cases.

- ✓ Prof. Shim (Secondary CSI) have studied the error/uncertainty propagation for neutronics calculations. He will work on this topic.

*H.J. Park, H.J. Shim, C.H. Kim, "Uncertainty Propagation in Monte Carlo Depletion Analysis," Nucl. Sci. Eng., 167, 196-208 (2011).

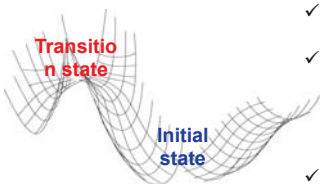
(Appendix) What else we may do from the theory

- ✓ In addition, the detrapping process is not a elementary process.
- ✓ The determined effective frequency (trial frequency for detrapping), $1x10^{12}$ s^{-1}, is only for H jump from [1] to [2].
- ✓ To be fully detrapped, the H atom has to move more away, like [4]. Some of H atoms arrived [2] may arrive [4], but most come back to [1].
- ✓ Considering this effect (which can be evaluated, although not done here), $1x10^{10}$ s^{-1} as the pre-exponential factor seems reasonable.



(Appendix) What else we may do from the theory

(Example) pre-exponential factor of detrapping: $1x10^{10}$ s^{-1} is too small?



- ✓ The frequency of vibrational motions of H is ~$5x10^{13}$ s^{-1} in metals.
- ✓ However, the frequency of trial for detrapping (= the effective frequency, ν_0) should include the effect of potential surface shape (curvature) around the transition state.
- ✓ Then the effective frequency (within harmonic transition state theory*)

$$\nu_0 = \left(\prod_{i=1}^{3N} \nu_i\right) \Big/ \left(\prod_{i=1}^{3N-1} \nu_i'\right)$$

The product of all vibration modes at the initial state (local/global minimum)

The product of all vibration modes at the transition state (1st-order stationary point)

- ➢ For example, for detrapping of H atom from a vacancy in bcc-Fe, the effective frequency is around $1x10^{12}$ s^{-1}. (using a potential model, not DFT)

*G.H. Vineyard, J Phys. Chem. Solids 3 (1957) 121, for example.

Summary

Our research objective is

(i) to establish a computational methodology to simulate the tritium behavior in irradiated tungsten which contains radiation defects, then

(ii) to acquire estimates of the tritium inventory buildup in plasma-facing tungsten under some proposed fusion reactor conditions.

To achieve this overall goal, we work on 3 subjects for 3 outcomes:

(1) a high-quality potential model for W-H system
(2) a kinetics database relevant to the tritium behavior in irradiated W
(3) estimates of the tritium retention (code development) in plasma-facing W under some proposed fusion reactor conditions

We are happy to work on something to fill uncovered gaps among what other CRP participants cover.