Multi-scale modeling on hydrogen permeation and leakage behavior
水素透過・漏洩のマルチスケールモデリング

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Monte Carlo simulation was performed in order to analyze the effects of barrier coverage on the performance of permeation barrier. It was observed that the barrier performance was not linearly dependent on barrier coverage and that it was necessary to maintain the barrier coverage of 99.9% in order to reduce the permeation by 90%. Therefore, it is important to keep a high coverage condition to reduce the permeation adequately.

1. Introduction

In fusion reactor design, it is important to minimize tritium leakage from the reactor (mainly through metals/alloys) in order to realize a safety and efficient fuel cycle. Permeation through metals/alloys is one the key steps in the leakage.

Hydrogen-metal interactions have been paid attention to due to its practical importance, e.g. hydrogen embrittlement and hydrogen storage. Hence, basic properties such as diffusion coefficient and solubility of hydrogen isotopes have been determined in most metals and various alloys. These data may be applied to estimate tritium leakage amount. However, since not only diffusion or solution in the bulk but also surface reactions is involved in hydrogen permeation, it is necessary to integrate these fundamental steps reasonably.

In the case of experiments, hydrogen permeation can be directly analyzed at specific conditions. However, control of all parameters governing permeation is difficult. In addition, numerous efforts are required to determine the importance of each parameter and the correlation among these parameters. Hence, modeling study is needed to deepen understandings effectively and systematically.

In the previous study, we developed a code to model permeation behavior of hydrogen isotopes by means of Monte Carlo (MC) technique [1]. Because the number of available data sets is adequate in both experiment and calculation, bcc Fe was used as a test material. In this code, a potential energy curve was described according to *ab initio* calculation results of density functional theory (DFT) [2, 3]. Then, permeation behavior of hydrogen isotopes under this potential curve was simulated by MC. Consequently, the simulation results well reproduced hydrogen permeability observed by experiment [4].

In the present study, effects of permeation barrier located on surfaces were studied using that Monte Carlo code, with focusing on the influence of breaking of permeation barrier.

2. Method

Permeation behavior of hydrogen isotopes was divided into 4 steps; (1) adsorption, (2) migration between surface and bulk, (3) diffusion and (4) desorption. Model parameters such as diffusion barriers and adsorption energy were mainly derived from refs. 2 and 3. In these studies, *ab initio* calculations by density functional theory (DFT) with a GGA-PBE functional were conducted. Prepared potential energy curve used in MC simulation is given in Fig. 1.

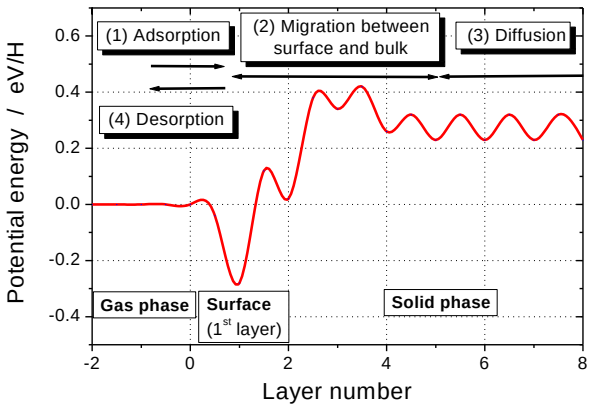

Fig. 1. Potential energy curve obtained by DFT calculations (most data were from refs. [2, 3])

In the present study, we fixed the following simulation conditions:

- ✓ Permeating species: hydrogen molecule (H_2)
- ✓ Temperature: 773 K
- ✓ System: 100×100 slab models of 2D periodic boundary conditions (the number of layers along z direction is changed to check depth dependence of simulation results)

A schematic drawing of simulation system is given in Fig. 2. The yellow-colored area indicates the surface where permeation barrier does not exist (or is broken), while the blue-colored area the surface where permeation barrier exists. In order to check effects of barrier coverage, we changed the ratio of [yellow-colored area] to [whole area] (yellow + blue). In addition, the number of layers along z direction was changed to check depth dependence of simulation results. We have confirmed that 100×100 slab model is enough large to neglect effects of periodic boundary conditions.

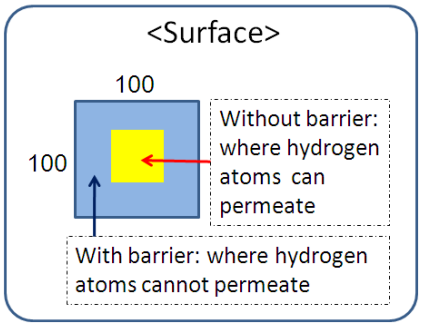

Fig. 2. A simulation system with/without barriers

3. Results & Discussion

The simulation results about effects of barrier coverage on normalized permeation coefficient are given in Fig. 3. The barrier covers the entrance side of surfaces (higher pressure side). It is assumed that this permeation barrier can prevent hydrogen permeation perfectly if the barrier exists. In Fig. 3, it is clear that the barrier performance is not linearly dependent on barrier coverage. Specifically, even 10% of barrier breaking largely degrades the performance of barrier, and permeation was not adequately decreased. In order to reduce the permeation by 90%, it is necessary to maintain the barrier coverage of 99.9%.

The effects of barrier coverage are also dependent on the number of layers along z direction. As the number of layers increases, the barrier breaking degrades the barrier function more significantly. Therefore, in a realistic case, it is important to keep a high coverage condition.

In the case of exit-side barrier (lower pressure side), the degree of degradation due to barrier breaking is slightly weaker than entrance-side barrier (higher pressure side). The details will be given in the presentation.

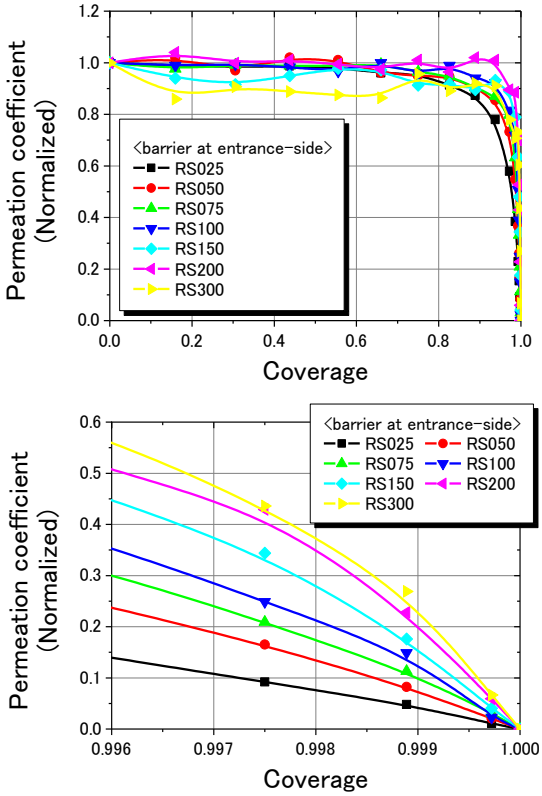

Fig. 3. Effects of barrier coverage on normalized permeation coefficient. The lower figure is magnified one focusing at the coverage of 0.996-1.000. For example, "RS075" indicates that the number of layers along z direction is 75.

4. Summary

Monte Carlo simulation was performed in order to analyze the effects of barrier coverage on the performance of permeation barrier. It was observed that the performance was not linearly dependent on barrier coverage, and that it was necessary to maintain the barrier coverage of 99.9% in order to reduce the permeation by 90%. Therefore, it is important to keep a high coverage condition to reduce the permeation adequately.

References

[1] T. Oda, Y. Oya, K. Okuno, S. Tanaka: Fusion Sci. Technol. **54** (2008) 537.
[2] D.E. Jiang et al.: Phys. Rev. B **70** (2004) 064102.
[3] D.C. Sorescu: Catalysis Today **105** (2005) 44.
[4] O.D. Gonzales: Trans. Metall. Soc. **245** (1969) 607.