Hybrid Simulation between Molecular Dynamics and Binary Collision Approximation Codes for Hydrogen injection onto Carbon Materials

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Molecular dynamics (MD) simulation with modified Brenner’s reactive empirical bond order (REBO) potential is a powerful tool to investigate plasma wall interaction on divertor plates in a nuclear fusion device. However, MD simulation box’s size is less than several nm for the performance of a computer. To extend the size of the MD simulation, we develop a hybrid simulation code between MD code using REBO potential and binary collision approximation (BCA) code. Using the BCA code instead of computing all particles with a high kinetic energy for every step in the MD simulation, considerable computation time is saved. By demonstrating a hydrogen atom injection on a graphite by the hybrid simulation code, it is found that the hybrid simulation code works efficiently in a large simulation box.

I. INTRODUCTION

To understand a mechanism of chemical and physical interactions between hydrogen plasmas and divertor plates in a nuclear fusion device, it is necessary to study an elementary processes of the reactions. In molecular dynamics (MD) simulations, the equations of motion of atoms are solved numerically. We investigated plasma-wall interaction on the divertor plates made of carbon materials by the MD simulation with modified Brenner’s reactive empirical bond order (REBO) potential\textsuperscript{1,2} in the previous works\textsuperscript{3–5}. A typical scale length of the MD simulation box in these works are several nm.

In order to expand the simulation box to more realistic scale length, i.e., several µm, we develop a hybrid simulation code between the MD simulation code with the REBO potential and atomic collision in any structured target (AC\textsubscript{∀}T) code\textsuperscript{7}. In the AC\textsubscript{∀}T code, which uses binary collision approximation (BCA), a two-body interaction is calculated instead of computing all particles for every step in the MD simulation. Therefore computation time is saved. We choose the AC\textsubscript{∀}T code as the partner of the MD simulation for its fast calculation of a location and a velocity of the particle. The main problem of hybridization between the two simulation codes is how to define a threshold kinetic energy of the injection particle. For higher kinetic energy of particle than the threshold kinetic energy, we calculate the trajectory of the particle by the AC\textsubscript{∀}T code. As the kinetic energy is dissipated to its surroundings and attaches to the threshold energy \(E_{th}\), we give a position and a velocity of the projectile to the MD simulation code as the initial condition. Then we perform the MD simulation. Generally speaking, because a multi-body interaction with the neighborhood of the projectile must be calculated in the MD simulation code with the REBO potential, computation time becomes longer than in the AC\textsubscript{∀}T code. To make the performance of the simulation better, it is necessary to shorten the MD simulation and extend the AC\textsubscript{∀}T simulation box. However, it is known that the AC\textsubscript{∀}T code cannot calculate exactly the particle motion in a low energy. Comparing the MD simulation with the AC\textsubscript{∀}T simulation, two simulation results are consistent in higher kinetic energy than 200 eV\textsubscript{12}. Thus we set \(E_{th}=200\) eV at which a simulation algorithm changes from the AC\textsubscript{∀}T simulation to the MD simulation.

II. AC\textsubscript{∀}T-MD HYBRID SIMULATION

A concept of hybrization between the MD simulation and the AC\textsubscript{∀}T simulation is that the AC\textsubscript{∀}T simulation works when a kinetic energy of a projectile is higher than the threshold energy \(E_{th}=200\) eV, while the MD simulation works when the kinetic energy becomes lower than the threshold energy \(E_{th}\).

To explain the hybrid simulation more concretely, let’s consider a hydrogen atom injection onto a graphite (Fig. 1). An injection kinetic energy of the hydrogen atom is set to 1 keV.
which is higher than $E_{th}$. While the kinetic energy of the hydrogen atom is higher than $E_{th}$, trajectories of the hydrogen atom and the surrounding carbon atoms are calculated by the AC/$\mathrm{T}$ simulation. The hydrogen atom dissipates its kinetic energy by interacting with carbon atoms in diamond and then the kinetic energy becomes lower than $E_{th}$. At that moment, the MD simulation starts to calculate the motions of the hydrogen and the carbon instead of the AC/$\mathrm{T}$ simulation. The master process (4) informs the splitter process of the completion of the AC/$\mathrm{T}$ process with the list of moved atoms. The splitter process updates its material data with the list of moved atoms. (5) Then, the MD simulation box are picked up from the entire target material and they are sent to the MD processes. The MD simulation is started in each MD processes. (6) We will explain the algorithm of the AC/$\mathrm{T}$ simulation, the MD simulation and the splitting program in the following subsections.

A. AC/$\mathrm{T}$ Simulation

A binary collision between a projectile and a target atom is necessary in the AC/$\mathrm{T}$ simulation\(^2\). During the binary collision, a total energy, a total momentum and a total angular momentum are conserved. Due to these conservations, the final position and velocity of the projectile and the target atom are obtained analytically on a potential $V(r)$, where $r$ is the distance between the projectile and the target atom. In the AC/$\mathrm{T}$ code similarly to the ACAT code, the Moliere approximation is adopted and then $V(r)$ is given by the Thomas-Fermi potential as follows:

$$V(r) = \frac{Z_1 Z_2 e^2}{r} \Phi(r/a),$$

$$\Phi(x) = 0.35e^{-0.3x} + 0.55e^{-1.2x} + 0.10e^{-6.0x},$$

where $a$ is a screening length, $Z_1$ and $Z_2$ are atomic numbers of the projectile and the target atom, respectively. Unlike the MD simulation, the AC/$\mathrm{T}$ simulation directly gives us the asymptotic trajectory of the projectile collided with the target. In the AC/$\mathrm{T}$ simulation, the projectile hits the target atoms and then it brings about cascade shower in the large simulation box.

B. Molecular Dynamics Simulation

The equation of motion in the MD simulation is written by

$$r_i = \frac{p_i}{m_i}, \quad \dot{p}_i = -\frac{\partial U}{\partial r_i},$$

where $r_i$, $p_i$ and $m_i$ are the position, momentum and the mass of the $i$-th atom. The function $U$ is total potential energy. In our MD simulation, the modified REBO potential is used for the potential $U$ which is defined as:

$$U \equiv \sum_{i,j>i} U_{ij}, \quad U_{ij} \equiv V_{ij}^R(r_{ij}) - \tilde{b}_{ij}\{r_i\}, \{\theta^B\}, \{\theta^{\mathrm{DH}}\} V_{ij}^A(r_{ij}),$$

where $r_{ij}$ is the distance between the $i$-th and the $j$-th atoms. The functions $V_{ij}^R$ and $V_{ij}^A$ represent repulsion and attraction, respectively. The function $\tilde{b}_{ij}$ generates a multi-body force. The bond angle $\theta^{\mathrm{DH}}_{kij}$ is the angle between the vector from the $i$-th atom to the $j$-th atom and the vector from the $i$-th atom to the $k$-th atom. The dihedral angle $\theta^{\mathrm{DH}}_{kijkl}$ is the angle between the plane passing through the $k$-th, $i$-th, $j$-th and $l$-th atoms. Moreover, the parameters $\{r_i\}$, $\{\theta^B\}$, and $\{\theta^{\mathrm{DH}}\}$ denote all sets of $r_{ij}$, $\theta^{\mathrm{DH}}_{kijkl}$, and $\theta^{\mathrm{DH}}_{kijkl}$, respectively (for details of the modified Brenner REBO potential, see Refs.\(^2\) and\(^6\)). The second-order symplectic integration\(^{12} is used to execute the time integration of the equation of motion where the time step is set to $5 \times 10^{-18}$ s.

Unlike the ACAT simulation, the MD simulation cannot treat a large scale material. To calculate efficiently, local boxes 14 Å on a side are picked up, and the simulation for the atoms...
in each box is executed in parallel. To obtain precise forces acting on all atoms in the MD simulation boxes 14 Å on a side, the bond energy $U_{ij}$ in Eq. (4) is calculated in a larger cubic box 26 Å on a side. Furthermore, the function $\bar{b}_{ij}$ depends on more outer covalent bonds. Consequently, the MD process needs the positions of atoms in a cubic box 34 Å on a side. Namely, in the MD process, the equations of motions are solved about atoms in the cubic box 14 Å on a side, and forces acting on the atoms are calculated by aggregating surrounding atoms in a cubic box 34 Å on a side as shown in Fig. 3.

C. Detamination of MD Simulation Box

By the MPMD method, one CPU is allocated into the MD process treating one cubic box 34 Å on a side. However, there is the special case that two or more cubic boxes overlap because the moving particles are close. These overlapping boxes are regarded as a combined MD simulation box for one CPU process. The determination for the MD simulation box is carried out by the splitting program (See Fig. 4) as follows. The simulation box in the AC∀T code is divided into cubic cells 2 Å on a side, where 2 Å is the cutoff length of covalent bond in the REBO potential. $17 \times 17 \times 17$ cells, which are corresponding to the cubic box 34 Å on side, are marked up. Only a center cell of the marked cells is a member of the MD simulation box, initially. The center cell is set to the first target cell and the member of the MD simulation box is selected by the following recursive routine: if adjoining cells of the target cell has been marked up, they are added into the member of the MD simulation box and become next target cells, while if they have not been marked up, the recursive routine is stopped.

III. NUMERICAL DEMONSTRATION

We demonstrate an AC∀T-MD hybrid simulation for a hydrogen atom injection onto a graphite, whose size is 321 Å long, 347 Å wide and 335 Å depth which consists of 100 graphene. Periodic boundary conditions are imposed on the horizontal direction. The initial temperature of the graphite is set to zero Kelvin. The hydrogen atom is injected normally onto the surface at 1 keV. 900 simulations with the same initial material and randomly changed injection position have been performed. As a benchmark, stand alone AC∀T simulations have also been performed. Table I shows the average and the standard deviation of the depth $d_\perp$ and the horizontal distance $d_\parallel$ from the surface to the final positions in the result of both the AC∀T-MD hybrid simulation and the AC∀T simulation. Although the averages of depth in the both cases are almost equal, the average and the standard deviation of horizontal distance by the AC∀T-MD hybrid simulation are smaller than that of the AC∀T simulation. Figure 5(a) and (b) show 30 samples chosen randomly in 900 trajectories calculated by the AC∀T and the AC∀T-MD hybrid code, respectively. Figure 5(b) shows the trajectories in the same initial condition calculated by the AC∀T-MD hybrid simulation. The gray and blue lines in Fig. 5(b) show the trajectories of which indicate the AC∀T contribution and the MD contribution, respectively. Compar-
not treat the binding from surrounding atoms. The MD simulation includes it by the multi-body force. Therefore, the atom is not possible to move a long distance for low kinetic energy in the ACvT-MD hybrid simulation as shown in Fig. 5(c). In our previous research, it was shown that there is an adsorption site 1.1 Å above each of the carbon on a graphene sheet. The hydrogen calculated by the MD dissipates its kinetic energy quicker than the ACvT case and finally be trapped at the adsorption site.

IV. SUMMARY

The ACvT-MD hybrid simulation code has been developed to treat large materials, long time and wide range kinetic energy in PSI. The motion of hydrogen atom is solved by BCA method using the ACvT code while its kinetic energy is higher than 200 eV. When the kinetic energy becomes lower than 200 eV, the MD code is invoked around atoms moved by the ACvT simulation.

The ACvT-MD hybrid simulation was performed and compared with stand alone ACvT simulations of 900 hydrogen atom injections onto a graphite. Their results suggested that the ACvT-MD hybrid simulation tends to derives a shorter trajectory than the ACvT simulation does. This fact is caused by the binding from the surrounding atoms, which is ignored in the ACvT simulation.

V. ACKNOWLEDGEMENT

Our deep appreciation should go to Prof. Takahiro Kenmotsu of Faculty of Life and Medical Sciences, Doshisha University. Numerical simulations were carried out using the Plasma Simulator at the National Institute for Fusion Science, Japan. This work is supported by the National Institutes of Natural Sciences’ undertaking Forming Bases for Interdisciplinary and International Research through Cooperation Across Fields of Study and Collaborative Research Program (No. NIFS09KEIN091) and Grants-in-Aid for Scientific Research (No. 19055005) from the Ministry of Education, Culture, Sports, Science and Technology, Japan. This work is performed with the support and under the auspices of the NIFS Collaboration Research program (NIFS09KTAN008).

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1 D. W. Brenner, O. A. Shenderova, J. A. Harrison, S. J. Stuart, B. Ni, and S. B. Sinnott: J. Phys.: Condens. Matter 14 (2002) 783.
2 A. Ito and H. Nakamura, Commun. Comput. Phys., 4 (2008) 592.
3 A. Ito and H. Nakamura: J. Plasma Phys. 72 (2006) 805.
4 A. Ito, H. Nakamura and A. Takayama, J. Phys. Soc. Jpn. 77 (2008) 114602.
5 S. Saito, A. Ito and H. Nakamura, ‘Incident angle dependence of reactions between graphene and hydrogen atom by molecular dynamics simulation’, Plasma and Fusion Research Special Issue, Accepted for Publication (2010).
6 M. Suzuki, J. Math. Phys., 26, 601 (1985).
7 A. Takayama, S. Saito, A. M. Ito, T. Kenmotsu, ‘Extension of the simulation code ACAT to treat real atomic positions’, submitted to JJAP. [arXiv:1005.3344]
8 A. Takayama, K. Shimizu, Y. Tomita and T. Takizuka, ‘A new

FIG. 5: 30 sample trajectories of incident hydrogen that move in graphite calculated by the (a) stand alone ACvT code and (b) ACvT-MD hybrid simulation, respectively. (c) Detailed image of the MD simulation part for trajectory ‘A’ in (b).
framework for integrated simulation model using MPMD approach’, JPFRS., (accepted) (2010).

9 A. Ito and H. Nakamura: Jpn. J. Appl. Phys. 47 (2008) 4715.
10 A. Ito, H. Nakamura, and A. Takayama: J. Phys. Soc. Jpn. 77 (2008) 114602.
11 M. Suzuki: J. Math. Phys. 26 (1985) 601.

12 A. Ito, T. Kenmotsu, Y. Kikuhara, K. Ohyu, K. Inai, Y. Wang, S. Irle, K. Morokuma and H. Nakamura, The 4th IAEA-TM on the Theory of Plasma Instabilities, May 18-19, 2009, Kyoto University, Japan, P2-23.