Molecular Dynamics Simulation of Xenon at High Temperature for a Nuclear Fuel

Je-Yong Oh, Yang-Hyun Koo, Jin-Sik Cheon, Byung-Ho Lee, Dong-Seong Sohn
Korea Atomic Energy Research Institute, P.O. Box 105, Yuseong, Daejon, 305-600, tylor@kaeri.re.kr

1. Introduction

Since the fission gas products in a fuel pellet could affect the swelling of a fuel and the properties of a fuel pellet, an accurate prediction of a fission gas behavior is necessary for an evaluation of the integrity and performance of a fuel rod. As most of the elements of a fission gas are Xenon (Xe) atoms and the estimated equilibrium pressure of a gas bubble in a fuel pellet is not so high, either the Van der Waals equation or the ideal gas law have traditionally been used due to their simple equation forms [1].

However, there is a possibility that the Van der Waals equation would not be applicable to the gas bubbles in a rim structure which has been observed at the periphery of a high burnup UO_2 fuel pellet [2]. As it is very hard to reach an equilibrium state of the gas bubbles in the rim structure due to its low temperature, the pressure of rim bubbles is predicted to be from tens to hundreds MPa, which is much higher than the equilibrium pressure [2,3]. Although the reliability of the equations of state can be confirmed by a comparison with experimental data, the experimental data are very limited at a high temperature and pressure for a nuclear fuel pellet.

To fill the lack of experimental data, the pressure-volume-temperature relationship of Xe was simulated by the molecular dynamics (MD) for a nuclear fuel operating temperature in this paper.

2. Molecular dynamics simulation

In this section the assumptions and model used for the molecular dynamics simulation are described.

2.1 Interatomic potential

The interatomic potential between atoms is the most important factor for the MD simulation because it describes the force between atoms and the MD simulation is carried out by the integrate Newton's equations of motion.

The general interatomic potential function for an inert gas is the Lennard-Jones (n,m) potential,

$$U(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^n - \left(\frac{\sigma}{r}\right)^m\right] \qquad (1)$$

where ε and σ are the Lennard-Jones energy and length parameters, and r is the interatomic distance. The n and m are exponents for the Lennard-Jones potential. The Lennard-Jones (n,m) potential is suitable for inert gases near the equilibrium condition, but it is not suitable for a very small r, i.e. very close distance between atoms, where the repulsive term is more dominant [4].

Hence we use the exponential-six (exp6) potential [5] instead of using the Lennard-Jones potential,

$$U(r) = \varepsilon\left\{\frac{6}{\alpha-6}\exp\left[\alpha\left(1-\frac{r}{\sigma}\right)\right] - \frac{\alpha}{\alpha-6}\left(\frac{\sigma}{r}\right)^6\right\} \qquad (2)$$

where α is the fitting parameter for the exponential-six potential. α =13.0, ε /k = 243.1K, σ = 4.37Å are used, and k is the Boltzamann constant.

2.2 Condition for Simulation

The molecular dynamics (MD) simulation was performed in the NVT ensemble (constant N-number of atoms, V-volume, T-temperature) with the modified MXDORTO program [6] for this study. To find an appropriate system size, the effect of numbers of simulated atoms was investigated from 100 to 2000 atoms. Although 500 atoms were enough for most cases, the condition of 2000 atoms was selected for a reduction of the deviation. And Xe atoms were distributed randomly. Δt (time step) was 0.002 ps and the number of time steps was 10000.

3. Results and Discussions

When a molecular dynamic simulation is carried out at a low pressure where the gas atoms obey the ideal gas law, the interatomic potential hardly affects the behavior of the gas atoms due to the large distances between the atoms. Hence a comparison of the calculated pressures with the measured ones at a high pressure should be made to verify the MD simulation results. As there is no proper experimental data for Xe at a high temperature and pressure in a nuclear fuel pellet, experimental data at a room temperature [7] was used for a verification of the high pressure condition. Table 1 shows that the calculated pressures are in good agreement with the measured data.

Table 1. Comparison of the simulated pressures with the measured ones at room temperature

Volume [cm^3/mol]	Measured Pressure [GPa]	Calculated Pressure [GPa]
32.10	0.80	0.83
31.53	0.95	0.93
27.51	2.39	2.04
25.98	3.02	2.83

The MD results were compared with the equation of state for Xe at 900K as shown in Fig. 1 and 1600K as shown in Fig. 2. The temperature of 900K was chosen as the typical temperature for the rim structure and the temperature of 1600K for the centerline temperature in the fuel pellet. The ideal gas law underestimated the pressures at both temperatures because it did not consider the Van der Waals force (repulsive force). The Van der Waals equation generally overestimated the pressure and had a certain threshold to apply.

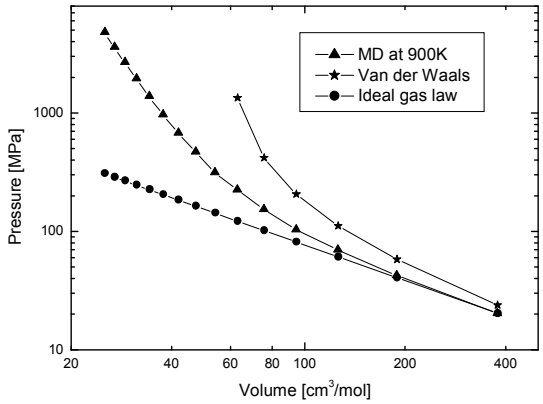

Fig. 1. Comparison of the MD data with the equations of state for Xe at 900K.

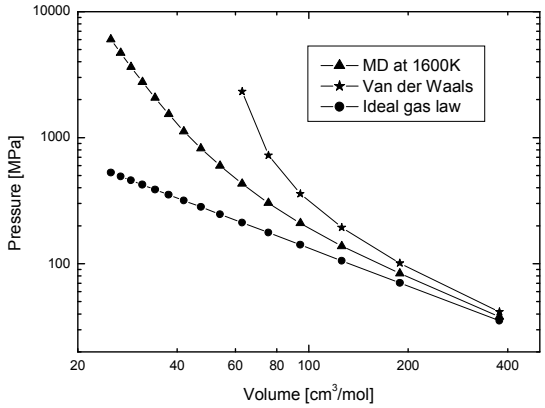

Fig. 2. Comparison of the MD data with the equations of state for Xe at 1600K.

4. Conclusion

The molecular dynamics simulation of Xenon was carried out at the nuclear fuel operating temperatures with the exponential-six potential. The simulation results were verified by a comparison of the experimental data, and it showed a good agreement.

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