Multiscale simulation models of Xe bubble formation in irradiated Mo

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OUTLINE

Multiscale simulation models for Xe bubble nucleation and growth in irradiated Mo were developed that consist *Ab-initio* calculations of the interatomic potentials for the Mo and Xe-Mo systems, atomistic MD simulations of the kinetic rate coefficients of radiation defects, and nucleation mechanisms of Xe bubbles in Mo. Simulations of various Xe concentrations, temperatures and pressures were carried out. A critical concentration of Xe atoms was determined at which the nucleation occurs spontaneously.

INTRODUCTION

One of the major problems in the nuclear fuel development is fuel swelling due to fission gas production during fission reactions, which is a challenging experimental and theoretical task for the nuclear fuel technology. Noble gases are practically insoluble in the metal fuels and are always rejected from the solid matrix, and their normal state is mostly gaseous [1]. Xenon in the fuel either gets released into the plenum or forms gas bubbles in the fuel matrix. Fuel swelling as a result of gas bubble accumulation is destructive for the fuel and will eventually lead to fuel and cladding failure.

Small bubbles of Xe and Kr were observed both in accelerator experiments and in reactor experiments with a variety of nuclear materials. The inert gas bubbles can also migrate, coalescence and grow by interacting with irradiation-induced point defects such as vacancies and dislocation loops. It was also recognized that fission gas bubbles could serve as very efficient traps for dispersed gas atoms in nuclear materials [1].

The ultimate goal of the fission gas simulation is to predict the concentration of the atomically dispersed gas atoms in the matrix, depending on the fuel element location and the duration of irradiation; the distribution function of the bubbles over their sizes in the matrix and on extended defects, such as grain boundaries and dislocation lines; the temperature dependence of the diffusion coefficient of the fission gases in the matrix and the amount of the gas released from the fuel.

Molecular dynamics is a powerful atomistic simulation method that can predict the nucleation rate of Xe and is a useful tool for the development of nuclear fuels in predicting Xe bubble formation and lattice swelling [2,3]. However, the accuracy of this method strongly depends on the quality of the interatomic potentials.

In this paper, we present our preliminary simulation results of Xe gas nucleation characteristics that were obtained by atomistic molecular dynamics with a new interatomic potential developed at Argonne for a binary Mo-Xe system by using an *Ab-initio* DFT simulation method and published in a previous paper [4].

COMPUTATIONAL APPROACH

Nucleation of Xe bubbles in pure Mo were investigated by molecular dynamic simulation performed at various initial Xe concentrations; temperatures, and gas pressures by large-scale parallel calculations with LAMMPS simulation package [5].

The calculation cell contained 432000 Mo atoms and Xe atoms were added at interstitial positions randomly. The calculation cell size was determined by the requirement of setting up the total pressure in the system equal to zero, at the initial time instant. Such equilibration procedure was efficiently adding several vacancies per Xe atom, depending on the Xe concentration. The simulations without equilibrating the pressure were not showing bubble formation. The total simulation time was equal to 10 ns. A spontaneous nucleation mechanism similar to the spinodal decomposition was obtained at concentrations of Xe atoms above a critical one. The temperature of simulations was set at a low initial temperature and was equilibrated at 700K. During the bubble formation process, the latent heat release was removed via a velocity rescaling thermalization procedure.

RESULTS

The estimated critical concentration of Xe atoms at 600K was obtained to be 1.6 at. % ($n_{Xe} = N_{Xe}/N_{total} = 0.016$). Typical final atomic arrangements obtained in this simulation are shown in Fig. 1. At low Xe concentration of 0.8 at. %, bubble formation was not observed and the lattice was almost not distorted. At Xe gas concentration of 5.8 at. %, the average diameter of the bubbles is 1.25 nm, and the lattice was slightly distorted. The diameter increases to 2.25 nm at the gas concentration of 11.1% and the lattice is heavily deformed.

Our preliminary simulation results showed that Xe bubble formation is triggered at a Xe concentration of about 1.6 at. %. Simulation of bubble formation with and without adding vacancies to the system showed that bubble formation at low temperatures was significantly accelerated if several vacancies per Xe atom were efficiently added into the simulation system.

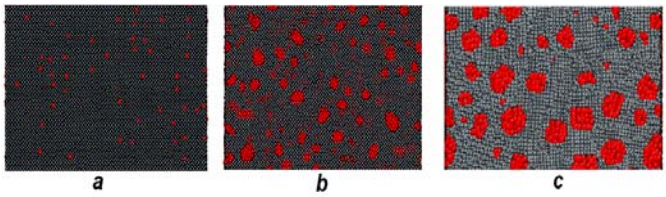

Figure 1. The snapshots in this figure shows examples of the arrangement of atoms in calculation cell at different Xe gas concentrations: gray atoms – Mo; red atoms – Xe. The various situations are simulated: a) – n_{Xe} = 0.8 at. %, T_0 = 600 K, no nucleation was observed; b) – n_{Xe} = 5.8 at. %, T_0 = 600 K, T_{final} = 1200 K, modest nucleation with an average bubble diameter of 1.25 nm, the lattice is slightly deformed; c) - n_{Xe} = 11.1 at. %, T_0 = 600 K, T_{final} = 2100 K, strong nucleation observed, with the average bubble size of 2.2 nm, the lattice is heavily deformed.

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