Atomistic simulation of defects formation and structure transitions in U-Mo alloys at swift heavy ion irradiation

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Abstract. At irradiation of swift heavy ions, the track formation frequently takes place in nuclear materials. There is a large interest to understanding of the mechanisms of defects/track formation at this phenomenon. In this work, the atomistic simulation of defects formation and melting in U-Mo alloys at irradiation of swift heavy ions has been carried out. We use the two-temperature atomistic model with explicit account of electron pressure and electron thermal conductivity. This two-temperature model describes ionic subsystem by means of molecular dynamics while the electron subsystem is considered in the continuum approach. The various mechanisms of structure changes at irradiation are examined. In particular, the simulation results indicate that the defects formation may be produced without melting and subsequent crystallization. Threshold stopping power of swift ions for the defects formation at irradiation in the various conditions are calculated.

1. Introduction
At irradiation of swift heavy ions (e.g. Xe ion - the typical fission fragment), the track formation frequently takes place in nuclear materials. The mechanism of the formation and structure of ion track has been a subject of a discussion since the late 50s of the last century [1, 2]. This phenomenon has great significance in radiation material science and nuclear engineering because a typical example of swift heavy ion is a fission fragment in a nuclear fuel (the initial energy of such an ion is about 100 MeV). In addition, the great interest in track formation is caused by the importance of fundamental problems in this field which need to be resolved.

Uranium has received a lot of attention for its unique nuclear properties and its various applications in nuclear industry. A high-temperature γ-phase has the best technical properties for nuclear engineering because it has body-centered cubic (bcc) lattice. However, γ-phase is extremely unstable at low temperature. For this reason, uranium is alloyed with other metals, which have bcc structure. Mo exhibits a high solubility in γ-U. Compared with other high density uranium alloys and compounds, the low-enriched uranium alloys with 6-12 wt.% of Mo have attracted a great deal of attention and are recognized as the most prominent candidates for advanced research and test reactors, because they have a relatively larger γ phase region and present more stable irradiation performance.
Mo is a strong γ-stabilizer which provides stable swelling behavior in U-Pu-Mo fuels, has high thermal conductivity, low thermal expansion, and high melting points [3–5].

Phase and structural transformations during irradiation are important for describing the evolution of the nuclear fuel in the operating conditions and fuel fabrication because the physical properties of U-Mo alloy strongly depend on the material lattice structure. There are a lot of models describing track formation in different materials. In this work the ion track formation in U-Mo alloy is associated with defect formation as it is usually done in pure metals [6, 7]. The mechanism of defect formation during irradiation is usually associated with collision cascades [8–10] or with melting and subsequent crystallization of material [11, 12]. In this work it is shown that several number of defects may be produced without melting and subsequent crystallization.

This work is devoted to theoretical analysis of the phenomena taking place in a nuclear fuel material under operating conditions. The simulation of defects formation and structure transitions in U-Mo alloys at irradiation of swift heavy ions has been carried out using the two-temperature atomistic model [12–14] with explicit account of electron pressure and electron thermal conductivity [12]. The various mechanisms of structure changes at irradiation are examined.

2. Computational technique

The structures of U–Mo alloys and the ion subsystem were described using molecular dynamics simulation with the novel interatomic angular dependent potential [15]. This potential contains an angular-dependent term and was parameterized using the force-matching method [16, 17]. All calculations were performed with the LAMMPS code [18]. The simulation cell had the following size: 160α₀ × 160α₀ × 6α₀, where α₀ is the lattice parameter of a U-Mo alloy structure with the periodic boundary conditions in all directions.

The energy of the swift heavy ions transfers primarily into the energy of the electron subsystem. For a short time, this produces a two-temperature state of matter, where the electron temperature \( T_e \) may be several orders higher than the ion temperature \( T_i \). In this case the use of a two-temperature model is necessary. The ionic subsystem was described by the molecular dynamics approach, while the electron subsystem was considered in the continuum approach [12–14]. Energy transport within the electronic subsystem was solved according to the heat diffusion equation with added source terms for heat transfer between the subsystems:

\[
C_e \rho_e \frac{\partial T_e}{\partial t} = \nabla (\kappa_e \nabla T_e) - g_p (T_e - T_i)
\]

where \( C_e \) is the specific heat, \( \kappa_e = D_e \rho_e C_e \) is the thermal conductivity (\( \rho_e \) is an electron density and \( D_e \) is an electron diffusivity), \( g_p \) is the coupling constant for the electron-ion interaction.

The electronic specific heat is taken in the form:

\[
C_e = C_0 + C_1 T_e
\]

where \( C_0 \) is equal 1.3·10^{-10} eV/(K\( e \)), \( C_1 = 1.37·10^{-6} \) eV/(K²\( e \)).

Relaxation of strong electron excitations generated by swift ions is the main factor that determines the nature of the track regions. Thus the important task for the simulation of track formation is to estimate of the thermal diffusion coefficient \( D_e \) for the two-temperature model. It was obtained from the experimental data of the thermal conductivity \( \kappa_e \) [19].

Electronic pressure effects were included in the model to account for the blast force acting on ions because of electronic pressure gradient:

\[
\vec{F}_i = -\partial U/\partial r_i + \vec{F}_\text{lang} - \nabla P_e/n_i
\]

where \( F_\text{lang} \) is a force from Langevin thermostat simulating electron-phonon coupling, and \( \nabla P_e/n_i \) is the electron blast force. The electronic pressure is taken to be \( P_e = B \rho_e C_e T_e \) [12].
3. The track formation

Thermal spike model was used for the description of track formation in this work. In this model the process of swift heavy ions irradiation passes through two main stages. The first stage involves the energy dissipation of electrons and energy transfer by means of electron-electron collisions. So the energy input from swift heavy ions was modeled by setting the initial electron temperature profile. The profile was calculated using the following model:

\[ T_e = 300 + T_{\text{max}} \cdot \exp(-d/r) \]  

(4)

The electron temperature profile was set in the plane in the center of the simulation cell, \( r \) - track radius, \( d \) - distance from the track axis. The track simulations were carried out with the swift heavy ions velocity directed along the [0 0 1] direction of the U-Mo crystalline lattice.

The second stage consists in the movement of atoms due to the electron-phonon interaction, while a high density of nonequilibrium defects is produced.

The various mechanisms of structure changes at irradiation were examined. These mechanisms significantly depend on the initial state of U-Mo alloy and ion energy. The influence of ion energy is usually described in terms of so-called stopping power \( S = dE/dx \), which defines the rate of the kinetic energy loss of an ion per unit length along its path. In this work the values of the electron stopping power were ranged from 15 keV·nm\(^{-1}\) to 35 keV·nm\(^{-1}\), which are close to the experimental data.

The atomistic simulation of defects formation and melting in U-Mo alloys at irradiation of swift heavy ions has been carried out for different initial temperatures and molybdenum concentrations. The energy input from swift heavy ion results in the increasing of the system temperature near the center of the simulation cell. The simulation results indicate that the defects formation may be produced without melting and subsequent crystallization. Apparently this is due to the low formation energy of self-interstitial atom [15]. Since the structural changes in the alloy occur at the certain temperature, the initial temperature also affects the defects formation in addition to the energy input. Therefore, threshold stopping power of swift ions for the defects formation at irradiation decreases from 33 keV·nm\(^{-1}\) to 18 keV·nm\(^{-1}\) when the temperature of U-Mo alloy increases from 300 K to 900 K (fig. 1) for U-Mo alloy with 5 atomic percents of molybdenum. Increasing the molybdenum concentration from 5 to 50 atomic percent leads to the increasing of melting temperature of U-Mo alloy (threshold stopping power increases from 22 keV·nm\(^{-1}\) to 39 keV·nm\(^{-1}\) at \( T = 700 \) K).

\[ S \]  

keV·nm\(^{-1}\)

\[ T \]  

K

**Figure 1.** The dependence of threshold stopping power on temperature for U-Mo alloy with 5 atomic percents of molybdenum: 1 - threshold stopping power for defects formation, 2 - threshold stopping power for melting of ion subsystem.
Despite the mechanism of defect formation during irradiation is usually associated with melting and subsequent crystallization of material, the simulation results indicate that the defects formation in U-Mo alloy may be produced without melting and subsequent crystallization. The possible reason for this effect is small energy formation of Frenkel pair in U-Mo alloy [15]. Thus threshold stopping power for defects formation at irradiation is 2-3 keV·nm$^{-1}$ less than threshold stopping power for the melting of the ion subsystem (fig. 1).

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