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Molecular Dynamics probing of the energy spectrum of particles in radiation stimulated processes

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Abstract

The approach based on the classical molecular dynamics (MD) is developed that allows to probe the energy spectrum of particles in radiation induced processes. To simulate the effect of particles collisions in the selected interval of the energy spectrum the "shock function" is introduced to the standard scheme of MD. This function describes the forces acting on the lattice atoms by the incident particles in the selected energy interval. The approach is illustrated by modeling the ion bombardment of triatomic model crystal with significantly different atomic masses of constituents. It can be useful in particular for a prediction of clusters type defects formed in polyatomic crystals.

Keywords: Molecular Dynamics; computer modeling; cluster defects in solids.

1. Introduction

Molecular dynamics (MD) is a widely used method in the investigations of radiation induced processes. This method is especially important when the experimental studies are time-consuming and complex, for example in the case of exposure to reactor radiation. MD calculations allow tracking the trajectories of particles, studying mechanisms of materials destruction and finding the equilibrium configurations of structural defects [1-8]. Due to the application of MD method the peculiarities of the passage of fast particles through the matter have been established.

We developed the MD software that allows studying radiation induced processes in the selected range of energy spectrum of incident particles. Mechanisms of formation of radiation defects essentially depend on the type of incident particles and their energy. As a rule at the long-term unchanging irradiation conditions a stationary energy distribution of incident particles is set. In each interval of energy spectrum of these particles the specific mechanisms of defect formation are realized. The final radiation effect is determined by a superposition of radiation effects caused by incident particles in all intervals of their energy spectrum. Therefore to decrypt the mechanisms of formation of radiation defects it is important to clarify the mechanisms of radiation-stimulated processes in different intervals of the energy spectrum of incident particles.

To simulate the effect of atomic collisions in the selected interval of the energy spectrum the "shock function" is introduced. This function determines the forces (F_{SH}) acting on the lattice atoms as described earlier in [9]. The pulses that are transferred to lattice atoms during irradiation are characterized by special random function. This function shows which atom in the irradiated sample is knocked, what energy value is chosen from the selected energy interval, and what the direction of hit is.

It should be noted that due to the superposition of radiation effects caused by incident particles from different intervals of the energy spectrum, the result of the action of particles from selected energy range can be partly or fully hidden. Therefore to estimate the role of different mechanisms in the radiation induced destruction of material it is necessary using the proposed approach to carry out the suggested simulations in different intervals of the energy spectrum and take into account the contribution of each energy interval to the final radiation effect.

The developed approach is especially useful when the compound under irradiation consists of atoms with significantly different masses. For example we demonstrate that it is possible to obtain new information regarding the formation of defects in compounds that are used as nuclear materials ($U(Al, Si)_3$). Another example is a possibility to form the appropriate track structures for creation of electronic devices in track electronics [9 – 13].

The properties of track devices depend on the shape of tracks and the electronic structure of the internal track surfaces. The necessary properties of tracks can be obtained using the proposed approach.

2. Description of the approach

If radiation effects induced by the incident particles in the energy range (E_1, E_2) of the total energy spectrum are studied, the maximal energy transferred in elastic collisions to the lattice atom of mass M by ions with mass m_{ion} is expressed as:

$$\varepsilon_{1,2} = \frac{4 m_{ion} M}{(m_{ion} + M)^2} \cdot E_{1,2} \ , \qquad (1)$$

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In (1) the transferred energies ε_1 and ε_2 correspond to the energies of incident particles E_1 and E_2.

To simulate the elementary process induced by incident particles a random function (RF) is used. RF inserted to the computer program performs three tasks:

- Selects an atom in the target lattice which gets a hit;
- Selects an energy value from the interval (ε_1, ε_2);
- Selects an orientation of the pulse transferred to the target atom.

A linear congruent generator (LCG) for RF is an algorithm that yields a sequence of pseudo-randomized numbers calculated with a discontinuous piecewise linear equation. This generator presents one of the best-known pseudorandom number generator algorithms that are easily implemented and fast, especially on computer hardware which can provide modulo arithmetic by storage-bit truncation. These generators based on linear congruent method are especially useful for non-cryptographic applications, such as modeling. They are effective and most used in empirical tests and show good statistical characteristics.
The generator described in [14 – 16] is defined by the recurrence relation:

$$X_{n+1} = (aX_n + c) < m \qquad (2)$$

where X_0 is an initial value. In our model the parameters are: m = 232, a = 214013, c = 2531011.

It is assumed in the suggested model that the number of atoms simultaneously subjected to shock is proportional to the dose rate, and the number of steps is proportional to the irradiation dose. In order to determine the kinetic energy transferred to the lattice atom the scaling of the forces F_{SH} should be implemented. As a reference point we used the energy that is necessary for irreversible displacement of lattice atom to interstitial position in elastic collision (E_d). Then the following relation may be written:

$$\frac{(F_{SH}t)^2}{2M} = E_d \ , \qquad (3)$$

where t is duration of the action of the force in the process of one hit.

In computer experiment we gradually increased the value of the force F_{SH} to the point where the atoms begin to move irreversibly to interstitial positions. This magnitude of the force F_{SH} corresponds to $E_d \approx 25$ - 30 eV. Such procedure allows to define the interval for F_{SH} that corresponds to the selected interval (ε_1, ε_2) and respectively (E_1, E_2) as determined by Eq. (1).

3. Computer simulation of ion bombardment of polyatomic crystals

In the MD method [17-19] the classical equations of motion with an appropriate potential of the interaction between particles are solved. Verlet algorithm [20] is usually used to solve these equations. To simulate the action of the shock function, the total force acting on atom i is presented by expression:

$$\vec{F}_i^{\,n} = -\sum \frac{\partial \Phi(\vec{r}_{ij})}{\partial x_i} + \vec{F}_{SH} \ , \qquad (4)$$

where $\Phi(\vec{r}_{ij})$ is the interatomic potential and $r_{ij} = |\vec{r}_i - \vec{r}_j|$ is the distance between atoms i and j.

To apply the proposed approach we have constructed a model crystal with a cubic lattice that consists of three different types of atoms. This model crystal contained 8000 atoms. Masses of atoms correspond by convention to Si^{28}, Ba^{145} and U^{238}. Further we denote these atoms as m_1, m_2 and m_3, respectively. In Figure 1 a fragment of the model crystal is shown. The occupation of the lattice sites was chosen in such a way that each U or Ba atom has light Si atoms as nearest neighbors and each U (Ba) atom has Ba (U) atoms as second nearest neighbors. In our calculations, Lenard-Jones potential [21] was used with parameters taken from [22-24]. These parameters were slightly varied to stabilize the lattice of the model crystal. The potentials that describe the interaction of atoms of different types were taken as an average of constituent's interatomic potentials. Further simulations were devoted to the study of the influence of the ratio of atomic masses on the radiation effect under the bombardment of model crystal by Nitrogen (N) ions with energy in different energy intervals.

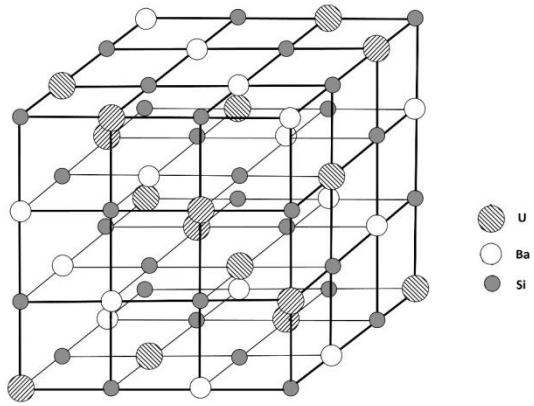

Figure1. Fragment of the model crystal.

In computer experiment the bombardment was performed in two energy (E) intervals: (I) 34 eV – 93 eV and (II) 93 eV – 142 eV determined according to expression (1).

It is clear that the ion bombardment in the energy interval (I) leads to displacement of only Si atoms into the

interstitial positions whereas a bombardment in the interval (II) leads to such displacement of both Si and Ba atoms.

4. Ion bombardment of triatomic crystal. Results and discussion

We started with the consideration of the action of particles in the first energy interval on radiation-induced structural changes in the target crystal. As a result of simulations it was found that the formation of clusters of heavier atoms (U and Ba) occurs. According to our definition, in *heavy clusters* heavy atoms occupy the nearest neighbor positions in the lattice. Such clusters are formed due to radiation-induced formation of vacancies in the sublattice of light atoms (Si). These vacancies stimulate the displacements of heavy atoms. As a result U and Ba move closer to each other forming heavy clusters. Radiation stimulated diffusion of heavy atoms under the bombardment of particles with energy less than the threshold energy of the atomic displacement also promotes the formation of such clusters.

The kinetics of the accumulation of clusters is shown in Figure 2. It may be seen that for the formation of heavy clusters some threshold dose exists. A consideration of atomic configurations in the model crystal showed that at higher doses the formation of three-atomic clusters starts by adding to diatomic clusters (m_2-m_3) one additional atom with the mass m_2.

This explains the saturation of the formation of diatomic (m_2-m_3) clusters demonstrated by the kinetic curve. With the further increase of the dose the accumulation of clusters containing m_2 and m_3 atoms stops and clusters containing only m_3 atoms are formed.

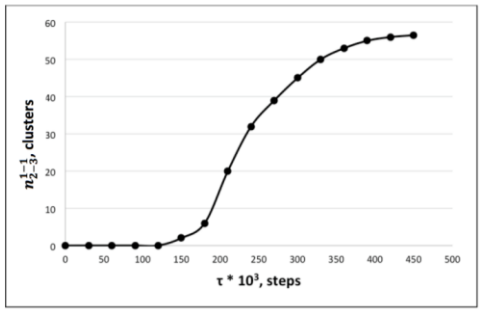

Figure 2. Kinetics of accumulation of m_2-m_3 clusters (the bombardment by ions in the first energy interval). Here n^{1-1}_{2-3} is the number of diatomic clusters formed by one atom with mass m_2 and one atom with mass m_3.

As a result of a bombardment of the model crystal by ions with the energy in the second energy interval we observed a formation of similar diatomic m_2-m_3 clusters which are further destroyed with accumulation of dose. Simultaneously with the destruction of m_2-m_3 clusters the diatomic clusters containing only atoms with the mass m_3 are formed as displayed in Figure 3.

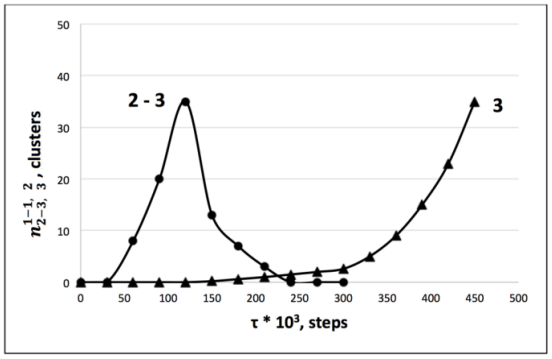

Figure 3. Kinetics of diatomic clusters formed by atoms with the masses m_2 and m_3 and by atoms with the mass m_3 (the bombardment by ions in the second energy interval, 300 hits/step). Here n^{1-1}_{2-3} is the same as in Figure 2, and n^{2}_{3} is the number of diatomic clusters formed by atoms with the mass m_3. Circles and triangles correspond to n^{1-1}_{2-3} and n^{2}_{3}, respectively.

We obtained that the kinetics presented in Figure 3 changes when the intensity of the ion beam is changed. As already mentioned, in our model the intensity of the ion beam is proportional to the number of atoms that get a hit during one calculation step. Figure 4 illustrates the results of formation of similar m_2-m_3 and m_3 diatomic clusters in the case of lower intensity of the ion beam than in the case shown in Figure 3. It can be seen that the formation of m_2-m_3 clusters starts later and their life time is longer. Respectively, the diatomic clusters of the atoms with the mass m_3 form also later.

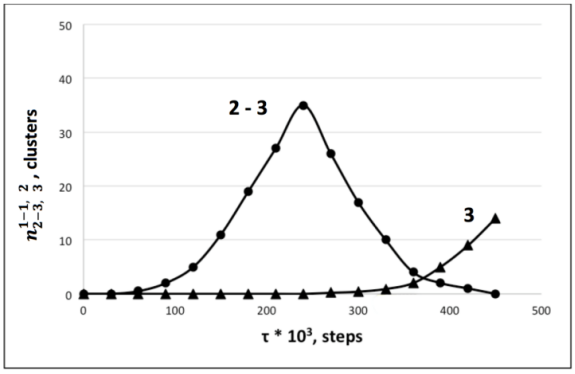

Figure 4. Kinetics of diatomic clusters m_2-m_3 and m_3 (the bombardment by ions in the second energy interval, 200 hits/step) Circles and triangles correspond to n^{1-1}_{2-3} and n^{2}_{3}, respectively.

Figure 5 illustrates that opposite conclusions are derived for the intensity of the ion beam that is in half larger than in the case shown in Fig. 3. As follows from Figure 5 the formation of m_2-m_3 clusters begins almost immediately after the irradiation starts and they are stable for a shorter period of time. Diatomic clusters, consisting of atoms with the

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large mass m_3, start to form much earlier than in the case shown in Fig. 3.

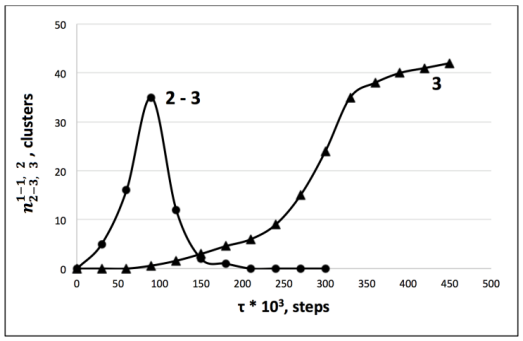

Figure 5. Kinetics of clusters m_2-m_3 and m_3 (the bombardment by ions in the second energy interval, 400 hits/step) Circles and triangles correspond to n_{2-3}^{1-1} and n_3^2, respectively.

Figure 5 shows that at larger dose rate the kinetics of accumulation of diatomic clusters of m_3 atoms tends to saturate. At these doses it was observed that three-atomic and four-atomic clusters of m_3 atoms start to form. It is obvious that they are formed not only by bringing together three or four massive atoms but also by attachment of additional atoms to already existing diatomic clusters of heavy atoms. So, the saturation stage of the kinetics of m_3 clusters may be reasonably explained by transition of diatomic clusters of atoms with the mass m_3 into polyatomic clusters of these atoms. It was mentioned that in the second energy interval (93 eV – 142 eV) the atoms with masses m_1 and m_2 are displaced from the lattice sites. At some dose these clusters are destroyed and two-atomic clusters, consisting only of atoms with the mass m_3, are formed. This is caused due to the ability of bombarding particles (in this energy interval) to displace m_2 atoms. Further we observed the increase of the number of atoms of m_3 type in the heavy clusters.

Figs. 6-8 demonstrate the dependence of the characteristic parameters of the formation and destruction of heavy clusters on the intensity of the ion beam. Fig. 6 depicts the dependence of the threshold dose for m_2-m_3 clusters formation under the ion irradiation in the first and in the second energy intervals on the dose rate of irradiation.

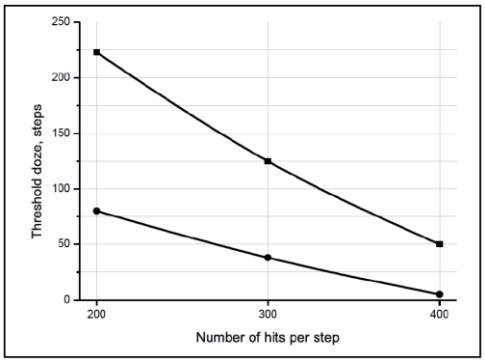

Figure 6. Dependence of the threshold dose for the formation of diatomic m_2-m_3 clusters on the dose rate under irradiation by ions in the first (squares) and second (circles) energy intervals.

The almost linear dependences of threshold doses for heavy clusters formation on the dose rate means that the accumulation of vacancies leads to significant decrease in the bonding of heavy atoms in the lattice. In its turn, this changes the equilibrium positions of heavy atoms in the lattice and leads to the formation of heavy clusters.

Fig 7 shows how the dose at which the m_2-m_3 clusters break up depends on the intensity of the ion beam. Fig. 8 illustrates the dependence of the threshold dose for the formation of diatomic clusters of m_3 atoms under irradiation by ions in the second energy interval on the dose rate. The non-linear dependences on the dose rate of the threshold doses for m_2-m_3 clusters destruction and m_3 clusters formation in this case are explained by the necessity of the preliminary breaking of m_2-m_3 clusters by incident particles.

Analysis of results of triatomic model crystal bombardment by incident particles from the second energy interval allows suggesting the features of the formation of clusters at higher energies. In this case, as a result of elastic collisions, all three types of atoms can be displaced. This facilitates the formation of different heavy clusters. Although simultaneously heavy clusters will collapse, the heaviest clusters will collapse to less extent. Thus, the radiation effect will lead to the predominance of the heaviest clusters.

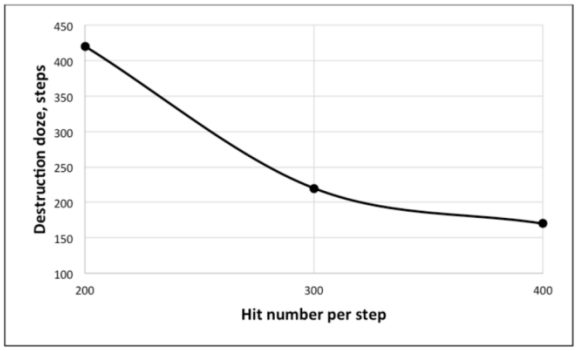

Fig.7. Dependence of the destruction dose on the dose rate for m_2-m_3 clusters under irradiation by ions in the second energy interval.

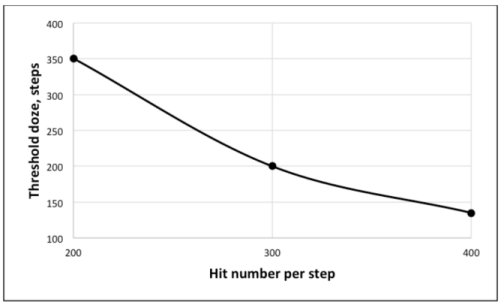

Fig. 8. Dependence of the formation dose on the dose rate for m_3 clusters under irradiation by ions in the second energy interval.

5. Summary

We report the MD approach that allows clarification of the contribution of incident particles in the selected interval of energy spectrum to the final structural damage of the target crystal. Its application for the case of triatomic model crystal is demonstrated. A formation of clusters caused only by atomic elastic collisions is considered. The results obtained for model crystal are of a general nature, and the developed approach can be applied for study of radiation effects in different materials including recently reported U-Al-Si phases [25].

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