Features of primary radiation damage in Fe–Cr alloy near free surfaces

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Abstract. The influence of interfaces on the primary radiation damage in the Fe–10Cr alloy was studied in the framework of the molecular dynamics method. The simulation of atomic displacement cascade evolution near free surfaces with crystallographic indices (110) and (111) was performed. It was revealed that the number of survived point defects sufficiently depends on the region of cascade generation. Atomic displacement cascade near the free surface generates approximately two times more defects than the cascade in the bulk sample with the ideal structure. After the irradiation of the free surfaces the samples contained more vacancies than interstitial atoms. Calculations showed that the crystallographic orientation of the irradiated free surface significantly affects the character of the primary radiation damage of the material. Craters are much more frequently formed at irradiation of the (111) surface. The 1/2⟨111⟩ dislocations are formed mainly at irradiation of the (111) surface, and the ⟨100⟩ dislocations mainly appear at loading the (110) surface.

1. Introduction
The primary radiation damage depends upon the internal structure of the material. Various interfaces such as free surfaces, grain boundaries define the features of the atomic displacement cascade development, the number and type of surviving radiation defects [1–5]. In [1, 2] it was shown that grain boundaries act as barriers to the propagation of atomic displacement cascades and accumulate in their region the largest number of generated defects. The largest-sized clusters of point defects are formed at the final stage of the cascade development in the grain boundaries. The evolution of atomic displacement cascades near the free surface of the metals has its own features due to the possibility of the escape of generated defects to the surface. Typically, the development of such displacement cascades is accompanied by the formation of adsorbed atoms (adatom islands) on the surface of the sample and sputtering a certain number of atoms as a result of collisions between the decay particle and atoms of the crystal lattice [5]. Furthermore, the evolution of near-surface cascades is characterized by the formation of craters on the irradiated surface and the generation of a sufficiently large number of vacancy type dislocation loops [4–9]. One might expect that the free surface effect on the primary radiation damage is the most significant for the atomic displacement cascades of low energy (less than 50 keV), as the evolution of cascades takes place directly in the near-surface region. Therefore, in the present study we carried out computer simulations of the nucleation and evolution of radiation-induced defects generated by the atomic displacement cascades in the Fe–10Cr crystallite near free surfaces with different crystallographic orientations.
Table 1. The number of survived radiation defects after generation of atomic displacement cascades of energy of 20 keV near the (110) and (111) free surfaces and in bulk Fe–10Cr.

| Cascade type          | Number        | Average size, vacancies |
|-----------------------|---------------|-------------------------|
|                       | Vacancies     | SIAs                    | Adatoms | Crater | 1/2 $\langle 111 \rangle$ loop | $\langle 100 \rangle$ loop |
| Bulk cascade          | 54 ± 3        | 54 ± 3                  | —       | —      | —                      | —                      |
| (110) surface cascade | 46 ± 3        | 26 ± 2                  | 176 ± 26| —      | 63 ± 8                | 101 ± 12               |
| (111) surface cascade | 53 ± 3        | 28 ± 2                  | 127 ± 18| 105 ± 16| 113 ± 18         | 75 ± 9                 |

2. Methodology
Evolution of atomic displacement cascades near free surfaces of Fe–10Cr alloy was studied on the basis of the molecular dynamics method [10–12]. Calculations were performed with use of the LAMMPS package [13]. The interaction between atoms was described by the many-body potential constructed according to the concentration-dependent embedded atom method [14]. As in [15, 16] the collision of decay particle with the crystallite was simulated by setting a momentum to the one of surface atoms—the primary knocked atom (PKA). Direction of PKA momentum was perpendicular to the free surface. Periodic boundary conditions were applied in other two directions. The PKA energy was equal to 20 keV. It should be noted that the average PKA energy for iron in the first wall of DEMO reactor is 18.8 keV [17]. Simulated crystallites had the cubic shape with 20 nm edges. The irradiated free surface had indices (110) or (111). The initial temperature of the crystallites was 300 K. Concentration of Cr was 10 at.% which is close to concentrations for majority of steels applied in nuclear power plants [18]. For each irradiated surface 40 calculations with different PKA positions were made. The occupancy of Wigner–Seitz cells was calculated to identify point defects in irradiated crystallites. Structural analysis of extended defects, such as craters and dislocation loops was based on the common neighbor analysis [19] and the dislocation extraction algorithm [20]. Visualization of obtained structures was performed in the OVITO software [21].

3. Results and discussion
Evolution of the atomic displacement cascade in the material is characterized by three stages: ballistic, recombination and primary damage state. The ballistic stage starts with the collision of the decay particle with the lattice atoms and ends when the atomic displacement cascade reaches the maximum size. Then the recombination stage follows, during which the number of radiation-induced defects is reducing due to the annihilation of self-interstitial atoms (SIAs) and vacancies. The primary damage state of a cascade begins with the moment when the number of radiation-induced defects reaches saturation. It fluctuates around some average value, which is associated only with thermal and diffusion processes in the material.

Calculations have shown that for the 20 keV atomic displacement cascade near the free surface the number of survived radiation defects is twice as much as in the case when the cascade with the same energy is generated in the bulk far away from any interfaces. This is due to the fact that in the first case specific structural defects are formed in the crystallites: craters, vacancy type dislocation loops, adatom islands, and, in addition, some of the atoms are sputtered from the free surface. It was found that the number of survived point defects is approximately 250 after the generation of displacement cascade in the surface region (this is without surface defects such as craters and adatom islands). About 110 point defects survive in the case of the cascade generated in the bulk.
Detailed information about the number of defects generated by atomic displacement cascades in the bulk as well as near the (111) and (110) free surfaces is shown in table 1. The table shows that the number of vacancies, not related to the vacancy loops, is roughly the same for cascades near both irradiated surfaces and for cascades in the bulk material. The number of survived SIAs in the surface regions was about two times less than SIA number after cascades generated in bulk. This is due to the escape of SIAs to the free surface and the sputtering of atoms from the irradiated surface on the ballistic stage of cascade development. The number of adatoms depends on the orientation of the irradiated surface and for the (110) surface it is somewhat higher than for the (111) surface.

Calculations showed that the generation of cascades for both irradiated surfaces leads to the formation of sufficiently large vacancy loops with the $1/2 \langle 111 \rangle$ and $\langle 100 \rangle$ Burgers vector. It is notable that $\langle 100 \rangle$ vacancy loops are mainly formed after the (110) surface irradiation, and $1/2 \langle 111 \rangle$ vacancy loops after the (111) surface exposure. We determined the loop size as the number of vacancies composing it. As shown in table 1, the average size of the two types of vacancy loops depends on the orientation of the irradiated surface. Atomic displacement cascades formed in the bulk do not generate vacancy loops (see table 1). It should be noted that irradiation of the (111) surface generally leads to the formation of craters on it. With regard to the (110) surface, the cases of crater formation upon it are considerably less. The frequency of the formation of craters is connected with the anisotropy of propagation of shock waves.
waves generated by atomic displacement cascades near free surface at the ballistic stage. The waves propagating along (111) close-packed directions have the higher amplitude than waves moving in (110) directions [22], so generation of crater on the (111) surface is more preferable.

The results obtained are in good agreement with theoretical [4,5] and experimental [6–9] data on the formation of craters and dislocation loops in the surface region after irradiation. It was shown experimentally that under Xe$^+$ irradiation of Ag, Au, In, Pb the number of craters per ion is larger for denser material [6]. 1/2 $\langle 111 \rangle$ and (100) vacancy loops were found in experiments on Fe and Fe-Cr irradiation by Fe$^+$ and Xe$^+$ ions of energies 100–150 keV [9]. The same types of dislocation loops were formed in molecular dynamics simulations of atomic displacement cascades near the (100) and (110) free surfaces in Fe [4,5].

Features of the atomic displacement cascade evolution near the (111) free surface are shown in figure 1. The figure 1(a) shows that cascade splits into five subcascades at the ballistic stage. Red color indicates displaced atoms that are in the region of the main subcascade, which is characterized by the greatest kinetic energy ($\sim$ 50% of the kinetic energy of the whole cascade). As a consequence, the longer time is needed for relaxation of the radiation-damaged region formed by the main subcascade, see figure 1(b). The largest-sized vacancy loop is formed in this region at the end of the recombination stage, see figure 1(c). One of the subcascades at the end of the recombination stage forms considerably smaller vacancy loop which is shown on the right in figure 1(c).

Characteristic differences in the primary radiation damage of the material structure after generation of atomic displacement cascades near the (110) and (111) free surfaces are shown in figure 2. The figure clearly shows the crater formed on the (111) surface and vacancy loops.

Analysis of the simulation results shows that there is a correlation between the size of vacancy loops and the number of adatoms. Figure 3 shows that the number of adatoms on the free surface is approximately equal to the size of vacancy loops for the majority of the calculations. Deviations from this ratio are associated with a small number of survived point defects and their
clusters in the bulk. Note that the number of adatoms on the (110) surface is slightly higher than the number of vacancies in the loop. For the (111) surface the loop size often exceeds the number of adatoms. Moreover a sufficiently large number of loops underneath the (111) surface escaped to the free surface during relaxation of the crystallite.

It is found that the size of the crater formed on the free surface correlates with the size of the surviving vacancy loop. The larger crater on the free surface, the smaller the size of the surviving vacancy loop. It can be concluded from the comparison of various calculations that the size of vacancy loops increases with increasing kinetic energy of the main subcascade.

Free surfaces are efficient sinks for point defects and their clusters. Therefore, the number of surviving SIA clusters for (110) and (111) irradiated surfaces is smaller than in the case of atomic displacement cascade generated in the bulk, see figure 4(a). The figure also shows that the clusters of the largest size are formed by the atomic displacement cascade in the bulk.
Clusters with the size up to 17 SIAs can survive for cascades in the bulk, almost twice the size of SIA clusters which survived in the vicinity of the free surfaces. Note that the number of SIA clusters is almost always larger for the (111) surface than the (110) surface. Number of single dumbbells is the same for the two surfaces. In view of the low mobility of vacancies the number of survived vacancies and their clusters of size $<13$ is approximately the same for bulk and near-surface cascades. However, the largest vacancy clusters (except dislocation loops) are formed in the bulk, see figure 4(b).

4. Conclusion
It was found that the number of survived radiation defects after generation of atomic displacement cascades near the free surface exceeds two times their number after cascade generation in the bulk material far away from the interfaces. This is because in the first case some of the atoms were sputtered from the surface region and the specific structural defects (craters on the free surface, dislocation loops of vacancy type in the surface region) were formed after sample relaxation. The crystallographic orientation of the irradiated surfaces has a significant influence on features of the primary radiation damage of the material. So, craters are much more frequently formed at irradiation of the (111) surface, than of the (110) one. The $\langle 111 \rangle$ dislocations are formed mainly at irradiation of the (111) surface, and the $\langle 100 \rangle$ dislocations mainly appear at loading the (110) surface. There is a correlation between the size of the vacancy loops and the number of adatoms on the free surface. The size of the vacancy loops exceeds the number of adatoms in the case of the (111) surface, and the inverse relationship is observed for the case of (110) surface. A significantly higher number of vacancy loops from the radiation-damaged region escaped to the (111) free surface than to the (110) surface.

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