Journal of Nuclear Materials 385 (2009) 83–87

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

A hybrid model of primary radiation damage in crystals

S.I. Samarin *, V.V. Dremov

Russian Federal Nuclear Center – Institute of Technical Physics, 13, Vasiliev Street, Snezhinsk, 456770 Chelyabinsk Region, Russia

A B S T R A C T

The paper offers a hybrid model which combines molecular dynamics and Monte Carlo (MD+MC) methods to describe primary radiation damage in crystals, caused by particles whose energies are no higher than several tens of keV. The particles are tracked in accord with equations of motion with account for pair interaction. The model also considers particle interaction with the mean-field potential (MFP) of the crystal. Only particles involved in cascading are tracked. Equations of motion for these particles include dissipative forces which describe energy exchange between cascade particles and electrons. New particles – the atoms of the crystal in the cascade region – have stochastic parameters (phase coordinates); they are sampled by the Monte Carlo method from the distribution that describes the classic canonical ensemble of non-interacting particles subjected to the external MFP. The introduction of particle interaction with the MFP helps avoid difficulties related to crystal stability and the choice of an adequate interparticle interaction potential in the traditional MD methods. Our technique is many times as fast as the traditional MD methods because we consider only particles which are involved in cascading and apply special methods to speedup the calculation of forces by accounting for the short-range pair potential used.

1. Introduction

Primary radiation effects in crystalline material are usually analyzed using Monte Carlo models implemented, for example, in such codes as MARLOW [1,2] and TRIM-91 [3]. Many Monte Carlo models rely upon the assumption that interactions are binary. Up to now, there has not been given an unambiguous answer to the question from what energies particle interaction can be considered binary, i.e., simultaneous interaction with several atoms from the nearest surrounding can be neglected. However, multi-body interaction may be essential, causing specific trajectories where moving particles have impact parameters close to those of several atoms from the atomic rows in the crystal. The authors of [2] used an improved binary model where kinematics of scattering allowed for inelastic energy losses and the procedure of choosing the partner for interaction was significantly improved to rule out repeated interactions with one and the same atom and to allow simultaneous interaction with several atoms. Such improvements allow the description of particle motion in the crystal channel, but underestimate energy losses in contrast with the dynamic models [4]. In Monte Carlo techniques, the new direction of the scattered particle and the recoil atom are calculated using the asymptotic behavior of scattering regardless specific surrounding. In reality, the atom always moves in a force field of surrounding atoms and its trajectory differs from that one constructed under the assumptions of local and binary interaction. In the study of Pu^{239}, self-irradiation where most damage falls on a U^{235} atom which is produced in the α-decay of Pu^{239} with an initial energy of $\sim$86 keV, the approximations used in Monte Carlo techniques may essentially distort the actual particle trajectory because its radius of curvature is comparable with the distance between atoms in lattice nodes.

With the above shortcomings of MC techniques borne in mind, it is safe to say that semi-dynamic and dynamic models fit better the simulation of cascades initiated by low-energy particles. The authors of [4] give a hierarchy of cascading models and suggest that dynamic models are on the top in the hierarchy of models describing primary radiation effects in crystals. In their view, the use of dynamic models for the simulation of the low-energy phase is absolutely indispensable because only these models can reliably track the chains of focused collisions and replacements that essentially contribute to mass and energy transfer in crystal.

Dynamic models have long been used to model cascades, but they are rather time-consuming which results in their uses mainly for low-energy particles (several tens of eV). In case of high-energy particles, the simulation of one cascade requires a very long time, not to speak of a number of cascades that would be sufficient to evaluate cascade statistics.

Another problem of dynamic models is the selection of an appropriate potential. It is difficult to find such a potential that would ensure crystallite stability with no disturbance of the lattice structure. As a rule, cascading is modeled using central interatomic

* Corresponding author. Tel.: +7 3514654730; fax: +7 3514655118.
E-mail address: s.i.samarin@vniitf.ru (S.I. Samarin).

0022-3115/$ - see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2008.09.054

potentials, i.e., $U(r_{ij}) = U(|\vec{r}_i - \vec{r}_j|)$, though this assumption does not hold for covalent crystals [4].

The above shortcomings of MC and MD models make it necessary to develop a semi-dynamic technique which would help reduce the time required for modeling primary radiation damage. The technique must be free of all drawbacks peculiar to MC models. It must provide a rather high speed of calculation and the free choice of potentials without disturbing crystal stability.

2. Basic assumptions

The technique proposed is based on the molecular dynamics method where the particle trajectory is constructed from the solution of the equation of motion for interacting particles subjected to the MFP. Considered are only particles involved in the cascade, i.e., the particles whose distance to cascade and excited particles is less than a specified interaction range. Cascade particles are the particles whose total energy is positive (free particles). Excited particles are the bonded particles (with a negative total energy) near the lattice node, whose energy is higher than a specified value. Equations of motion for these particles are solved with account for dissipative forces describing the electronic energy loss. When a new particle enters the region of already modeled particles, its parameters $(\vec{r}, \vec{v})$ are sampled from a distribution which describes the classic canonical ensemble of non-interacting particles subjected to the MFP:

$$f(\vec{r}, \vec{v}) = \frac{1}{Z} \cdot \exp\left(-\frac{m \cdot v^2/2 + U_{MFP}(\vec{r})}{kT_{cr}}\right), \tag{1}$$

where $\vec{r}$ is distance to the current lattice node. It has been shown in [5] that the classic description can be applied to most crystals in a wide temperature range. The MFP is a 'binding' potential in the form of non-overlapping spherical potential wells:

$$U_{MFP}(\vec{r}) = \begin{cases} \sum\limits_{\vec{n},\ell} \frac{\chi}{2} \cdot (\vec{r} - \vec{R}_{\vec{n},\ell})^2 - \varepsilon_{\max}, & |\vec{r} - \vec{R}_{\vec{n},\ell}| < R_{\max}, \\ 0, & |\vec{r} - \vec{R}_{\vec{n},\ell}| \geqslant R_{\max}, \end{cases} \tag{2}$$

where $\varepsilon_{\max}$ is the depth of the potential well; $\vec{R}_{\vec{n},\ell} = \vec{\kappa}_\ell + n_1 \cdot \vec{e}_1 + n_2 \cdot \vec{e}_2 + n_3 \cdot \vec{e}_3$ is the node position vector defined by the position vectors $\vec{\kappa}_\ell$, the basis vectors $\vec{e}_1, \vec{e}_2, \vec{e}_3$ and the integer vector $\vec{n} \equiv (n_1, n_2, n_3)$; χ is a dimension coefficient defined below; $R_{\max} = \sqrt{\frac{2 \cdot \varepsilon_{\max}}{\chi}}$ is the maximum effective range of the potential for the current node. The MFP defines the structure of the crystal, describes thermal displacements of atoms in the node and reproduces the average energy that is needed to displace an atom. The MFP must be binding and negative. The particles whose total energy is negative are assumed to be bonded (or excited particles in the cascade) and those whose total energy is positive are free (or cascade particles). Moreover, particles interact with each other by force of the repulsive pair potential describing the process of scattering:

$$U(r_{ij}) = \frac{Z_1 \cdot Z_2 \cdot e^2}{r_{ij}} \cdot \Phi\left(\frac{r_{ij}}{a}\right), \tag{3}$$

where a and $\Phi(\frac{r_{ij}}{a})$ are, respectively, a screening parameter and a screening function defined as

$$\Phi\left(\frac{r_{ij}}{a}\right) = \sum_{k=1}^{n} b_k \cdot \exp\left(-c_k \cdot \frac{r_{ij}}{a}\right), \tag{4}$$

$$a = \frac{0.88534 \cdot a_0}{\sqrt{(Z_1^{2/3} + Z_2^{2/3})}} \text{[6]},$$

$a_0 = 0.529$ Å is the first Bohr orbit, Z_1 and Z_2 are charges (atomic numbers) of interacting particles, $r_{ij} = |\vec{r}_i - \vec{r}_j|$ is distance between interacting particles. The model allows using an arbitrary potential of the form (3) by defining appropriate coefficients of the screening function, including Moliere, universal and other potentials [4]. At each time step, we find candidate particles from among the particles not yet involved in modeling for each modeled particle whose energy is higher than a specified value by searching the nearest nodes to its current position. After finding all candidates, we check whether the node has been initiated or not. If yes, the candidate particle is rejected. The model neglects structural changes resulted from the displacement of atoms from lattice nodes which apparently influences the MFP. It does not allow for changes in the basic parameters such as the mean-square displacement and the energy of atom displacement from the lattice node caused by the disturbed lattice spacing and the heating of the crystal. Thus MFP parameters do not change in the process of cascade formation. The same shortcomings are present in all MC techniques used to model cascading. So, the detailed modeling of cascade evolution requires that its fast stage be simulated with MD techniques. Our MD+MC technique is capable of describing, in some measure, the effects which can be hardly reproduced in the traditional MC methods, specifically, recovery of atomic displacements as some cascade particles 'settle down', the chain of collisions along crystallographic directions, the chain of replacements which essentially contribute to mass and energy transfer, and the cooling of the cascade region due to the electronic energy loss.

3. Simulation of atomic cascades caused by δ-plutonium self-irradiation

As mentioned above, to construct the mean-field potential used in the proposed method we need to know such parameters as the average energy required to displace an atom from the lattice node, $\langle E_{\text{dis}} \rangle$, and the mean-square thermal displacement of atoms $\langle u^2 \rangle$, and data on the crystalline structure. Data on the mean-square thermal displacement and the crystalline structure of plutonium (α, β, γ and δ phases) can be obtained experimentally from X-ray and neutron diffraction and from photon spectra obtained in inelastic neutron scattering experiments [11–13]. Values of $\langle E_{\text{dis}} \rangle$ for plutonium have not yet been measured reliably, but the author of [7] proposes an estimate derived from the empiric dependence $\langle E_{\text{dis}} \rangle \cong 175 \cdot k \cdot T_m$[K], where T_{melt} is the temperature of melting which equals 913 K for δ-plutonium. With this temperature we can estimate that the average displacement energy roughly equals 14 eV. Another value has recently been obtained in MD simulations with the potential [8] whose parameters were taken to be such as to fit the macroscopic characteristics of the crystal. This value equals 10 eV. Both the values were used in calculations.

Another important issue in cascade modeling is the description of the electronic energy loss which is of importance for not only our method, but for all methods that model radiation damage in material. Up to now, there has not been proposed a model which would be simple and accurately describe the electronic energy loss. In most cases, energy dissipation for low-energy cascade particles is described using the Lindhard–Scharff model, often with a correction factor [14] which is defined from correspondence between an integral parameter, say range, measured and calculated. Here, we have two parameters that can be varied: the average displacement energy and the correction factor of the Lindhard–Scharff stopping power. For the former, we have only two values recommended. Therefore, the correction factor is selected in accord with the average cascade size (or the average number of Frenkel pairs) in calculations. For $\langle E_{\text{dis}} \rangle = 10$ eV, the correction factor must be no smaller than three to ensure that the estimated cascade size is close to that in [7]: ~7.5 nm. In this case, the number of Frenkel pairs (~2000) roughly agrees with the estimate provided in [7], but the electronic

energy loss we obtain, specifically ~47% of the initial energy of the initiating particle, markedly differs from ~25%. Moreover, this factor does not ensure agreement with the coefficient of the dissipative force obtained by the Hakkinen model [10]; its estimate is provided in [15]. For $\langle E_{dis} \rangle = 14$ eV, the correction factor must be ~1. In this case, all calculated parameters are close to estimates obtained in [7]: an average cascade size of ~8.4 nm; ~2000 Frenkel pairs; and an electronic energy loss of ~30%. All the above allows us to simulate cascading with $\langle E_{dis} \rangle = 14$ eV until reliable experimental data are available. However, such parameters as the average displacement energy and the Lindhard–Scharff coefficient [9] need to be taken consistently with the pair potential used. In all calculations presented here, we used the Moliere potential (3) and (4).

3.1. Evolution of atomic cascades from δ-plutonium self-irradiation

Simulation results suggest that the entire process of cascade formation can be divided into a number of stages.

An active cascading stage involves collisions in which large portions of energy (about several keV) are transferred to recoil atoms which become rather energetic and leave their positions in lattice nodes (see Fig. 1). The reason for this stage is as follows. An initiating particle (U^{235} which is a Pu^{239} decay product) starts its motion from the node of the lattice slightly distorted by thermal oscillations in any direction (all directions are equiprobable). Since the particle has a high-energy, it inevitably reaches a scatterer atom with small impact parameters causing large energy redistribution between the scattered particle and the scatterer. The particle approaches the scatterer atom rather closely and after scattering, not one but two particles behave as initiating, but with lower energies. This process continues until the energy of the particle decreases to such a low value that the minimal approach distance between atoms becomes comparable with the half-distance between lattice nodes (~1–10 keV). Fast cascade particles intensively lose energy to recoil atoms and electrons. The number of particles involved in the cascade rapidly grows and the cascade region extends fast. This stage lasts for ~0.05–0.1 ps. The energy of cascade particles is $\sim 10^3$–10^4 eV on average. Then the cascade gradually enters the next stage.

A channeling stage is characterized by particle motion along crystallographic directions (channels) which causes a lot of displacements with small energy losses to recoil atoms and hence the slow energy loss by channeling particles (Fig. 2). Atoms displaced by a channeling particle also move along channels causing further displacements with yet smaller energy losses. The cascade gives rise to a great number of sub-cascades which often overlap. In this stage, high-energy cascade particles lose energy slower than in the stage of active cascading. The number of particles involved in the cascade also increases slower. The cascade region extends slower, but the number of displacements (Frenkel pairs) is rather high. The stage continues to ~0.3–0.4 ps; the energy of cascade particles is ~10–10^3 eV on average.

A replacement stage is characterized by a much slower growth of the cascade and the lower electronic energy loss. The number of cascade particles decreases due to annihilation of Frenkel pairs. The stage is featured by chains of atomic replacements (Fig. 3). The number of particles in the cascade grows due to mainly phonon excitation (with no production of Frenkel pairs). The stage exhibits effects of collision chains along crystallographic axes with no atom displacements, which usually terminate with Frenkel pair annihilation. Sub-cascades often overlap. The average energy of cascade particles decreases to several eV. Frenkel pairs annihilate when a cascade particle near a vacancy site transfers a portion of its energy to a neighbor atom and occupies the site. The neighbor atom remains excited, but does not create a vacancy. The process which occurs more often is the replacement of an atom in the lattice node by a cascade particle. One Frenkel pair annihilates to produce another, causing single replacements or replacement chains (Fig. 3A). The chains terminate with either annihilation of the Frenkel pair or birth of an interstitial atom. Cascade modeling to long times with the proposed method may distort processes in the deformed crystal because the model assumes that the crystalline lattice is perfect, while cascading disturbs lattice spacing and form which is not accounted for (except for the thermal deformation of the lattice). We can recommend that the proposed model be used to follow cascading to about 0.5–1 ps. As a rule, by these times the maximum energy of particles in the cascade decreases to ~20 eV, and further modeling can be done with the traditional molecular dynamics models.

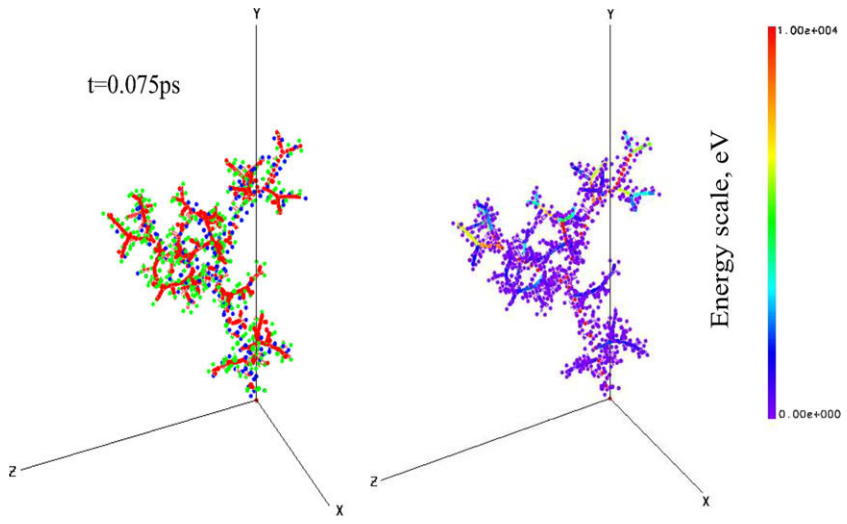

Fig. 1. The initial stage of cascading (to t = 0.075 ps) left: the red dots show the tracks of cascade particles and the blue and green ones show the tracks of weakly and strongly excited particles in lattice nodes. Right: particle tracks are painted with respect to their kinetic energy. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

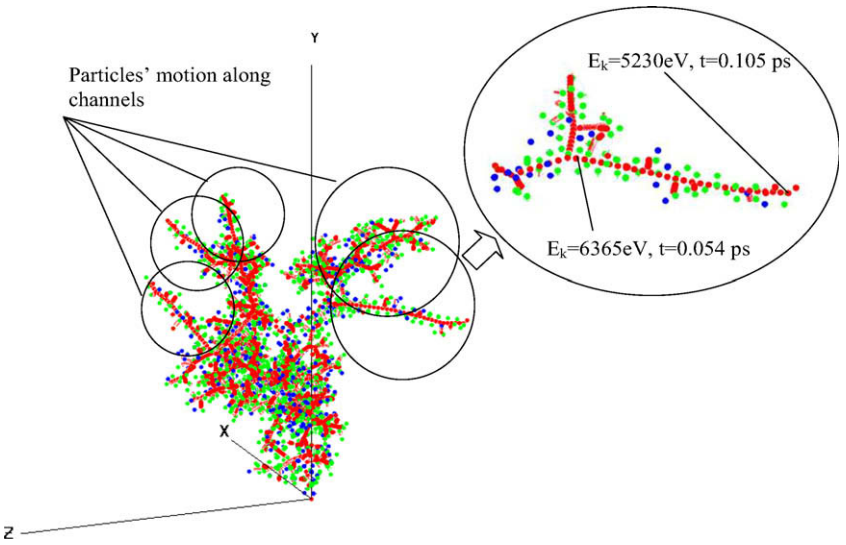

Fig. 2. The initial stage of channeling: particle tracks along channels. Circled are spectacular tracks of channeling particles. One of them is zoomed on the right. The kinetic energy of this particle is ~6365 eV at t = 0.054 ps and ~5230 eV at t = 0.105 ps.

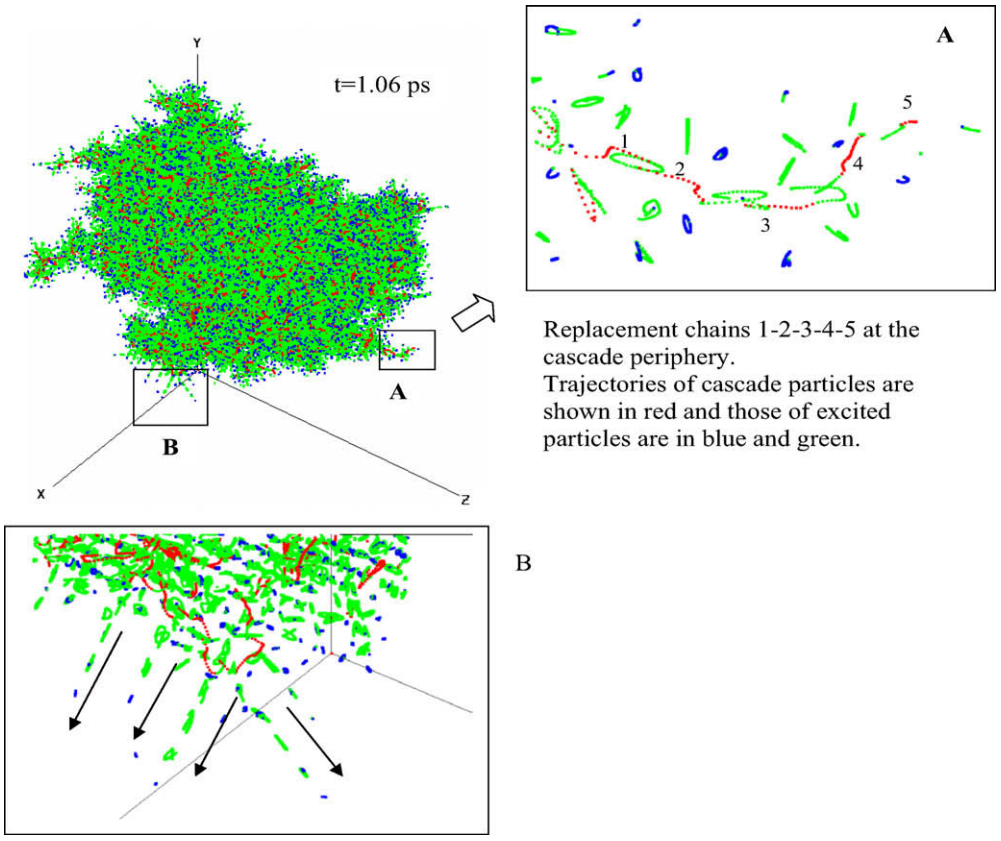

Fig. 3. Cascade tracks at the replacement stage; t = 1.06 ps. (A) A spectacular replacement chain; (B) tracks of collision chains (arrows) without displacement of atoms from lattice nodes.

3.2. Statistics of atomic cascades from plutonium self-irradiation

Cascading is a statistical process whose characteristics, such as the size of the damage area, the average energy of cascade particles and the number of cascade particles or Frenkel pairs, may strongly depend on the direction of the initiating particle. It is necessary to estimate cascade characteristics by averaging over a large number of cascades. The code MANDRAGOR was developed just for this purpose. It was used to obtain a number of characteristics which could help attain a better understanding of cascading processes. The characteristics averaged with the proposed model can be used to investigate structural changes caused by self-irradiation with respect to different alloying additions, temperature, pressure and other parameters. MANDRAGOR calculations show the current number of Frenkel pairs to be much smaller than the total number of replacements. So, the number of Frenkel pairs in δ-Pu is ~1000, while the total number of displacements produced in the cascade during the same time is ~2400. In calculations by the code TRCR2

Table 1
Total number of Frenkel pairs from U^{235} produced in the α-decay of Pu^{239}.

	δ-Pu	δ-Pu + Ga (5%)
TRCR2 [20]	2590	2534
MANDRAGOR	2440	2244
Wolfer [8]	2290	–

[15] in the same setup, the total number of displacements is larger than that obtained by MANDRAGOR. This may result from the fact that TRCR2 does not account for changes in the crystallite state which were induced by the cascade earlier. In other words, TRCR2 does not allow sub-cascades to overlap which leads to slightly overestimated total displacements. Table 1 contains the total number of Frenkel pairs produced by the cascade.

4. Conclusion

The paper proposes a technique to model cascades initiated by the recoil atom produced in charged-particle emission decay. Its capabilities are demonstrated through the simulation of atomic cascades in δ-plutonium, produced in the α-decay of Pu^{239}. Cascading processes are analyzed with respect to the energy of particles in the cascade. The technique has been shown to give a better description of low-energy cascade characteristics than that used in [15]. The technique allows an easy exchange of data on the modeled cascade with other MD codes for further modeling of primary radiation damage effects, making it possible to simulate self-irradiation-induced micro-scale processes in one run. The technique also gives statistical estimates which can be used to investigate structural changes from self-irradiation with respect to alloying additions, temperature, pressure and other parameters.

Acknowledgement

This work was supported in part and implemented under Contract 04783-000-99-35 (VNIITF–LANL), Task Order 009.

References

[1] M.T. Robinson, O.S. Oen, Appl. Phys. Lett. 2 (1963) 30.
[2] M.T. Robinson, I.M. Torrens, Phys. Rev. B 9 (1974) 5008.
[3] J.F. Ziegler, TRIM-91 User Manual, IBM-Research, 28-0 Yorktown, New York, 10598 (1991).
[4] A.F. Akkerman, Simulation of Charged Particles in Matter, Energoatomizdat Publishers, Moscow, 1991.
[5] V.V. Dremov, F.A. Sapozhnikov, M.S. Smirnova, Molecular dynamics investigation into elastic–plastic properties of solids, in: Fifth International Symposium High Dynamic Pressures, vol. I.P.331–340, 2003.
[6] O.B. Firsov, J. Exp. Theor. Phys. 33 (9) (1957) 698–699.
[7] W.G. Wolfer, Radiation effects in plutonium, Fundamental Plutonium Properties, VNIIEF, Sarov, 2003.
[8] A. Kubota, W.G. Wolfer, in: Proceedings of the V international workshop 'Fundamental Plutonium Properties', Snezhinsk, 12–16 September 2005.
[9] J. Lindhard, Sharff, Phys. Rev. 124 (1961) 128.
[10] H. Hakkinen, U. Landman, Phys. Rev. Lett. 71 (1993) 1023.
[11] B.I. Bennet, A.C. Lawson, B. Martinez, J.W. Richardson, J.A. Roberts, Los Alamos Sci. N 26 (2000) 142.
[12] A.C. Lawson, B. Martinez, J.A. Roberts, B.I. Bennet, J.W. Richardson Jr., Philos. Mag. B 80 (2000) 53.
[13] A.C. Lawson, A. Goldstone, B. Cort, R.I. Sheldon, E.M. Foltyn, J. Alloys Compd. 213–214 (1994) 426. tab1.
[14] J.P. Biersack, L.G. Haggmark, A Monte-Carlo computer program for the transport of energetic amorphous targets, Nucl. Instrum. Meth. 174 (1980) 257–269.
[15] V.V. Dremov, F.A. Sapozhnikov, S.I. Samarin, D.G. Modestov, N.E. Chizhkova, J. Alloys Compd. 444–445 (2007) 197–201.