Available online at www.sciencedirect.com
ScienceDirect
Journal of Nuclear Materials 362 (2007) 293–299
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Influence of self-interstitial mobility on damage accumulation in zirconium under fission irradiation conditions

C. Arevalo [a], M.J. Caturla [b,*], J.M. Perlado [a]

[a] *Instituto de Fusión Nuclear, ETSI Industriales, Madrid, Spain*
[b] *Departamento de Física Aplicada, Universidad de Alicante, Alicante E-03690, Spain*

Abstract

Irradiation of zirconium is studied using a kinetic Monte Carlo model. The initial cascade damage produced by 25 keV recoils at a temperature of 600 K obtained from molecular dynamics simulations is used in the calculations. The evolution of the microstructure under fission irradiation conditions has been followed for a number of displacements per atom (dpa) up to 0.5. In particular, the influence of self-interstitial cluster migration on the total defect concentration and size is analyzed. Results are compared with available experimental data.

PACS: 61.72.−y; 61.80.Az; 61.80.Hg; 61.72.Ji

1. Introduction

Advanced Zr alloys are proposed to be used for high burnup applications in current light water reactors as well as for cladding and internal components in future Generation IV reactors. Consequently, understanding the effect of irradiation on the mechanical properties of these alloys is crucial for safe operation of these reactors. However, knowledge of basic irradiation effects in Zr is very scarce in particular at a microscopic level. This work focuses on the description of microscopic effects of irradiation using computer simulations in a multiscale approach.

Molecular dynamics (MD) simulations performed by Voskoboinikov and Bacon [1,2] have provided information about the damage produced by displacement cascades in α-Zr with recoil energies up to 25 keV and different temperatures. These calculations show that point defect clusters created in simulations of displacement cascades in zirconium can be classified in three typical self-interstitial atoms (SIA) categories: SIA dislocation loops with Burgers vector 1/3 ⟨1 1 2 0⟩, triangular arrangement of SIAs within one basal plane, and three-dimensional irregular. Only the first one of these SIA-clusters is glissile while the other two types are sessile. There are also three typical vacancy categories, all of them sessile: triangular prism shape, pyramidal shape and the third type is best described as two components joined together.

On the other hand, MD simulations have also been used to study the mobility of defects in α-Zr

* Corresponding author. Tel.: +34 965903400x2056; fax: +34 965909726.
E-mail address: mj.caturla@ua.es (M.J. Caturla).

0022-3115/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2007.01.079

[3]. These simulations show that single interstitials move in one dimension in the basal plane. Some of the self-interstitial clusters also move in one dimension while others remain immobile. Monovacancies move in three dimensions in the crystal and vacancy clusters are immobile. The mobility of these self-interstitial clusters is a key parameter for developing damage evolution models in metals through either rate theory or kinetic Monte Carlo simulations. In particular, the work of Soneda [4] and of Domain et al. [5] in Fe have shown that the mobility of these clusters is crucial to be able to reproduce the experimental measurements in this material. However, there are still many unknowns mostly regarding the effect of impurities.

The damage accumulation in h.c.p. Zr under irradiation has been studied using a kinetic Monte Carlo (kMC) approach with information provided by MD calculations. We have considered fission reactor conditions, that is, damage (dpa) rate of $10^{-6}\,s^{-1}$ and a temperature of 600 K. The defect concentration as a function of displacements per atom as well as the cluster size distributions has been obtained from these calculations. In particular the influence of self-interstitial cluster migration on the final defect concentration and size is analyzed.

2. Simulation model

An object kinetic Monte Carlo model is used to study the evolution in time of those defects produced during irradiation. The database of displacement cascades produced by Bacon et al. [1,2] using MD is used as initial defect configuration. Cascades were produced by recoils from 10 to 25 keV in steps of 5 keV at 600 K. Information about migration and binding energies of different cluster types and sizes have also been obtained using MD [6,7] and used as input in the kMC [8]. In this model defects are allowed to execute random diffusion jumps (in one, two or three dimensions depending on the nature of the defect) with a probability proportional to their diffusivity. During the simulation, various kinetic processes (events) are allowed to take place: dissociation of a particle from a cluster, diffusive jump of a particle, recombination, association of two defects of the same type, annihilation of a defect at a sink, trapping or de-trapping of a defect at an impurity and introduction of a new cascade.

In order to understand the behavior of the microstructure for Zr, the study initially focused on the evolution of single cascades, and the diffusion and stability of clusters for long times (h), as well as the possibility of formation of clusters. The number of defects escaping recombination was obtained from these calculations, and therefore the number of defects that would interact with the microstructure [9]. These cascade aging simulations followed the same methodology than previous work for Fe [4]. Our simulations showed that dissolution of small vacancy clusters at 600 K is expected as well as migration of single vacancies. 1D migration of interstitials results in small recombination between interstitials and vacancies. The number of vacancies that move to the surface increases when PKA energy decreases because of vacancy clusters are small at low energy and dissolve. However, the number of interstitials that diffuse to the surface is constant with PKA energy.

Using 25 keV cascades, the evolution of the microstructure during irradiation under fission environment conditions has been studied. Periodic boundary conditions were used in this case. The size of the computational box was $100 \times 100 \times 100\ nm^3$. A database of 25 different cascades placed in random locations in the simulation box was repeated in order to simulate the damage accumulation until the total dose was reached. One-dimensional (1D) migration was considered for single interstitial atoms (SIAs) and mobile interstitial clusters, while three-dimensional (3D) motion was used for monovacancies. Vacancy clusters and several interstitial clusters (depending on the initial cascade configuration) are assumed to be immobile. In order to improve the simulation time a grain boundary approximation has been introduced: grain size of 1 μm. That is, if one defect moving in 1D makes 10^6 jumps without finding another one it disappears in a sink.

3. Results

Fig. 1 shows the concentration of visible clusters as a function of displacements per atom for vacancies (triangles) and for self-interstitials (circles). Those clusters that have more than 50 defects, which correspond to loops of about 2 nm, are considered as visible under TEM. In this way, the results of the simulations can be compared to experimental measurements as done earlier in [5,10]. At this temperature, 600 K, the concentration of visible interstitial clusters is very low, approximately $1 \times 10^{15}\ cm^{-3}$ for a dpa dose of 0.5, while the concentration of visible vacancy clusters is three orders of magnitude higher. Therefore, under these condi-

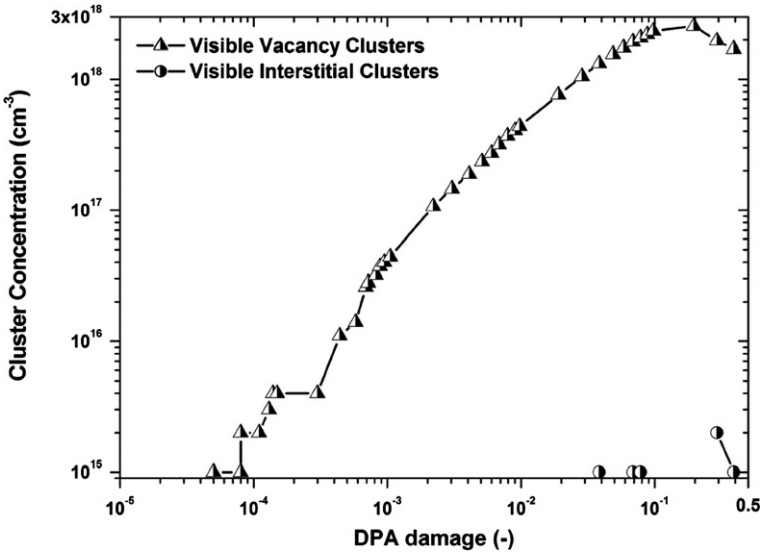

Fig. 1. Visible concentration of vacancies and self-interstitials as a function of dpa damage for a temperature of 600 K. All self-interstitial clusters are mobile. Clusters with more than 50 defects, equivalent to loops of 2 nm radius, are considered visible.

tions mostly vacancy clusters would be observed under the microscope. The reason for this difference is mostly the one-dimensional migration of self-interstitials. This results in very low recombination between self-interstitials and vacancies, as well as low recombination between self-interstitial clusters. Moreover, many of these SIA-clusters migrate to sinks and disappear, without encountering any other defects in their path.

At a dpa damage of 0.1 the concentration of defects seems to saturate. At this point there is cascade overlap such that when a new cascade is introduced, its defects and clusters can recombine with defects in the bulk or can help already existing clusters to grow, reaching equilibrium in the total concentration.

Fig. 2 shows the average cluster size for vacancies (triangles) and self-interstitials (squares) as a

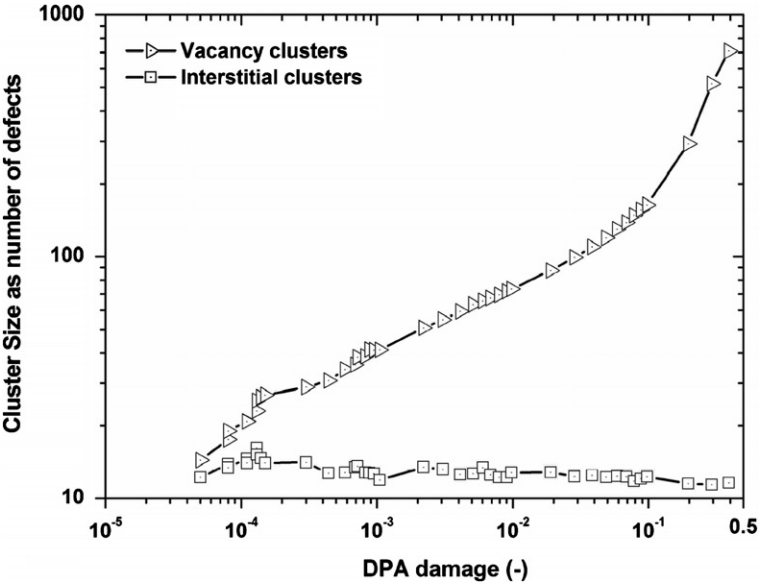

Fig. 2. Average cluster size of vacancies and self-interstitials as a function of dpa damage for 600 K irradiation temperature and all self-interstitial clusters mobile.

function of dpa. The average cluster size for interstitials is independent of dose, due to their stability and the lack of interaction between self-interstitials. Single interstitials and mobile interstitial clusters disappear at sinks without interacting with other self-interstitials, therefore these clusters do not grow during irradiation under these conditions. However, an increase in the average cluster size for vacancies with dpa is observed. This is due to the three-dimensional migration of single vacancies that increases the probability of recombination with other defects, and also due to the dissolution of small vacancy clusters at this temperature. When saturation is reached the average vacancy cluster size presents an increase because of overlap with previous vacancy clusters.

Fig. 3 presents the cluster size distribution for vacancies at a dpa of 0.01, before saturation, and at 0.1 when saturation occurs. This plot shows that the number of clusters of each size increases with dose, but also vacancy clusters move towards larger cluster sizes. The dissolution of small vacancy clusters, single vacancy migration and low recombination with self-interstitials produces this effect.

In the case of interstitials, as it can be seen in Fig. 4, the number of clusters for each size increases with dpa but the cluster size distribution is almost constant. The maximum cluster size at a dpa dose of 0.01 is 40 while it is 50 at a dpa dose of 0.1. Due to the one-dimensional migration interstitials do not interact with each other and as a consequence, there is not a significant growth of interstitial clusters. Moreover, many self-interstitials have disappeared in sinks as already mentioned above.

According to the work by Griffiths [11] both vacancy and interstitial loops are observed under neutron irradiation. The relative concentrations depend on temperature, at high temperatures (>675 K) most of the loops are vacancy type while at low temperatures (<575 K) most of them are interstitial type. Between 573 K and 723 K approximately equal numbers of both types of loops are observed experimentally. Although it is not possible to make a quantitative comparison with these experiments, some conclusions can be extracted. On one hand our calculations show the formation of these two types of defects under irradiation. However, there is a discrepancy regarding the ratio of vacancies to self-interstitial loops formed. Clearly, the low recombination between vacancies and self-interstitials in the model results in high vacancy concentrations, and the low recombination between self-interstitials results in small self-interstitial clusters.

Three possible parameters could influence the accumulation and growth of self-interstitial clusters: (1) the mobility of these clusters, which could be impeded by the presence of impurities (2) the interaction between them and (3) their one-dimensional

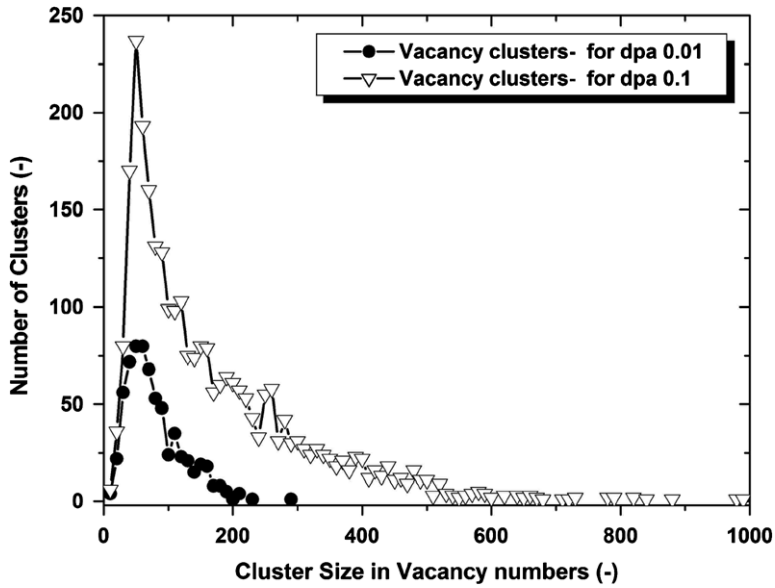

Fig. 3. Vacancy cluster size distribution for two dpa damage values, 0.01 and 0.1 for irradiation temperature of 600 K and all self-interstitial clusters mobile.

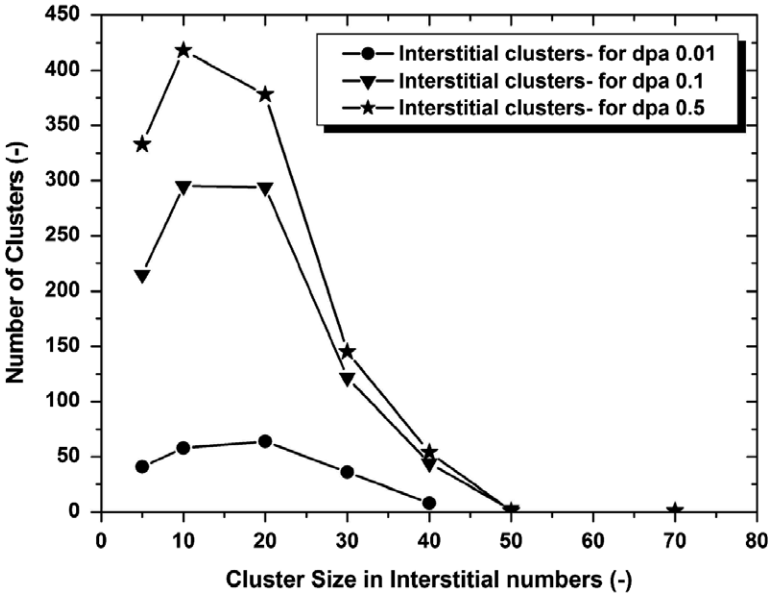

Fig. 4. Interstitial cluster size distribution for two dpa damage values, 0.01 and 0.1 for irradiation temperature of 600 K and all self-interstitial clusters mobile.

migration. Calculations have been performed to evaluate the influence of each of these parameters and to improve the agreement with the experimental observations.

In order to check the influence of self-interstitial cluster mobility, calculations were performed where all self-interstitial clusters are immobile and only single self-interstitials are mobile. Single self-interstitials move one-dimensionally as in the case above. Fig. 5(a) shows the visible vacancy cluster concentration as a function of dpa damage for this particular condition (circles), compared to the case above (squares). Fig. 5(b) shows the concentration of visible self-interstitials. There is a reduction in the concentration of visible vacancies due to higher recombination with the immobile self-interstitials, however the difference is not significant. In this case since only single self-interstitials are allowed to move there is a large population of small self-interstitial clusters. Larger self-interstitial clusters are formed compared with the case above, while before only clusters up to size 50 are formed at a dpa dose of 0.1, clusters up to size 70 are observed now. However, the total concentration of these visible clusters is not significantly different as shown in Fig. 5(b).

A second factor that could influence the size of the self-interstitial clusters formed is the existence of a bias for the interaction between them. Since the displacement field around an interstitial cluster could be larger that that around a vacancy cluster it is sometimes assumed a stronger interaction between self-interstitials and other defects. In the model presented here this interaction is effectively included through a difference in the capture radius between defects. The capture radius is modified by a factor Z, which corresponds to the interaction bias. We have included a value of $Z = 1.4$ for interstitials, while the value for vacancies remains $Z = 1$. Fig. 5(a) shows the visible vacancy cluster concentration obtained when this bias is introduced (triangles). In this calculation all self-interstitial clusters are considered to be mobile with a one-dimensional migration as in the first case discussed. No changes in the concentration of visible vacancy clusters are observed in this case. The concentration of visible self-interstitial clusters is slightly higher. For example at a dpa of 0.1 the visible self-interstitial cluster concentration is $4 \times 10^{15}\,\mathrm{cm}^{-3}$ when including the higher bias for interstitials while it is below $1 \times 10^{15}\,\mathrm{cm}^{-3}$ when no bias is included. The difference, nevertheless, is still not significant.

Finally we have studied the influence of the one-dimensional migration of interstitial clusters on damage accumulation. Simulations were performed considering all interstitial clusters mobile but following the normal three-dimensional diffusion. Results of these calculations for visible clusters of vacancies and self-interstitials are also included in

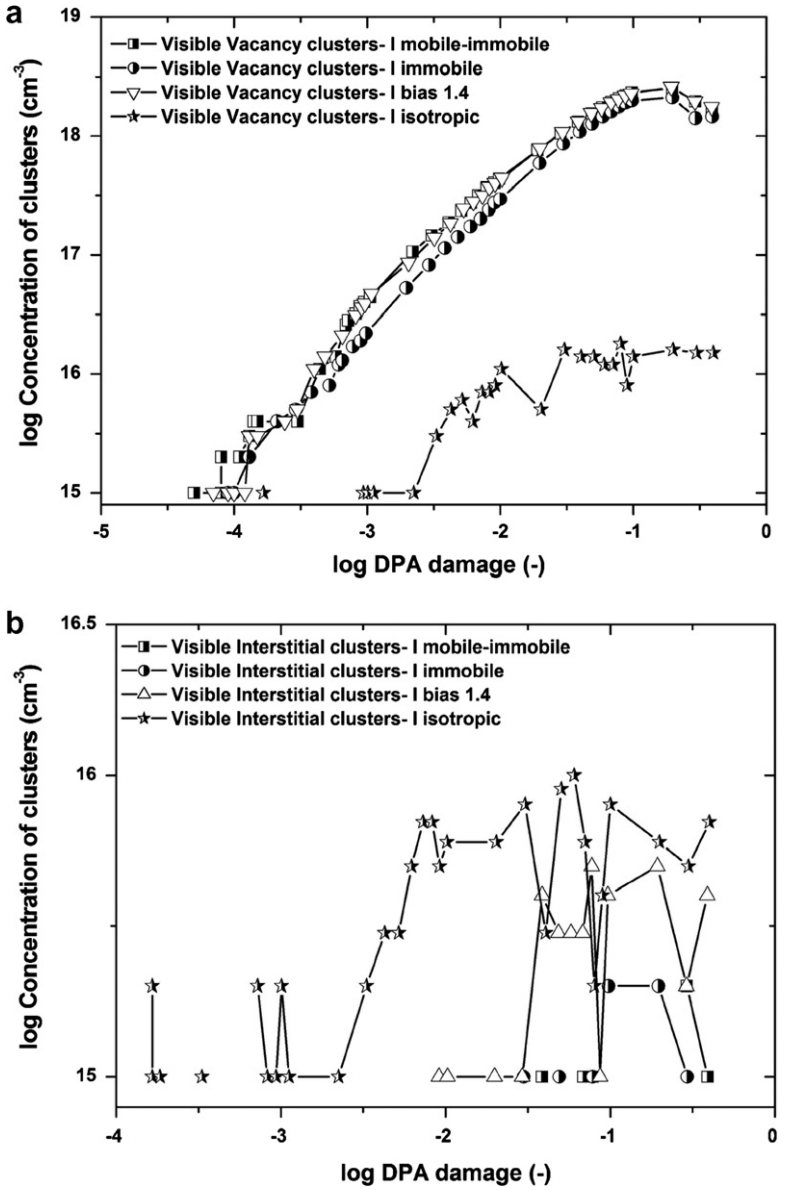

Fig. 5. Visible concentration of vacancies (a) and interstitials (b) as a function of dose for a temperature of 600 K. Clusters with more than 50 defects, equivalent to loops of 2 nm radius, are considered visible. Four different cases are presented: squares – clusters of interstitial mobile, circles – clusters of interstitials immobile, triangles – increasing the interaction bias between interstitials and stars – three-dimensional migration of SIAs.

Figs. 5(a) and (b), respectively (isotropic diffusion). Clearly the difference here of the visible vacancy cluster concentration as compared with the cases above is very significant, at a dose of 0.5 it has been reduced by two orders of magnitude. The concentration of visible interstitial clusters is also higher, although the difference is not as large as for vacancies. Notice that in this case the concentration of vacancy and self-interstitial clusters are similar for a dose of 0.5 in closer agreement to the experimental results. The cluster size distribution for both species is also similar, with maximum cluster sizes of 70–80 defects at a dose of 0.5. A high recombination between vacancies and interstitial (≈90%) dominates in this case. This results in significantly lower sizes for vacancy clusters as compared with the case of one-dimensional diffusion.

4. Conclusions

The influence of self-interstitial cluster migration on damage accumulation in zirconium has been studied. At 600 K the formation of both vacancy and self-interstitial clusters is observed under fission irradiation conditions. The relative concentration of vacancies and self-interstitials depends significantly on the type of diffusion assumed for self-interstitials. When self-interstitial clusters move one-dimensionally the concentration of self-interstitial clusters is very low compared to that of vacancies. At this temperature small vacancy clusters dissociate and single vacancies are mobile. These effects together with the low recombination with self-interstitials and the isotropic diffusion of vacancies, gives rise to vacancy clusters of sizes clearly visible through TEM. Self-interstitial clusters, however, are no able to grow due to the one-dimensional migration and reach with this model sizes much smaller than those observed experimentally.

Considering all self-interstitial clusters as immobile in the model or a higher interaction bias between self-interstitials does not increase the size of these clusters significantly and does not reduce the total defect concentration. Only when self-interstitials are considered to move three-dimensionally there is a significant growth of self-interstitial clusters, a higher recombination with vacancies and therefore a lower total defect concentration. Under these assumptions the concentration of self-interstitials and vacancies is similar, in better agreement with the experimental results. Therefore, it is possible that at this temperature, 600 K, the migration of these defects is not purely 1D but some migration in two dimensions within the basal plane, or outside the plane could be expected. In fact molecular dynamics simulations show that at this temperature these migration modes are not negligible [12]. The kMC model is currently being extended to include different diffusion probabilities along different directions to take this effect into account more accurately.

Acknowledgements

We want to thank N. de Diego, R. Voskoboinikov and D.J. Bacon for useful discussions and for providing the information on cascades and energetics of defects. Calculations were done under the 'Matcon' Linux-cluster at the UA. Thanks to J.L. McDonald for computer support. This work has been funded by the EURATOM-project FIKS-CT-2001-00137 (SIRENA) 'Simulation of radiation effects in Zr–Nb alloys: Application to stress corrosion cracking behavior in iodine-rich environment'. One of the authors (MJC) thanks the Spanish MEC for support through the Ramon y Cajal program.

References

[1] R. Voskoboinikov, Yu.N. Osetsky, D.J. Bacon, Nucl. Instrum. and Meth. B 242 (2006) 68.
[2] R. Voskoboinikov, Yu.N. Osetsky, D.J. Bacon, Nucl. Instrum. and Meth. B 242 (2006) 530.
[3] Yu.N. Osetsky, D.J. Bacon, N. de Diego, Metall. Mater. Trans. A 33 (2002) 777.
[4] N. Soneda, T. Diaz de la Rubia, Philos. Mag. 78 (1998) 995.
[5] C. Domain, C.S. Becquart, L. Malerba, J. Nucl. Mater. 335 (2004) 121.
[6] C. Arévalo, M.J. Caturla, J.M. Perlado, Proc. Mater. Res. Soc. 792 (2004) 19.
[7] D. Kulikov, L. Malerba, M. Hou, Nucl. Instrum. and Meth. B 228 (2005) 145.
[8] M.D. Johnson, M.J. Caturla, T. Diaz de la Rubia, J. Appl. Phys. 84 (1998) 1963.
[9] C. Arévalo, M.J. Caturla, J.M. Perlado, Nucl. Instrum. and Meth. B 228 (2005) 176.
[10] M.J. Caturla, N. Soneda, E. Alonso, B.D. Wirth, T. Díaz de la Rubia, J.M. Perlado, J. Nucl. Mater. 276 (2000) 13.
[11] M. Griffiths, J. Nucl. Mater. 159 (1988) 190.
[12] B.J. Whitting, D.J. Bacon, MRS Proc. 439 (1997) 389.