ELSEVIER

Journal of Nuclear Materials 295 (2001) 290–294

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Effect of defect sink strengths on the radiation induced segregation in binary alloys

M.V. Sorokin *, A.E. Volkov

Russian Research Centre, Kurchatov Institute, Kurchatov Sq. 1, 123182 Moscow, Russia

Received 10 November 2000; accepted 7 February 2001

Abstract

The radiation induced segregation (RIS) of alloy components at a flat free surface is investigated analytically and numerically. The segregation reversal, where depletion of a component at the surface is changed by its accumulation, is investigated. It is shown that the criterion for RIS reversal can be given in terms of some critical temperature. Dependencies of the critical temperatures on the partial damage efficiencies as well as on defect sink strengths are investigated in detail. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

Irradiation with fast particles creates in solids a supersaturated solution of point defects. Decay of the solution results in diffusion currents of point defects from the interior of the material to the sample surface. The coupling of the currents of point defects with those of atoms and different mobilities of alloy components results in the segregation of the components at the surface [1,2].

Competition between generation of point defects and their annihilation at volume sinks and the sample surface can lead to the reversal of the radiation induced segregation (RIS), i.e., where accumulation of an alloy component with lower diffusion ability at the surface is replaced by its depletion [3–6].

The dependencies of this effect on the partial generation rates and irradiation temperature have been investigated recently [5,6], but an effect of different defect sink strengths on the reversal of RIS was not taken into account there. In this paper, we describe the reversal of RIS in terms of the dependence on the defect sink strengths.

2. Formulation of the problem

We describe in this paper a semi-infinite irradiated binary alloy with a flat free surface. We consider the alloy as an ideal solution with random distribution of A- and B-atoms. Diffusion of the alloy atoms results from jumps of point defects (vacancies and interstitials) through the lattice. Trying to describe the reversal of RIS in the framework of the simplest analytical model, we do not take into account more complicated mechanisms of the defect migration or those whose validity for the atomic transport during irradiation is not finally established now (e.g., migration of small interstitial clusters [7] vs [8]).

Defect accumulation during irradiation is described by generation rates, both for vacancies and interstitials, $G_v = G_i = G_A + G_B$ (where G_Y are the partial generation rates describing displacements of Y-atoms from lattice sites ($Y = A,\ B$)). These generation rates are considered as input parameters. We suppose that the partial damage rates G_Y are given by the following equation [6]:

$$G_Y = \frac{\eta_Y C_Y G_v}{\eta_A C_A + \eta_B C_B} = \eta_Y C_Y G, \tag{1}$$

where η_Y is the partial damage efficiency, and the effective generation rate G is defined as

$$G = \frac{G_v}{\eta_A C_A + \eta_B C_B}. \tag{2}$$

---

* Corresponding author. Tel.: +7-095 196 9766; fax: +7-095 882 5804.

E-mail address: sorokin@dni.polyn.kiae.su (M.V. Sorokin).

0022-3115/01/$ - see front matter © 2001 Elsevier Science B.V. All rights reserved.
PII: S 0 0 2 2 - 3 1 1 5 (0 1) 0 0 5 0 4 - 9

2.1. Governing equations

At the temperatures and dose rates which are relevant to applications, recombination of point defects can be neglected [9,10] and excess defects created by irradiation annihilate mainly due to their absorption at the sample surface and volume sinks consisting of voids and dislocations. Coupling between the diffusion currents of defects and alloy components results in both the RIS of the components at the surface and the movement of this surface due to creation/dissolution of new layers by the defect currents [1–6].

Due to the adiabatic principle [5,6,11], defect profiles adjust themselves to the current position of the sample surface. Hence, it is possible to introduce a coordinate system related with the moving surface of the sample where the steady-state diffusion of point defects is described by [6]

$$-\omega\nabla\overrightarrow{j_{\mathrm{v}}} + G_{\mathrm{v}} - k_{\mathrm{v}}^2 D_{\mathrm{v}}(C_{\mathrm{v}} - \overline{C}_{\mathrm{v}}^{\mathrm{eq}}) = 0, \tag{3}$$

$$-\omega\nabla\overrightarrow{j_{\mathrm{Y_i}}} + G_{\mathrm{Y}} - (k_{\mathrm{Y_i}})^2 D_{\mathrm{Y_i}} C_{\mathrm{Y_i}} = 0; \quad \mathrm{Y} = \mathrm{A},\ \mathrm{B}. \tag{4}$$

Here C_{v} and $C_{\mathrm{Y_i}}$ are the atomic concentrations of vacancies and Y-interstitials respectively; $\overrightarrow{j_{\mathrm{v}}}$ and $\overrightarrow{j_{\mathrm{Y_i}}}$ are the diffusion currents of point defects; ω is the atomic volume. $D_{\mathrm{v}} = d_{\mathrm{A_v}} C_{\mathrm{A}} + d_{\mathrm{B_v}} C_{\mathrm{B}}$ is the diffusion coefficient of vacancies; $d_{\mathrm{Y_\alpha}} = (1/6)\,\lambda_\alpha^2 z_\alpha \nu_{\mathrm{Y_\alpha}}$ ($\alpha = \mathrm{v},\ \mathrm{i}$) are the partial diffusion coefficients [1]; λ_α, z_α are, respectively, the diffusion jump length and the number of nearest neighbors for an α-defect; $\nu_{\mathrm{Y_\alpha}} = \nu_{\mathrm{Y_\alpha}}^0 \exp(-\Phi_{\mathrm{Y_\alpha}}^{\mathrm{m}}/T)$, $\nu_{\mathrm{Y_\alpha}}^0$ is the attempt frequency of atomic jumps; $\Phi_{\mathrm{Y_\alpha}}^{\mathrm{m}}$ is the Gibbs free energy of the Y-atom migration via the α-defect. For simplicity equal partial diffusion coefficients of different alloy species via interstitials ($D_{\mathrm{Y_i}} = d_{\mathrm{Y_i}} = d_{\mathrm{i}}$) are assumed below. $\overline{C}_{\mathrm{v}}^{\mathrm{eq}}$ is the equilibrium vacancy concentration at surfaces of the volume sinks averaged over the ensemble of volume sinks (corresponding interstitial concentration is negligible). The sink strengths, k_β, have the following form: $k_\beta^2 = Z_\beta^{\mathrm{d}}\rho_{\mathrm{d}} + 4\pi N_{\mathrm{V}}\langle Z_\beta^{\mathrm{V}} R_{\mathrm{V}}\rangle$ [12]. $Z_\beta^{\mathrm{d/V}}$ are the bias factors for the absorption of β-defects at the dislocations/voids; N_{V} and R_{V} are the void volume density and the void radius respectively; $\langle\ \rangle$ denotes the average over the ensemble of voids; ρ_{d} is the dislocation density.

The diffusion currents of point defects in Eqs. (3) and (4) are given by the equations, where the coupling of the vacancy diffusion with that of the alloy components is

$$\overrightarrow{j_{\mathrm{v}}} = \overrightarrow{j_{\mathrm{A_v}}} + \overrightarrow{j_{\mathrm{B_v}}}, \tag{5}$$

where $\overrightarrow{j_{\mathrm{Y_v}}}$ are the vacancy currents via Y-atoms.

$$\omega\overrightarrow{j_{\mathrm{Y_v}}} = d_{\mathrm{Y_v}}(-C_{\mathrm{Y}}\nabla C_{\mathrm{v}} + C_{\mathrm{v}}\nabla C_{\mathrm{Y}}), \tag{6}$$

$$\omega\overrightarrow{j_{\mathrm{Y_i}}} = -d_{\mathrm{i}}\nabla C_{\mathrm{Y_i}}. \tag{7}$$

At the sample surface ($x = 0$) the equilibrium concentrations of point defects are maintained:

$$C_\beta(0) = \overline{C}_\beta^{\mathrm{eq}} = \exp\left(\frac{S_\beta^{\mathrm{f}}}{k}\right)\exp\left(-\frac{H_\beta^{\mathrm{f}}}{kT}\right), \tag{8}$$

where S_β^{f} and H_β^{f} are the defect formation entropy and enthalpy respectively.

We also demand that all concentrations are bounded above and all diffusion currents tend to zero in the interior of the material:

$$\left.\overrightarrow{j_\beta}\right|_{x\to\infty} = 0. \tag{9}$$

Neglecting the lattice sites occupied by voids, the conservation law for the lattice sites is

$$C_{\mathrm{A}} + C_{\mathrm{B}} + C_{\mathrm{v}} = 1. \tag{10}$$

A difference between evaporation and absorption of defects results in the movement of the surface with the velocity V:

$$V = \omega\big(j_{\mathrm{A_i}} + j_{\mathrm{B_i}} - j_{\mathrm{v}}\big)_{x=0}. \tag{11}$$

Taking into account Eq. (5), Eq. (11) can be reduced to the following:

$$V = \omega(j_{\mathrm{A}} + j_{\mathrm{B}})_{x=0}, \tag{12}$$

where

$$\left.\overrightarrow{j_{\mathrm{Y}}}\right|_{x=0} = \left(\overrightarrow{j_{\mathrm{Y_i}}} - \overrightarrow{j_{\mathrm{Y_v}}}\right)_{x=0} \tag{13}$$

are the total diffusion currents of alloy species at the sample surface.

3. Reversal of RIS

When no segregation exists ($\nabla C_{\mathrm{Y}} = 0$), Eqs. (3)–(6) are reduced to the following:

$$D_{\mathrm{v}}\nabla^2 C_{\mathrm{v}} + G_{\mathrm{v}} - k_{\mathrm{v}}^2 D_{\mathrm{v}}(C_{\mathrm{v}} - \overline{C}_{\mathrm{v}}^{\mathrm{eq}}) = 0, \tag{14}$$

$$d_{\mathrm{i}}\nabla^2 C_{\mathrm{Y_i}} + G_{\mathrm{Y}} - (k_{\mathrm{Y_i}})^2 d_{\mathrm{i}} C_{\mathrm{Y_i}} = 0; \quad \mathrm{Y} = \mathrm{A},\ \mathrm{B}. \tag{15}$$

Solving Eqs. (14) and (15) with equilibrium boundary conditions (8)–(9) one obtains defect currents at the sample surface ($x = 0$):

$$\omega \left. j_{\mathrm{Y_i}}\right|_{x=0} = -\frac{G_Y}{k_{\mathrm{Y_i}}}, \tag{16}$$

$$\omega \left. j_{\mathrm{Y_v}}\right|_{x=0} = -\frac{d_{\mathrm{Y_v}} C_Y G_{\mathrm{v}}}{D_{\mathrm{v}} k_{\mathrm{v}}}, \tag{17}$$

$$\omega \left. j_{\mathrm{v}}\right|_{x=0} = -\frac{G_{\mathrm{v}}}{k_{\mathrm{v}}}. \tag{18}$$

Because no segregation exists in the investigated case ($\nabla C_Y = 0$) the diffusion currents (16)–(18) maintain the initial concentrations of the alloy species at the sample surface:

$$\left.\frac{j_A}{j_B}\right|_{x=0} = \frac{C_A^0}{C_B^0}. \tag{19}$$

Substituting Eqs. (13), (16) and (17) into Eq. (19) we obtain the criterion of the segregation absence:

$$\frac{d_{A_v} - d_{B_v}}{D_v} = \frac{k_v G}{G_v}\left(\frac{\eta_A}{k_{A_i}} - \frac{\eta_B}{k_{B_i}}\right). \tag{20}$$

Taking into account Eqs. (11) and (16)–(18) we also obtain that in the case when no segregation occurs at the sample surface the surface velocity is given by

$$V = G\left(\frac{\eta_A C_A + \eta_B C_B}{k_v} - \frac{\eta_A C_A}{k_{A_i}} - \frac{\eta_B C_B}{k_{B_i}}\right). \tag{21}$$

4. Discussion

The obtained criterion of RIS absence is based on the coincidence of the concentrations of alloy species in new layers at the sample surface formed by the diffusion currents and the concentration in the interior of the sample (see Eq. (19)). The diffusion currents depend on partial mobilities of alloy components. Because these mobilities are temperature dependent, the temperature is an important parameter governing the 'direction' of RIS. From this point of view it is reasonable to formulate the criterion in terms of the temperature.

It should be noted that at equal defect sink strengths ($k_{A_i} = k_{B_i} = k_v$) Eq. (20) is reduced to the criterion obtained in [6]:

$$\frac{d_{A_v}}{d_{B_v}} = \frac{\eta_A}{\eta_B}. \tag{22}$$

One can see that similar to Eq. (22), criterion (20) can be reformulated in terms of some critical temperature T_c, which can be evaluated from (see Fig. 1) and does not depend on the generation rate G

$$\frac{d_{A_v}(T_c) - d_{B_v}(T_c)}{d_{A_v}(T_c)C_A + d_{B_v}(T_c)C_B} = \frac{k_v}{\eta_A C_A + \eta_B C_B}\left(\frac{\eta_A}{k_{A_i}} - \frac{\eta_B}{k_{B_i}}\right). \tag{23}$$

Indeed if the parameters of the partial mobilities ($\nu^0_{Y_\alpha}$ and $\Phi^m_{Y_\alpha}$) are fixed, the critical temperature might depend on the effective generation rate G, the partial damage efficiencies η_Y, and the sink strength k_{Y_i}, k_v. But one can see, when no segregation exists, the defect concentrations as well as diffusion currents of alloy species depend linearly on the effective generation rate G (see Eqs. (16)–(18)) and we obtain that the critical temperature depends only on the ratio η_B/η_A and the sink strength k_{Y_i}, k_v. Moreover, it is clear from Eq. (23) that the effect of different sink strengths for different interstitial atoms k_{Y_i} can be taken into account by corresponding modifications of the particular damage efficiencies η_Y. In this case the dependencies of the critical temperature are reduced to those on the ratios η_B/η_A and k_i/k_v.

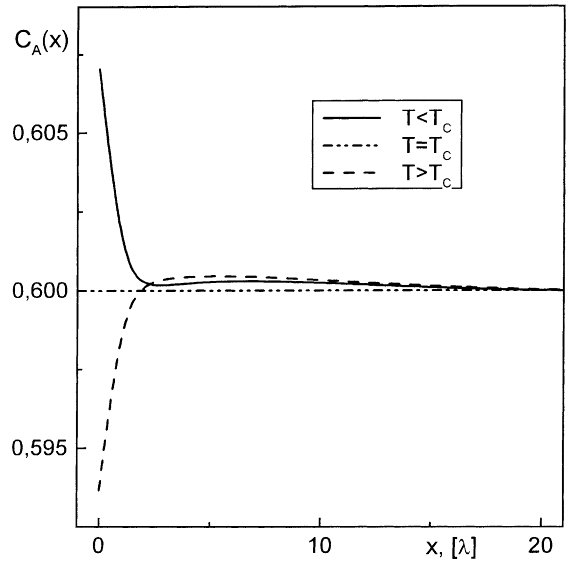

Fig. 1. Reversal from the depletion to accumulation of an alloy component at the precipitate surface at the critical temperature [6].

Using the numerical solution of the general diffusion equations governing RIS at the sample surface, we defined the dependencies of the critical temperature on the sink strengths for the parameters summarized in Table 1 [6]. In Fig. 2 we present the dependencies of the concentration C_A ($x = 0$) at the sample surface on the irradiation temperature and effective generation rate at different ratios of the sink strengths k_i/k_v and fixed η_B/η_A. The critical temperatures correspond to the value $(C_A\ (x = 0)/C_A^0) = 1$. One can see that the critical temperature T_c depends only on the ratio k_i/k_v and not on the effective generation rate G. In Fig. 3 we show a comparison between the critical temperature obtained from the analytical criterion (23) and that defined from numerical solution of the RIS problem [6]. Good agreement is achieved for all values of the ratios k_i/k_v.

In Fig. 4. we demonstrate the analytical dependence of the critical temperature on the ratio of the partial damage efficiencies η_B/η_A and defect sink strengths k_i/k_v. One can see from this figure that for any ratio of the sink strengths, k_i/k_v, the critical temperature increases monotonically when η_B/η_A decreases whereas, depending on η_B/η_A, $T_c(k_i^2/k_v^2)$ can either increase or

Table 1
Parameters used in the numerical calculations

Parameter	Notation	Value	
		Y = A	Y = B
Vacancy jump rate	$\nu^0_{\mathrm{Y_v}}$ (s^{-1})	3.5×10^{13}	1.5×10^{13}
Interstitial jump rate	$\nu^0_{\mathrm{Y_i}}$ (s^{-1})	0.5×10^{13}	0.5×10^{13}
Vacancy migration Gibbs energy	$\Phi^{\mathrm{m}}_{\mathrm{Y_v}}$ (eV)	1.2	1.14
Interstitial migration Gibbs energy	$\Phi^{\mathrm{m}}_{\mathrm{Y_i}}$ (eV)	0.9	0.9
Equilibrium interstitial concentration	$C^0_{\mathrm{Y_i}}$	0	0
Formation enthalpy of a vacancy	$H^{\mathrm{f}}_{\mathrm{v}}$ (eV)	1.4	1.4
Formation entropy of a vacancy	$S^{\mathrm{f}}_{\mathrm{v}}$	$5k$	$5k$
Dislocation density	ρ_{d} (m^{-2})	1×10^{14}	1×10^{14}
Vacancy absorption bias factor	Z_{v}	1	1
Interstitial absorption bias factor	Z_{i}	1.2	1.2
Initial concentrations of alloy species	C^0_{Y}	0.6	0.4
Damage rate	G (dpa/s)	$(1–3) \times 10^{-4}$	
Damage efficiency	η_{Y}	1	1.1
Temperature	T (K)	700–800	

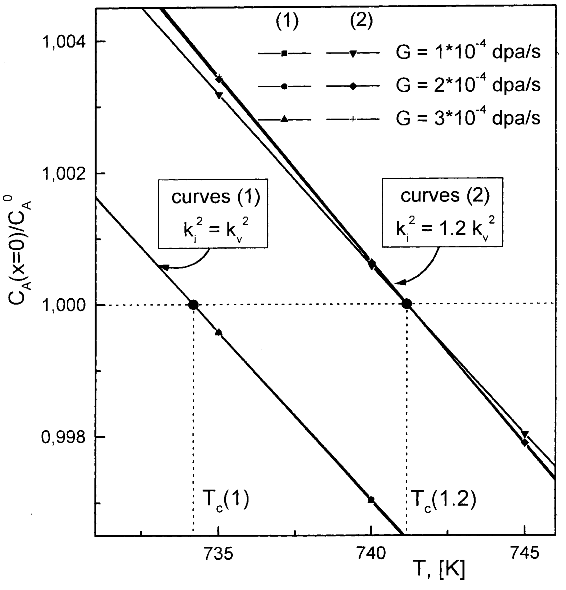

Fig. 2. Dependencies of the concentration C_{A} $(x = 0)$ at the sample surface on the irradiation temperature and effective generation rate obtained from the numerical solution of the general diffusion equations governing RIS in binary alloys for different ratios of the sink strengths $k_{\mathrm{i}}/k_{\mathrm{v}}$ [6]. The critical temperatures correspond to the value $(C_{\mathrm{A}}\ (x = 0)/C^0_{\mathrm{A}}) = 1$. Parameters used in the numerical calculations are summarized in Table 1.

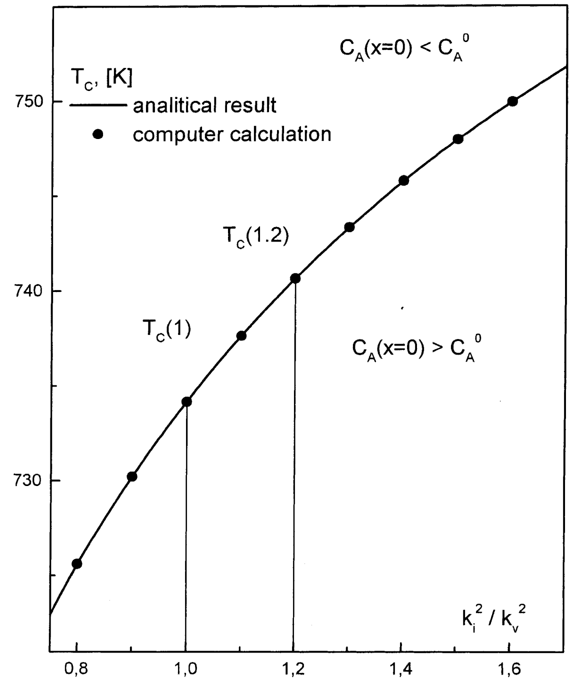

Fig. 3. Comparison between dependencies of the critical temperature obtained from the analytical criterion (23) and from the numerical solution of the RIS problem [6]. Parameters used in the numerical calculations are summarized in Table 1.

decrease when $k_{\mathrm{i}}^2/k_{\mathrm{v}}^2$ increases (see also Fig. 5). Indeed, when interstitials are preferentially absorbed by sinks $(k_{\mathrm{i}} \gg k_{\mathrm{v}})$, RIS is governed by vacancy diffusion currents to the sample surface. In this case no segregation exists when the mobilities of atoms via vacancies are equal and the critical temperature tends to that providing equal mobilities of the alloy components via vacancies: $T_{\mathrm{c}} \to T_{\mathrm{c}}(d_{\mathrm{A_v}} = d_{\mathrm{B_v}})$. At $k_{\mathrm{i}} \sim k_{\mathrm{v}}$, in order to compensate the diffusion currents of interstitials to the sample surface one has to change the critical temperature in order to modify the ratio of mobilities of alloy species via vacancies. Hence, when the generation of B-interstitials

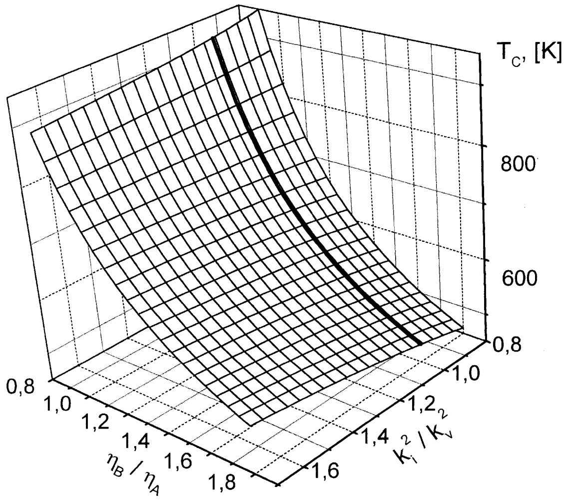

Fig. 4. Dependence of the critical temperature on ratios of the partial damage efficiencies η_B/η_A and defect sink strengths k_i/k_v.

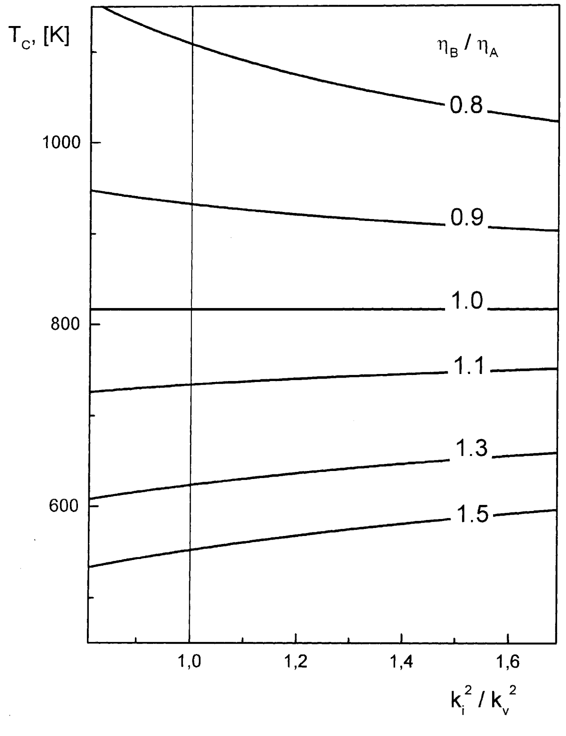

Fig. 5. Dependence of the critical temperature on the ratio of the defect sink strengths k_i/k_v for different η_B/η_A.

exceeds that of A-interstitials ($\eta_B > \eta_A$), in order to increase d_{B_v} the critical temperature has to be decreased when k_i decreases. And to the contrary ($\eta_B < \eta_A$) the critical temperature increases when k_i decreases.

5. Conclusions

A criterion for the absence of RIS is described in terms of the balance of the diffusion currents of alloy components at the surface.

The criterion is reduced to that for the critical temperature depending on the ratios η_B/η_A of the particular damage efficiencies and defect sink strengths k_i/k_v.

References

[1] H. Wiedesich, P.R. Okamoto, N.Q. Lam, J. Nucl. Mater. 83 (1979) 98.
[2] A.D. Marwick, J. Nucl. Mater. 135 (1985) 68.
[3] S. Watanabe, N. Sakagushi, N. Hashimoto, H. Takahashi, J. Nucl. Mater. 224 (1995) 158.
[4] H. Takahashi, N. Hashimoto, S. Watanabe, Ultramicroscopy 56 (1994) 193.
[5] A.E. Volkov, A.I. Ryazanov, J. Nucl. Mater. 256 (1998) 108.
[6] M.V. Sorokin, A.E. Volkov, J. Nucl. Mater. 282 (2000) 47.
[7] B.N. Singh, S.I. Golubov, A. Serra, Y.N. Osetsky, H. Trinkaus, A.V. Birashev, J. Nucl. Mater. 251 (1997) 107.
[8] V.A. Borodin, A.I. Ryazanov, J. Nucl. Mater. 256 (1998) 47.
[9] A.D. Braisford, R. Bullough, J. Nucl. Mater. 44 (1972) 121.
[10] A.D. Braisford, R. Bullough, Philos. Trans. R. Soc. London 302 (1981) 78.
[11] H. Haken, Synergetics. An Introduction, Springer, Berlin, 1983.
[12] M.F. Wehner, W.G. Wolfer, Philos. Mag. A 52 (1985) 189.