

AMORPHIZATION KINETICS UNDER ELECTRON IRRADIATION

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A model is brought forward of the kinetics of amorphization occurring under electron irradiation. The model is based on consideration of the phase transformations controlled by the structural relaxation of small-scale unstable atomic configurations excited by electrons.

INTRODUCTION

Electron irradiation can modify the kinetics of phase transformations in solids not only due to the diffusion enhancement as a result of the Frenkel pair production, but also due to irradiation-induced activation processes occurring on the crystal-amorphous interface. Research into those processes is of great importance for interpretation of the results of simulation tests, employing electron irradiation, of the structural materials to be used in molten-salt and high-temperature nuclear reactors. This work proposes a new model of the electron irradiation-induced phase transformations. We apply it to the description of amorphization of the intermetallic compound Zr_3Fe , because that process was studied experimentally and such theoretical models were proposed for its description that were based on considerations not accepted by us.

Motta and Olander [1] computed the contribution of point defects and compositional disordering to the free energy of the ordering crystal, and they assumed, as the necessary condition for amorphization, the following criterion:

$$\langle G_c \rangle \geq G_a \quad (1)$$

Here, $\langle G_c \rangle$ is the averaged free energy of irradiated crystal, G_a is the amorphous phase free energy. They calculated the kinetics of accumulation of point defects and chemical disordering in the ordering intermetallic compound $Zr(Cr, Fe)_2$ vs. time and found, employing the criterion (1), that the amorphization had to take place only after total compositional disordering and the vacancy concentration reaching about 0.009. This result was not supported by a number of studies, in which a partial compositional order was observed in amorphous alloys of the metal-metal type [2, 3].

This work proposes an alternative model of amorphization caused by electron irradiation. It is assumed that the amorphous phase nucleation occurs owing to the aggregation of radiation damage in such regions that may include point defect complexes, a sector of dislocation core, a boundary, a triple junction or some other

structural defects that lead to a considerable local increase of the free energy. The amorphous phase growth is controlled by both the diffusion and relaxation of the electron irradiation-excited non-equilibrium states of structural elements on the crystal-amorphous (C-A) phase boundary. According to observations of the field ion microscopy, the C-A boundary is of the order of interatomic distance wide [4]. The electrons, while being scattered on the boundary layer, excite and bring groups of atoms into a non-equilibrium state. The excited atomic configurations then relax into either amorphous or crystalline state. The Addendum considers the elementary phenomenological model that accounts for the nature of probabilities of the relaxation of excited atomic configurations into one phase or the other. Thus, within the framework of the given model, the amorphous phase can grow even in such regions where the criterion (1) is not fulfilled, if the radiation-stimulated diffusion process occurs faster than the reverse process of relaxation does into the crystalline state that has a lower free energy. Within the limits of the developed model, the critical electron flux for amorphization at a fixed temperature is calculated. Also, the required dose to amorphization is calculated vs. the values of electron flux and temperature in accordance with the Kolmogorov-Avrami theory of phase transformations.

A comparative study is made between theory and experiment on amorphization of the intermetallic compound Zr_3Fe , occurring under electron irradiation [5].

A MODEL OF MOTION OF THE INTERPHASE BOUNDARY

The observations made via field ion microscopy [4] indicated that the C-A boundary was narrow with the width of the order of the atomic scale. In our model, an assumption was made that atoms or groups of atoms near the interphase boundary could move into the other phase by a way of collective rearrangements with the average displacement $l \leq a$ (where a is the interatomic distance), causing the interphase boundary motion in this region.

Contributions to the rate of growth (dissolution) of precipitates of the amorphous phase are made both by irradiation and thermodynamic driving force. We shall designate those contributions in terms of u_{irr} and u_{th} , respectively:

$$u \equiv \frac{dR}{dt} = u_{irr} + u_{th}, \quad 2$$

where R is the amorphous phase precipitate radius.

The amorphous phase precipitate growth rate due to the action of the thermodynamic driving force has the following form:

$$u_{th} = r^{-1} D_{\Sigma} [\exp(\Delta g \beta) - 1], \quad 3$$

$$\Delta g \approx U_c - U_a + \xi T$$

where r is the size of the atomic group participating in the elementary act of interphase boundary rearrangement (of the order of several interatomic distances), U_c and U_a are the potential energy values of this group in the potential relief minima, corresponding to crystalline and amorphous states, respectively, ξ – configuration (at $T \rightarrow 0$) entropy of the group in amorphous state, $\beta = 1/k_B T$, k_B is the Boltzmann constant, D_{Σ} is the diffusion coefficient on the interphase boundary.

From (3) one can see that the amorphous phase precipitate growth rate under the action of thermodynamic driving force is positive when $\Delta g > 0$. When $\Delta g < 0$, the value of u_{th} is negative, but the growth rate can still be positive due to the contribution from u_{irr} (see, 4 below). It should be taken into account that the C-A boundary acts as a sink for point defects. For this reason, the crystalline phase layers adjacent to the boundary are free from point defects. One can also expect that, in an ordering alloy, the chemical order in these layers is higher than in the crystal bulk, far from the point defect sinks. That is why, it should be considered that $\Delta g < 0$ and $u_{th} < 0$.

The peculiarities of electron irradiation-induced damage in the vicinity of the interphase boundary are somewhat different from those in the bulk of the crystalline or amorphous phase. In the bulk of a solid, the electrons produce stable or unstable Frenkel pairs. The threshold energy of the Frenkel pair production in metals is usually about 30 eV. In the boundary layer the irradiation generates different unstable atomic configurations possessing the free energy higher than that in the amorphous state. The structural relaxation of these configurations may bring to either non-crystalline or crystalline structure. If σ_e is the cross-section of formation of the unstable atomic configuration in the boundary layer under the impact of electron irradiation, while J_e is the electron flux, then the generation rate of these configurations would be

$$\frac{dN_e}{dt} = 4\pi R^2 d J_e \sigma_e n, \quad 5$$

where N_e is a number of unstable configurations, n is the atomic concentration in the boundary layer and d is the layer width. The unstable atomic configuration may overlap both the crystalline and non-crystalline regions. Its structural relaxation leads to formation of the crystalline or amorphous phase. Let us denote by w_a and w_c the probabilities of the unstable configuration relaxation into the amorphous or crystalline state respectively and, assuming that $d \approx r$, we come to

$$u_{irr} \approx J_e \sigma_e n (w_a - w_c) r^4. \quad 6$$

As noted above, the amorphous phase precipitate growth rate u can be positive, even if $u_{th} < 0$, if $u_{irr} > 0$ and

$$u_{irr} > -u_{th} \quad 7$$

or

$$J_e > J_{e^i} = \frac{D_{\Sigma} [1 - \exp(\Delta g \beta)]}{\sigma_e (w_a - w_c) n r^5}. \quad 8$$

The derived expression (7) is the criterion of amorphization under the impact of electron irradiation.

KINETICS OF AMORPHIZATION UNDER ELECTRON IRRADIATION

The critical amorphous phase nuclei appear in crystal as heterophase fluctuations with concentration

$$w(k) \sim \exp(-\Delta G(k) \cdot \beta) \quad 9$$

$$\Delta G(k) = G_a(k) - G_c(k),$$

where $G_a(k)$ and $G_c(k)$ are the free energies of a group of k atoms in amorphous and damaged crystalline state respectively.

The rates of crystal-amorphous and amorphous-crystal transformations of k atoms are proportional to the appropriate self-diffusion coefficients:

$$I_{ca} \sim \exp(-G_c(k) \cdot \beta) \quad 10$$

$$I_{ac} \sim \exp(-G_a(k) \cdot \beta)$$

In a perfect crystal $I_{ca} \ll I_{ac}$ and the amorphous nuclei are unstable, but if there are stable structural defects in a crystal, then the value $\Delta G(k)$ can be small or even negative in a defect region that includes, for instance, point defect complexes, a dislocation core sector, an intergrain boundary, a triple junction or some other structural defect. In such regions, equilibrium precipitates of the amorphous phase may be formed. So, if the condition (1) is met locally, in a region whose size is greater than the critical size of the nucleus, then a heterogeneous amorphous phase nucleation may take place. Quite obviously, this nucleation cannot bring

about any noticeable amorphization of a sample, if the volume fraction of the regions where the condition (1) is met is small and the growth rate is controlled just by thermodynamic driving force. However, as noted above, the electron irradiation can have a considerable effect on the amorphous phase growth rate.

Let us designate as $V_d/V = n_d$ the volume fraction of the regions where the condition (1) is met and where the amorphous phase nucleation may occur (V_d is the volume occupied by extended defects or (and) irradiation-produced point defect complexes where the condition (1) is met, V is the total volume). If one designates the critical amorphous phase nucleus volume as V_a , then the value

$$N_d = n_d / V_a \quad (11)$$

is the volume density of the amorphous phase nucleation centers. The value N_d is time-dependent:

$$\frac{dN_d(t)}{dt} = -I_{ca} N_d(t) + I_{ac} [N_d(t) - N_d(0)] + I_{ca} N_d(0) \quad (12)$$

where h_d is the generation rate of point defect complexes under the impact of electron irradiation.

Equation (11) has the following solution:

$$N_d(t) = N_0 e^{(I_{ac} - I_{ca})t} + \frac{I_{ac} N_0 - h_d}{I_{ac} - I_{ca}} \left\{ 1 - e^{(I_{ac} - I_{ca})t} \right\} \quad (13)$$

Expressions obtained for the nucleation center concentration (12) and amorphous phase precipitate growth rate (2), (3), (5) can be used to describe the irradiation-affected amorphization kinetics within the framework of the Kolmogorov-Avrami theory. To simplify the final expressions, we impose the condition $|I_{ca}| \gg |I_{ac}|$ for nucleation centers and consider two cases.

In the first case, the initial extended structural defects play a decisive role in the amorphous phase nucleation, the value h_d in 14 being negligible. In this case, the amorphous phase volume fraction $X(t)$ is a time variable in accordance with the following expression [6]:

$$X(t) = 1 - \exp \left[-\frac{8\pi u^3 N_0}{I_{ca}^3} \left(e^{-I_{ca}t} - 1 + I_{ca}t - \frac{(I_{ca}t)^2}{2} \right) \right] \quad (15)$$

Asymptotically, in the limit $I_{ca}t \gg 1$, the expression 16 transforms into

$$X(t) = 1 - \exp \left[-4\pi N_0 u^3 t^3 / 3 \right] \quad (17)$$

From (14), one can see that the characteristic time of amorphization in this approximation is:

$$\tau_{ca} \sim (N_0^{1/3} u)^{-1} \quad (18)$$

while the dose-to-amorphization is

$$dpa_{ca} \sim \sigma_F J_e (N_0^{1/3} u)^{-1} \quad (19)$$

σ_F being the stable Frenkel pair generation cross-section.

In the second case considered here, the initial extended defects play no role and one can put $N_0 = 0$ in (12). Then, in the limit $I_{ca}t \gg 1$, we have:

$$N_d(t) \approx h_d / I_{ca} \quad (20)$$

and (see [6])

$$X(t) = 1 - \exp \left\{ -\frac{\pi h_d u^3 t^4}{3} \right\} \quad (21)$$

From (18), one can deduce the characteristic time of amorphization in this approximation:

$$\tau_{ca} \sim (h_d u^3)^{-1/4} \quad (22)$$

while the dose-to-amorphization is

$$dpa_{ca} \sim \sigma_F J_e (h_d u^3)^{-1/4} \quad (23)$$

In the both cases considered, we have a lag time of amorphization $t_{lag} \sim 1/I_{ca}$. At $t \gg t_{lag}$, the C-A transformation is described by the asymptotic expressions, (14) or (18).

NATURE OF DIFFUSION PROCESSES ON INTERPHASE BOUNDARY

As regards the conventional thermally activated diffusion, the diffusion coefficient expression has the form:

$$D_{th} = r^2 v_0 \exp(-E\beta) \quad (24)$$

where r is the length of the jumps (in our case, it is the size of the atomic group transiting from one phase to the other), v_0 is the frequency of attempts (of the order of the Debye frequency), E is the activation energy value.

Yet, the final value of the experimentally measured critical electron flux needed for amorphization at low temperatures, as shown in Fig.1, comes out in favor of a supposition that there should be a certain supplementary athermal diffusion process leading to interphase boundary rearrangements. We assume that this athermal diffusion process should be controlled by quantum-mechanical tunneling. In this case, the athermal diffusion coefficient expression acquires the form:

$$D_{ath} = r^2 v_0 D \quad (25)$$

where the coefficient of transmission under potential barrier is

$$D = \exp\left(-\frac{2}{\hbar} \sqrt{2 m E l}\right),$$

m is the tunneling complex mass, E is the potential barrier height, l is the distance between equilibrium positions, \hbar is the Planck constant.

Another possible contribution to the interphase boundary diffusion can be made from the radiation-stimulated processes as a result of the so-called "radiation shaking" [7]. This term implies the influence of phonons produced by the annihilation of the irradiation-generated unstable Frenkel pairs with the mean radius being of the order of the interatomic distance and lifetime $10^{-12} - 10^{-11}$ s. The radiation-stimulated diffusion coefficient in this case is determined by the expression

$$D_r = \frac{4\pi J_e N \sigma_F r^2 n}{3} \left(\frac{K \Omega_F \Omega_{ac}}{6\pi E a^2} \right)^3, \quad 26$$

where N is the number of unstable pairs relative to that of stable ones, K is the bulk modulus, Ω_F is the volume of dilatation center corresponding to the unstable Frenkel pair, Ω_{ac} is the volume of dilatation center corresponding to the unstable atomic configuration on the interphase boundary.

Then, since the thermally activated, athermal and radiation-stimulated diffusion processes are independent, the expression for the total diffusion coefficient on interphase boundary would have the form:

$$D_\Sigma = \sqrt{D_{th}^2 + D_{ath}^2 + D_{r,2}^2}. \quad 27$$

DISCUSSION OF RESULTS

A comparison is given below of the predictions of our theory with experimental data published in [5]. Note at once that the experimental data agrees better with the expressions (19), (20) than with (15), (16), for the reasonable choice of the fitting parameters.

We choose the tunneling coefficient D to correspond to the transition of a pair Fe+Zr of atoms between two noncoincident lattice sites on the interphase boundary, which are separated by a distance $l \approx 3.6 \cdot 10^{-11} m$, under the potential barrier of 1675 K. We assume that as soon as such a transition takes place, the whole region with radius $r \approx 2a$ becomes unstable and tends to relax into the adjacent phase by the means of "radiation shaking" process. So, the tunneling of a pair of atoms between two noncoincident sites acts like a trigger for such region.

The values used for the fitting parameters are given in Table. They were chosen on the basis of the criterion of the best agreement with experiment, as given in Figures 1-4.

Values of fitting parameters

Parameter	N of formula	Value (SI)
$U_c - U_a$	28	$-2200 \cdot k_B (J)$
ξ	29	$6 \cdot k_B (J \cdot K^{-1})$

r	30	$5 \cdot 10^{-10} (m)$
σ_e	31	$2.5 \cdot 10^{-25} (m^2)$
$w_a - w_c$	32	0.2
v_0	33	$3 \cdot 10^{13} (s^{-1})$
D	34	$e^{-33.65}$
E	35	$6700 \cdot k_B (J)$
h_d	36	$0.77 \cdot J_e (m^{-3} s^{-1})$
N	37	100
σ_F	38	$5 \cdot 10^{-27} (m^2)$
n	39	$6.4 \cdot 10^{28} (m^{-3})$
K	40	$10^{11} (Pa)$
Ω_F	41	$1.6 \cdot 10^{-29} (m^3)$
Ω_{ac}	42	$0.8 \cdot 10^{-29} (m^3)$
a	43	$2.5 \cdot 10^{-10} (m)$

From Fig. 1 one can gather that theory agrees with experiment satisfactorily in the low temperature region, where, as we believe, tunneling diffusion plays a major role in the kinetics of the C-A boundary. In the region $T \geq 175 K$, where the contribution from thermally activated processes is substantial, a flatter progression of the experimental curve than calculated calls for additional analysis.

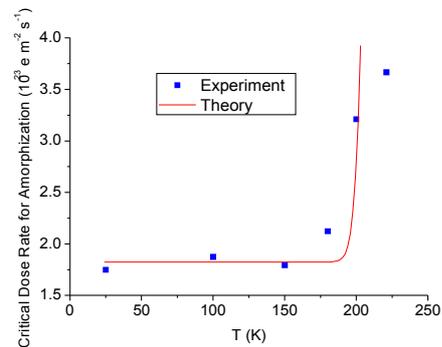


Fig. 1. The critical value of electron flux for amorphization vs. temperature (7)

One can see from Fig. 2 that our model produces the right results at $T = 28 K$ in the region of relatively large fluxes $J_e \gg J_{e,i} (T = 28 K)$.

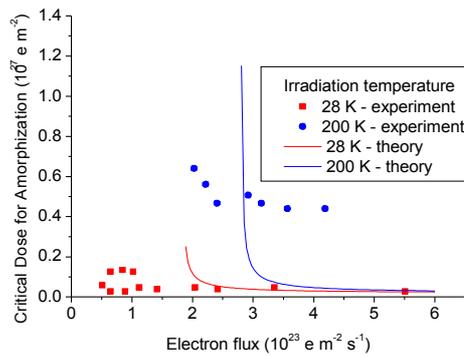


Fig. 2. Dependence of dose-to-amorphization (20) on electron flux density

The flatter progression of the experimental curve than predicted theoretically at $T \approx 200 K$, as we suppose, must have the same explanation that the discrepancy of our theory with experiment in this region in Fig. 1. The dose needed for amorphization, while approaching from the upper level downward to the critical flux value at the given temperature (see (7) and Fig. 1), must increase to infinity. We point out the discordance between the experimental data in Fig. 1 (Fig. 4 in [5]), where the critical flux values for amorphization at $T=28 K$ and $T=200 K$ are respectively about $1.8 \cdot 10^{23} e \cdot m^{-2} \cdot s^{-1}$ and $3.2 \cdot 10^{23} e \cdot m^{-2} \cdot s^{-1}$, and Fig. 2 (Fig. 6 in [5]) showing the finite values of the doses-to-amorphization at flux values that are smaller than the above-mentioned.

In the presented theory (Fig. 3), in the high temperature region, the value $dpa_{ca} \cdot \sqrt{J_e \sigma_F}$ does not depend on dose absorption rate, similarly to the theory [5].

The experimental observation of the dependence of this value on the dose absorption rate calls for additional analysis for the right explanation to be made.

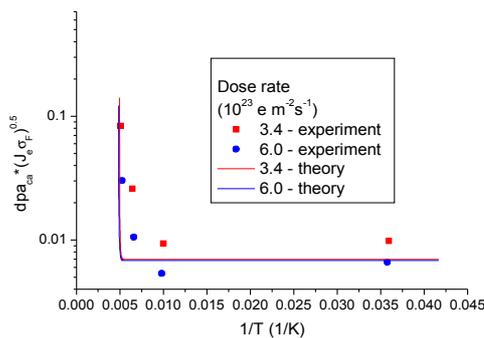


Fig. 3. Dependence of product "dose-to-amorphization (dpa) * square root of dose absorption rate (dpa/s)" vs. reverse temperature

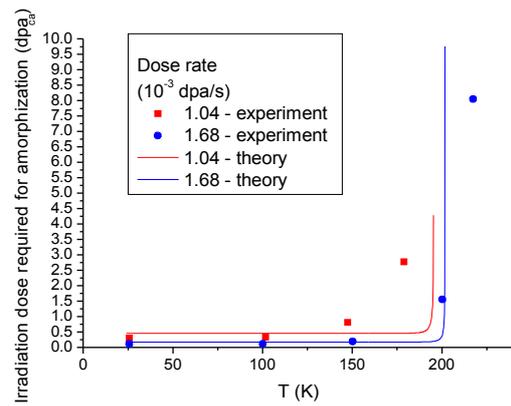


Fig. 4. Dependence of dose-to-amorphization (dpa) on temperature (20)

In the low temperature region, our theory agrees well enough with the experimental data on the dose-to-amorphization (Fig. 4). The flatter progression of the experimental curve than theoretically predicted, especially at low fluxes, at high temperatures, must be accounted for in the same way that the discrepancy of our model with the experimental data in the high temperature region in Fig. 1.

ADDENDUM. DYNAMICS OF OSCILLATOR WITH FRICTION IN TWO-WELL POTENTIAL UNDER ACTION OF RANDOM COERCIVE FORCE

Let us formulate the simple model in order to account for the relaxation dynamics of the atom on the C-A boundary. Let us consider the 1D particle motion in the two-well potential, as depicted in Fig. A1a. We shall also take into account the effects of thermal noises, dissipation and rare strong perturbations that imitate the radiation ambience. The phase portrait of this system in the absence of the dissipation and perturbations is given in Fig. A1b.

Here, L_1 and L_2 are the widths of the left- and right-hand separatrices, respectively; E_s and ΔE are the depth of the right-hand well and difference in the depths of the right- and left-hand wells, respectively; S_1 and S_2 are the phase space areas limited by the appropriate separatrices.

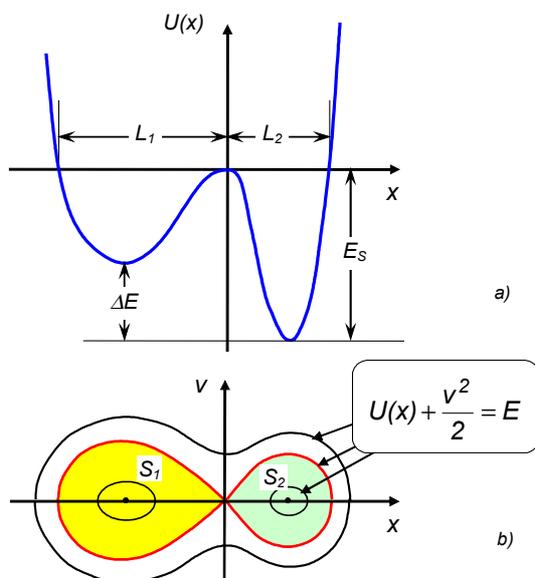


Fig. A1. Potential energy (a) and phase portrait (b) of 1D two-well potential

For the particle with unitary mass the equation of motion will be written as follows:

$$\frac{d^2 x}{dt^2} + 2\gamma \frac{dx}{dt} + \frac{dU(x)}{dx} = \Phi(t) + \sum_i g\delta(t-t_i) \quad (44)$$

Here, γ is the friction coefficient; $\Phi(t)$ is the random coercive force; g is the intensity of rare (on the scale of the period of oscillations) strong pulses imparting to the particle the energy $E \gg E_s$. The equation (25) differs from the Langevin one by the presence of the last term in the right-hand part that simulates rare strong pulses from electrons.

The value $\Phi(t)$ describes the “thermal noise” and has the following statistical properties:

$$\langle \Phi(t) \rangle = 0; \quad \langle \Phi(t)\Phi(t') \rangle = \kappa\delta(t-t') \quad (45)$$

$\kappa \sim T$.

In the absence of the right-hand part in the equation (25), the phase trajectory of the system will, depending on the initial conditions, tend to one of the focuses on the phase portrait.

At the condition $g=0$ (over large times the system is in equilibrium with the thermostat) and $T \ll E_s$ (the system spends most of its time in the regions limited by the separatrices), the probabilities for the system to be found in the regions S_1 and S_2 (in the left- or right-hand well, respectively) obey the following well-known relationship:

$$w_1^e/w_2^e = \exp(-\Delta E/k_B T), \quad (46)$$

the time spent by the system in one or the other well (lifetime) being

$$\tau_{1,2} \sim \exp(const \cdot \gamma E_s / \omega_{1,2} \kappa). \quad (47)$$

Here, $\omega_{1,2}$ is the frequency of oscillations in the left- or right-hand well, respectively.

To derive the expressions describing the probabilities to find the system in the region S_1 or S_2 at the condition $g^2 \gg E_s$, we consider two limiting cases:

1) Strong friction case (non-periodic regime)

$$\gamma > \omega_{1,2}, \quad \langle |t_i - t_{i+1}| \rangle \ll \tau_{1,2}. \quad (48)$$

In this case, the probabilities of staying in the left- or right-hand well are proportional to the widths (over the coordinate x) of the appropriate separatrices:

$$w_{1,2} \approx \frac{L_{1,2}}{L_1 + L_2}. \quad (49)$$

The obvious generalization of this result to the 3D case under consideration leads to the formula:

$$w_a - w_c \approx \frac{\rho_c}{\rho_a} - 1, \quad (50)$$

where ρ_c and ρ_a are the densities of the substance in crystalline or amorphous phase, respectively.

2) Weak Friction Case (Oscillatory Regime)

$$\gamma \ll \omega_{1,2}, \quad \langle |t_i - t_{i+1}| \rangle \ll \tau_{1,2}. \quad (51)$$

In this case, the probabilities of staying in the left- or right-hand well are proportional to the phase space areas limited by the appropriate separatrices [8, 9]:

$$w_{1,2} = \frac{S_{1,2}}{S_1 + S_2}. \quad (52)$$

In practice, instead of the phase volume, inside of which the system is confined, the entropy of the system, which is the value equal to the logarithm of the latter, is oftener used. In terms of the entropy the expression (33) acquires the form:

$$w_{1,2} = \frac{\exp(s_{1,2})}{\exp(s_1) + \exp(s_2)}, \quad (53)$$

where s_1 and s_2 are the entropies of the system in the regions 1 and 2. In this case, in order to compute the probabilities of the system staying in one or the other region, one needs to know the difference of its entropies in these regions.

In the specific case under consideration, the entropies of the amorphous and crystalline phases that are under the same external conditions differ by the value $\zeta \sim 1$, the co-called configuration entropy of the amorphous state, that has to do with the absence of the long-range order in the amorphous state. Then

$$w_a - w_c = \tanh \frac{\zeta}{2}. \quad (54)$$

From Table 1 one can see that the value of the fitting parameter $w_a - w_c = 0.2$ conforms better to the weak friction case, because the opposite case of the strong friction yields $w_a - w_c \ll 1$, since the densities of the amorphous and crystalline phases differ not more than by several percent.

CONCLUSIONS

This paper proposes a model of a polymorphous transformation under electron irradiation, taking into account both the processes of inhomogeneous nucleation of a new (in our case, amorphous) phase and the contribution from the relaxation of atomic configurations, excited on the interphase boundary by electron irradiation, into the process of the growth of precipitate of the new phase. The example of the simple dynamic system is used for clarification on the nature of probabilities of the structural relaxation. The quantitative comparison of the predictions of the proposed theory with experimental data on amorphization of the intermetallic Zr_3Fe under electron irradiation results in a reasonable agreement of theory with experiment. However, certain quantitative discrepancies with experimental data call for additional analysis.

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КИНЕТИКА АМОРФИЗАЦИИ ПОД ЭЛЕКТРОННЫМ ОБЛУЧЕНИЕМ

А.С. Бакай, А.А. Борисенко, К. Расселл

Приведена модель кинетики аморфизации под электронным облучением. Модель основывается на рассмотрении фазовых превращений, которые контролируются структурной релаксацией мелкомасштабных нестабильных атомных конфигураций, которые возбуждаются электронами.

КИНЕТИКА АМОРФИЗАЦІЇ ПІД ЕЛЕКТРОННИМ ВИПРОМІНЮВАННЯМ

О.С. Бакай, О.О. Борисенко, К. Расселл

Наведено модель кінетики аморфізації під електронним випромінюванням. Модель ґрунтується на розгляді фазових перетворень, які контролюються структурною релаксацією дрібномасштабних нестабільних атомних конфігурацій, які збуджуються електронами.