Propagating self-sustained annealing of radiation-induced interstitial complexes

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Abstract. A propagating self-sustained annealing of radiation induced defects as a result of thermal-concentration instability is studied. The defects that are considered in the model are complexes. Each of them consists of one atom of impurity and of one interstitial atom. Crystal with defects has extra energy which is transformed into heat during defect annealing. Simulation of the auto-wave of annealing has been performed. The front and the speed of the auto-wave have been obtained. It is shown that annealing occurs in a narrow region of time and space. There are two kinds of such annealing behaviour. In the first case the speed of the auto-wave oscillates near its constant mean value and the front of temperature oscillates in a complex way. In the second case the speed of propagation is constant and fronts of temperature and concentration look like sigmoid functions.

1. Introduction

The irradiation of materials with high-energy particles typically causes deterioration of their operational characteristics [1]. This leads to the necessity for frequent replacement, decontamination and disposal, which in itself is a difficult and expensive task. But the problem is particularly acute for bulky parts and assemblies, and especially for the reactor vessel. Therefore, exploring opportunities to extend the service life through the restoration of the operational characteristics of structural materials is extremely important, especially given the current state of nuclear power when the projected service life for many nuclear power plants is approaching the end.

It is known that one of the ways of restoring material characteristics after irradiation is annealing. However, restoring the material characteristics of bulky structures is difficult with conventional annealing techniques. The idea of extending the life of nuclear reactor vessels by annealing is though being actively discussed. Traditional annealing by heating to the necessary temperature of the entire reactor vessel requires a large amount of energy, a long period of time and can consequently cause irreversible phase-structural changes of the material.

For this reason, it is of interest to study the particularities of annealing by means of a temperature auto-wave, as it was proposed in [2]: annealing as a result of passing a short pulse of high temperature occurs locally and is devoid of the drawbacks discussed above. It is more precise, although associated with the setting up of a more complex auto-wave mechanism of temperature propagation. This mechanism
is based on the self-sustaining annealing of radiation defects. Consider a radiation damaged crystalline sample, which contains the radiation-induced lattice defects. Due to these defects, the crystal is not in a state of thermal equilibrium. In the formation of defects, the regular bonds are broken and new, irregular ones are established. As a result, stresses appear in the crystal and its elastic energy increases. Although the state of the crystal becomes metastable, its relaxation to equilibrium can take a long time, due to the high values of activation barriers.

The energy of a radiation-damaged sample after irradiation is significantly higher than its energy prior to irradiation. The magnitude of the difference between these energies is of the order of energy of the defect formation. For instance, the formation of a Frenkel pair requires from 5 to 7 eV. The energy stored in defects does not contribute to the thermal energy of the crystal, but it can be converted into thermal energy in the process of the annealing of defects, for instance, in the case of the recombination of vacancies and interstitial atoms. As a result, the temperature of the crystal increases, and since the intensity of defect annealing depends exponentially on temperature, the intensity of annealing is further boosted. Thus, a positive feedback loop is established. Under certain conditions, this positive feedback, which we will call thermal-concentration feedback, leads to the development of self-oscillations of temperature and of defect concentration [3, 4]. However, if the annealing is initially localized, then by thermal conduction and diffusion of the defects it may spread in space and again, under certain conditions, form a self-sustained wave.

Despite the topicality, the practical importance and a huge number of publications, the kinetics of annealing of radiation defects is studied insufficiently. This applies primarily to inhomogeneous annealing of, especially, multicomponent materials, and to nonlinear relations that can be realized in the process of annealing and lead to a wide variety of relaxation processes. It should also be noted that the study of the auto-wave annealing is of considerable academic interest because it allows the establishing of mechanisms of development of auto-waves in active media of a different nature.

2. Problem statement
Let us investigate the propagation of the annealing auto-waves caused by the thermal-concentration feedback using as an example the simplest model that describes the dynamics of the annealing inactive complexes. The complex binds an impurity atom, which is a trap for the defect, and one interstitial atom. Let the concentration of traps be \( n_t = \text{const} \), and \( n_0 = n_0(t) \) be the number of available traps that are not bound to interstitial atoms, and \( n_1 = n_1(t) \) be the number of traps bound with one interstitial atom. Both \( n_0 \) and \( n_1 \) change during the annealing process and are functions of time, but their sum remains constant: \( n_t = n_0(t) + n_1(t) \).

Since the concentration of the complexes is comparable with the concentration of traps [1, 4], the formation energy of the interstitial atoms is several times higher than one of the vacancies, then for high densities of dislocations, which are typical for irradiated materials, and for the characteristic temperatures of annealing, we can neglect the contribution of the vacancies. Note that this approximation reduces the energy accumulated in the defects and slows the development of the auto-wave annealing process in its initial stage.

Then the evolution of defect concentrations \( n_i, n_1 \) and of temperature \( T \) can be described by the system of equations:

\[
\frac{dn_i}{dt} = -\frac{n_i}{\tau_i} - \gamma_i n_0 n_i + \frac{n_1}{\tau_1}, \tag{1}
\]

\[
\frac{dn_1}{dt} = +\gamma_i n_0 n_i - \frac{n_1}{\tau_1}, \tag{2}
\]

\[
c \rho \frac{\partial T}{\partial t} = \kappa \frac{\partial^2 T}{\partial x^2} + \theta_i \frac{n_i}{\tau_i} + \theta_b \gamma_i n_0 n_i - \theta_b \frac{n_1}{\tau_1} - h(T - T_e). \tag{3}
\]

The first term in (1) describes the absorption of interstitial atoms by dislocations, the second and third terms in (1) and the first and the second terms in (2) describe the creation and the thermal destruction.
of complexes. Equation (3) describes the change in the crystal temperature with respect to thermal conductivity, heat sink and heat dissipation during annealing of defects and complexes. The last term in equation (3) describes the dissipation of heat. Here \( \gamma_i = \mu D_i \) is the complex creation rate constant, \( \tau_i^{-1} = z_i \rho_d D_i \) and \( \tau_i^{-1} = \mu D_i \nu \) are the inverse lifetimes of interstitial atoms with respect to absorption by dislocations and complexes with respect to thermal destruction. \( D_i = D_i \exp (-E_m / kT) \) is the diffusion coefficient, \( \rho_d \) is the density of dislocations and \( z_i \) is the preference of dislocation absorption of interstitial atoms. \( c \) and \( \rho \) are the specific heat capacity and the density of the crystal and \( \kappa \) is the heat transfer coefficient. \( E_m \) is the migration energy, \( \theta_i \) is the energy converted to heat in the process of one annealing of one interstitial atom, which is approximately equal to the energy of its creation and \( \theta_b \) is the release (absorption) of energy in destruction (creation) of the complex, \( E_b \). \( \theta_e \) is the environment (heat sink) temperature, \( k \) is the Boltzmann constant. \( \nu \) is the number of atoms in the unit volume. The lifetime of a complex with respect to thermal destruction, \( \tau_1 \) can be obtained from the principle of detailed balance: \( \gamma_i \nu n_i = n_i \gamma_i \).

The system (1) to (3) is essentially nonlinear, as the typical lifetimes of defects depend exponentially on temperature. Taking into account that the lifetime of the interstitial atom is much smaller than that of the complex, i.e. \( \tau_i \ll \tau_1 \), we set \( dn_i / dt = 0 \). Also take into consideration that the loss of interstitial atoms as a result of complex formation dominates. Then instead of (1) to (3), we obtain a system of two differential and one algebraic equation:

\[
\frac{dn_i}{dt} = -\frac{n_i}{\tau_1}, \quad (4)\\
\rho \frac{\partial T}{\partial t} = \kappa \frac{\partial^2 T}{\partial x^2} + (\theta_i - \theta_b) \frac{n_i}{\tau_1} - h(T - \theta_e), \quad (5)\\
\frac{n_i}{\tau_1} = \frac{n_1}{\tau_1}. \quad (6)
\]

Let the coordinate axis \( Ox \) be perpendicular to the surface and be directed into the medium. Consider the boundary conditions of the second kind: we assume zero heat flux across the boundary \( \Gamma: \frac{dT}{dx}|_{\Gamma} = 0 \). The initial concentration of complexes is given by: \( n_i (0, x) = n_{i,0} \). We will assume that the material temperature is initially equal to the environment temperature, \( \theta_e \), except for a narrow boundary layer of width \( \Delta \) in which, as a result of external heating, an elevated temperature, \( \theta_0 \), is attained and maintained (possibly for some finite period): \( T(t, x) = \theta_0 \) if \( 0 \leq x \leq \Delta \) and \( T(t, x) = \theta_e \) if \( x > \Delta \).

The problem of radiation defect annealing is similar to the well-known combustion problem [5], however unlike the latter the structure of radiation damages is non-stationary. Defects are continually annealed at a non-zero rate and the auto-wave can propagate to final distances only.

Non-linear system (4) – (6) was solved numerically for a finite interval of length \( L \) with the Crank-Nicolson finite difference scheme. This discretisation scheme yields a system of nonlinear equations, which was solved with Newton’s iterations at each time step. The Jacobian matrix used in the Newton method was obtained analytically and the system of linear equations was also analytically reduced to a tridiagonal form, which can be efficiently solved with the Thomas algorithm.

Calculations were performed for parameters of steel: \( c = 460 \) J/(kg·K), \( \rho = 7800 \) kg/m³, \( \kappa = 74.4 \) W/(m·K), \( z_i = 1.1, D_i = 10^{-5} \) m²/s, \( E_m = 0.3 \) eV, \( \rho_d = 10^{12} \) m⁻², \( \theta_i = 5.9 \) eV, \( \theta_b = E_b = 1 \) eV and \( \mu = 8 \cdot 10^{-8} \) m. Geometrical parameters and initial conditions were chosen as follows: \( L = 10 \) cm, \( \Delta = 1 \) mm, \( \theta_e = 300 \) K, \( \theta_0 = 425 \) K, \( n_{i,0} = 6 \cdot 10^{28} \) m⁻³ and \( n_{i,0} = 8 \cdot 10^{26} \) m⁻³. However, in order to investigate mechanisms of propagation of a self-sustained wave of radiation defect annealing, the parameters were varied in wide ranges.

3. Results and discussions

The obtained results are presented in figures 1 and 2. Note that in this paper, we will report defect concentration normalized by a constant factor, \( C = \frac{(\theta_i - \theta_b)n_1}{(c\rho)} \). The dimension of parameter \( C \)
Figure 1. Variation of temperature (a) and of normalized concentration (b) during the self-sustaining annealing without dissipation.

is kelvin and it has a physical meaning of temperature increase of an isolated piece of matter when all defects (complexes) are annealed.

From the results it follows that the development of thermal-concentration instability causes a rapid annealing of defects (propagating self-sustaining annealing of defects), when the defect density and the temperature, remaining almost constant throughout the depth of the sample, change dramatically in a narrow region that is moving with some constant average speed. In this area, the defect density falls to zero, and the temperature increases to some maximum value, which is then, taking into account the dissipation, reduced to ambient temperature.

The rapid change of the derivative of temperature on a short interval means a high contribution of thermal conductivity. Hence, in the annealing zone one can neglect the contribution of convective heat

Figure 2. Propagation of temperature (top curves) and of normalized concentration (bottom curves) fronts of self-sustaining annealing with a variable (a) and a constant speed (b), without dissipation.
flow changes in the heat transfer equation (5) as compared to the contribution of the thermal conductivity
(in the reaction zone the temperature differences are small, because of the strong dependence of lifetime
on temperature the defects are essentially annealed at a maximum temperature).

It was found that the propagation of annealing has specific features. Unlike the classical model auto-
waves (presented in figure 2b), the wave does not propagate at a constant speed for realistic values of
the annealing parameters. The speed oscillates in a complicated manner near its average value, which is
of the order of a few centimetres per second. The front of the defect concentration looks like a step or sigmoid function, while the temperature front oscillates in a complicated way in the area of the annealing,
as is illustrated in figures 1a, 1b and 2a.

Such behaviour may be associated with pre-heating of the material in the vicinity of the annealing
zone by thermal conduction. Because of the exponential dependence of the rate of annealing on the
temperature, a certain combination of parameters leads to a bifurcation and a stationary regime of heat
propagation is replaced by an oscillating one. This is supported by the fact that, for some parameters, the
temperature front also has the shape of a step or sigmoid function, and the speed of the annealing front
in this case is almost constant as illustrated in figure 2b.

In the case when heat dissipation is introduced, the temperature decreases slowly down to the
environment temperature. The temperature front of the self-sustaining annealing has the shape of a
moving hump with a steep climb and a more gentle (almost linear) descent, and not the shape of a sigmoid
function as in the system without dissipation of energy.

Due to the instability of defects, along with intensive annealing in the front of the auto-wave, a more
gradual (because of high activation energy) spontaneous annealing of defects takes place all over the
damaged volume; their density decreases and the condition for the self-sustaining defect annealing is
violated, especially if this is accompanied by heat removal.

Comparison of a traditional and of a self-sustaining propagating annealing of defects demonstrates that
the heating of a sample of finite size, to a certain temperature, by the usual heat conduction requires a larger
(asymptotically infinite) time, while heating time by propagating self-sustaining annealing is limited. Thus
the material will be exposed to an elevated temperature in a local spatial region for a significantly shorter
time, which to some extent can prevent the heat-induced phase-structural transformations of the material
and thereby prevent the change of its properties in the process of annealing. The self-sustaining annealing
of defects requires significantly less of externally supplied energy. Nevertheless, the propagation of
self-sustaining annealing of defects occurs when the stored energy of defects is sufficiently large, and
requires special conditions of initiation, which are not always achievable.

4. Conclusions
It was found that thermal-concentration instability constitutes the mechanism of spreading self-sustaining
annealing of defects, which occurs in a narrow region in a short time. It is shown that there are two
types of such annealing behaviour: in the first case the annealing propagation speed oscillates around a
constant mean value and the thermal front in the field annealing oscillates in a complicated way; in the
second case the velocity is constant, and the temperature and concentration fronts are of the form of a
sigmoid function.

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