Breaks of dose dependence of transient creep as result of competing influence of defects’ fluxes on climb of dislocations

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Abstract. In the framework of climb-glide model a theoretical approach is developed to describe transient creep under irradiation. It is obtained the explicit expression for creep rate which describes experimentally observed breaks of dose dependence of creep. It is shown that the breaks arise as result of competition of radiation and thermal fluxes of defects to dislocation. When interstitial and vacancy fluxes become equal, the dislocation cannot overcome the obstacle via climbing and cannot continue glide. Climb-glide mechanism does not contribute to the creep. The creep rate drops. Numbers of breaks depend on initial state of material and conditions of irradiation. Dose (time) of break appearance are obtained.

1. Introduction
Under loading and irradiation the resource of structural materials is largely determined by creep. New mechanisms of creep are developed under irradiation and lead to noticeable increase in creep. But it is more important there is a new behavior of creep under irradiation. Therefore often one distinguishes thermal and radiation creep [1,2].

If instantaneous deformation of the material is neglected, we can distinguish three stages of creep: transient (creep rate changes with time), stationary (creep rate is constant) and destroyed (accelerated growth of creep until destruction of material). The first two stages are the most important for problems of radiation materials science because to extend resource material we are interested in material before its destruction. In what follows we consider only the first two stages.

At high temperatures and low stresses the most part of deformation is accumulated during stationary stage of creep. When temperature decreases, the stationary stage shrinks. At not very high temperatures which correspond to operating temperatures of some nuclear power plants, the most part of deformation is accumulated during transient stage of creep [3]. Saturation of creep can be reached at large enough doses (more than hundreds dpa). In some cases self-oscillations of creep rate are observed [4,5] or the rate of creep can tend to different stationary values [6]. It means there are several different stationary regimes of creep. Sometimes the creep has only transient stage. Beside it is experimentally obtained the dose dependence of creep has complex behavior with several breaks [1,4,8-13].

In operating conditions of nuclear power plants the major contribution to the creep of irradiated materials gives "glide-climb" mechanism which is connected with dislocation motion. The motion of the dislocation combines glide and climb. The glide of the dislocation can be interrupted and blocked by an obstacle, such as clusters of impurity atoms, complexes, voids, loops, etc. In this case the dislocation can climb to a glide plane that does not intersect the obstacle and continue its glide in this plane.
Without irradiation climb of dislocations is provided by thermal vacancies. Under irradiation there is heating of the irradiated material due to relaxation of various radiation-induced excitations, for example excitations of electron and phonon subsystems. Consequently, the concentration of the thermally activated defects increases, and the creep rate increases.

However hallmark creep under irradiation is that the density of mobile radiation defects can be sufficiently high to provide climb of dislocations at virtually any temperature because vacancies and interstitials atoms are continuously generated by irradiation. Firstly, the irradiation generates along with the vacancies about the same amount of interstitial atoms. Interstitial atoms are considerably more mobile than vacancies. The activation energy of the interstitial atom is about 0.1 eV, which is several times smaller than the vacancy. Secondly, generation rate of the radiation defects do not depend on temperature. Thermal generation of vacancies increases exponentially with temperature and becomes noticeable at temperatures greater than about half the melting temperature. Interstitial atoms are not activated thermally. Third, the decreasing of the diffusion with decreasing temperature is compensated by increasing the density of defects, thus their fluxes on dislocation varies slightly. All these peculiarities influence on dynamics of point defects accumulation.

In this connection it is of interest to investigate peculiarities of the transient creep, which is connected with density of point defects and, therefore, is a function of irradiation conditions and the properties of the irradiated sample.

Here we use only one parameter of irradiation, namely defect generation rate, and don’t investigate dependence of defect generation rate on the other parameters of irradiation because it is the defect generation rate that is determined creep in framework of climb-glide mechanism. But we take into account that the defect fluxes to dislocations, and so rate of defect accumulation are functions of the temperature. Thus the creep in framework of the climb-glide model can be described by only defect generation rate, temperature and parameters of the irradiated materials.

2. Physical model and principal equations

In this paper, we develop a theoretical approach [14], which was proposed to describe the stationary creep under irradiation by quantitative way.

Under irradiation due to radiation-induced generation of a large number of interstitial atoms a dislocation can climb an obstacle by absorbing both vacancies and interstitial atoms [1,2]. When there is excess of absorption of vacancies, the dislocation overcomes the obstacle by virtue in dissolution of the extra-plane and vacancy regime of creep is realized. When there is excess of absorption of interstitial atoms, the dislocation overcomes the obstacle by virtue in expanding of extra-plane. The interstitial regime of creep takes place. To continue glide it does not matter in which way the dislocation overcomes obstacle: due to preferential absorption of interstitial atoms or vacancies. The rate of the creep must be positive any way. Thus the rate of creep is proportional to the modulus of the difference of absorption of vacancies and interstitials.

Suppose the material contains no obstacles non-radiation origin. In this case propagation and immobilization of dislocations are precluded. Thus the dislocation density can be considered as constant, and the creep rate is given by [14]

\[
\dot{\epsilon} = v[\alpha_i C_i - \alpha_v C_v]
\]  

(1)

Here \(C_i\) and \(C_v\) are the concentrations of interstitial atoms and vacancies, \(\alpha_i = z_d \rho_d D_i\), \(\alpha_v = \rho_d D_v\), \(\rho_d\) is density of dislocations. \(z_d\) is their preference coefficients. \(D_i = D_i^0 \exp(-E_i^m/kT)\) and \(D_v = D_v^0 \exp(-E_v^m/kT)\) are the diffusion coefficients for interstitial atoms and vacancies respectively. \(E_i^m\) and \(E_v^m\) are the migration energies. \(T\) is the temperature of irradiated crystal, \(k\) is Boltzmann constant. \(v=v/L/l\). Let us consider case of the constant stress when the height and concentration of the obstacles are constant parameters of model and dependence of equilibrium vacancy concentration on stress is negligible.

The system of equations for vacancies and interstitial atoms has typical form
\[
\frac{dC_i}{dt} = K - \beta_i C_i - \gamma C_i C_v
\]

\[
\frac{dC_v}{dt} = K - \beta_v (C_v - C_v^e) - \gamma C_i C_v
\]

where \( K \) is the point defect production rate. It is the main parameter of the irradiation for considered mechanisms. \( \beta_i = (z_d \rho_d + z_k \rho_k) D_i \) and \( \beta_v = (\rho_d + \rho_k) D_v \) are inverse life-times of vacancies and interstitial atoms which are determined by all (dislocation and non dislocation) sinks for defects, \( \rho_k \) is density of all sinks excluding sinks providing creep, \( z_k \) is their preference coefficients, \( \mu \) is recombination coefficients, \( \mu \) is constant. \( C_v^e = C_v^0 \exp\left(-E_v^f / kT\right) \) is the thermodynamic equilibrium concentration of vacancies. \( E_v^f \) is energy of vacancy formation. \( C_i^0 \) and \( \mu \) are material constants. Thermodynamic equilibrium concentration of interstitials is small and can be neglected. \( C_i^0 \) are initial concentrations of interstitial atoms and vacancies. Typically, the initial concentrations of defects are equal to their thermodynamic equilibrium concentrations. However, in a special way, such as pre-irradiation, the initial concentration of defects can be created different from the equilibrium. The thermal equilibrium concentration of vacancies and diffusion coefficients are exponential functions of temperature. In view of radiation heating the equation for the variation of crystal temperature has the form

\[
c_p \frac{dT}{dt} = Q + \theta_i \beta_i C_i + \theta_v \beta_v (C_v - C_v^e) + \theta_n \gamma C_i C_v - h(T - T_e)
\]

Here \( Q \) is rate of radiation heating, \( c_p \) is heat capacity of the sample per volume. Parameter \( h \) describes heat exchange between irradiated sample and environment. The environment temperature is constant and equal to \( T_e \). Heat exchange depends on its nature and geometry of the sample. Without irradiation the temperature of the sample is equal to the temperature of environment. \( \theta_i \), \( \theta_v \) and \( \theta_n \) are formation energies of one interstitial atom, one vacancy and the Frenkel pairs respectively. The rate of radiation heating is proportional to defect generation rate: \( Q = \xi \theta_n K \). Parameter \( \xi \) shows in how many times more irradiation energy goes into heating of the sample than for defect generation.

3. Results and discussion

Let consider change of creep rate from beginning of irradiation till stationary stage in qualitatively way.

During irradiation the concentrations of defects go to their stationary values. Creep tends to stationary stage, and the creep rate tends to a constant value. The stationary concentration of interstitial atoms is reached much faster because the characteristic lifetime of interstitial atoms is noticeably shorter than the lifetime of the vacancies.

In beginning of irradiation the concentrations of defects are equal to thermal equilibrium values, and glide-climb mechanism is provided by thermal vacancies. Thus this is a vacancy regime of creep. The radiation-induced vacancies and interstitial atoms appear since beginning of irradiation. The concentration of interstitial atoms is growing rapidly, concentration of vacancies - slowly. Accordingly, the flux of interstitial atoms to the dislocation grows faster and the creep rate decreases. If the generation rate of defects is large enough, at some point in time flux of interstitial atoms to the dislocation exceeds the flux of vacancies. Thus interstitial regime of creep will replace the vacancy regime. At this time, the contribution of glide-climb mechanism into the creep is absent. After that the interstitial regime of creep is developed and creep begins to increase. Increasing in the concentration of interstitial atoms is rapidly saturated, and the concentration of vacancies continues to increase. Further behavior of transient creep depends on conditions of irradiation. For example the flux
of vacancies to dislocations can again become more than flux of interstitial atoms and then exceed it. Rate of creep again becomes to equal zero and vacancy regime of creep again takes place. As the concentration of vacancies increases monotonically, and the concentration of interstitial atoms is almost constant, in further creep regime does not change. The creep will occur in the same regime as in the stationary stage. Thus, one or two dips of creep rate can take place depending on the regime of stationary creep.

Regimes of stationary creep without irradiation heating were investigated in [15] where it was shown that at low rate of the defect generation there is a vacancy regime of creep and at large - an interstitial regime. When the rate of defect generation is equal to critical value, the stationary creep rate equals zero. The critical value depends on the temperature of the sample. To account for the effect of radiation heating we take into account (4). Since the formation energy of the Frenkel pairs is approximately equal to the sum of the energies of vacancy and interstitial atom, we put

\[ v_i = v_f. \]

Then for stationary temperature of irradiated sample we obtain

\[ hK(T) = \frac{1}{K} \left( C_v^e(T) + K \right) \exp(-\beta_v t) \]

where \( \rho_i = \rho_v + \rho_k \). To obtain an expression for rate of a transient creep as a function of irradiation condition (rate of defect generation, radiation heating, temperature of environment and so on) we have to solve system (2) - (4) and to substitute obtained values as function of parameters into equation (1). The explicit solution of equations (2) - (4) can be obtained for low intensity irradiation when recombination and radiation heating are neglected. For this approximation we find

\[ T(t) = T_e \]

\[ C_i(t) = \frac{K}{\beta_i} \left( \frac{K}{\beta_i} - C_i^0 \right) \exp(-\beta_i t) \]

\[ C_v(t) = \frac{K}{\beta_v} + C_v^e - \left( \frac{K}{\beta_v} + C_v^e - C_v^0 \right) \exp(-\beta_v t) \]

To take into account recombination the equations (2) - (4) are solved numerically. The numerical solution shows that the influence of recombination leads to a no monotonic change of concentration of interstitial atoms: in the beginning it sharply increases, reaches a maximum and then decreases to a stationary value. Vacancy concentration monotonically approaches to its stationary value. Under certain conditions, the positive feedback between concentration of defects and temperature of the sample can lead to the development of self-oscillations of temperature [4,5], but here this effect is not considered.

Substitute (6) - (8) into (1) we obtain

\[ \dot{\epsilon}(t) = vK \left( \frac{\alpha_i}{\beta_i} - \frac{\alpha_v}{\beta_v} + \frac{\alpha_v C_v^e}{K} \right) \exp(-\beta_v t) \]

where \( \dot{\epsilon}_0 = K \left( \frac{\alpha_i}{\beta_i} - \frac{\alpha_v}{\beta_v} + \frac{\alpha_v C_v^e}{K} \right) \) is the rate of stationary creep.

When initial concentrations of defects are thermodynamic equilibrium values, the expression (9) is transformed into
Let rate of creep reach the first zero at time $t_1$. We can estimate its approximate value taking into account that vacancy concentration grows slower than concentration of interstitial atoms. Therefore we can put $\exp(-\beta_v t_1) \approx 1$ in (10) and find

$$t_1 \approx \frac{1}{\beta_v} \ln \left( \frac{v \rho_d \rho_i K}{\rho_i \dot{\varepsilon}_v + v \rho_d K} \right)$$

(11)

The first zero rate of creep takes place when the inequality $\alpha_i C_i^{\nu} - \alpha_v C_v^{\nu} > 0$ is satisfied.

Let us estimate approximate value of $t_2$ for which the second zero of creep rate is happen. Owing to the concentration of interstitial atoms reaches its stationary concentration much more quickly we can use stationary concentration of interstitial atoms at time $t_2$. Therefore we can put $C_i \approx C_i^{\nu}$ and $\exp(-\beta_v t_2) \approx 0$ in (10) and obtain

$$t_2 \approx \frac{1}{\beta_v} \ln \left( K \left( \beta_v C_v^{\nu} + \left( 1 - \frac{\alpha_i \beta_i}{\alpha_v \beta_v} \right) K \right) \right)$$

(12)

The second zero rate of creep takes place when the next inequalities are satisfied:

1. $\alpha_i C_i^{\nu} - \alpha_v C_v^{\nu} > 0$ (the second minimum takes place only when the first minimum takes place);
2. $\alpha_i C_i^{\nu} - \alpha_v C_v^{\nu} < \alpha_v C_v^{\nu}$, this condition coincides with the condition for the implementation of the vacancy regime of stationary creep.

Time of appearance of the first and the second dip of dose dependence was estimated for model with next parameters: $D_i^0 = 3 \times 10^{-5}$ m$^2$s$^{-1}$, $E_m^{\nu}=1.4$ eV, $C_i^{\nu}=1$, $E_f^{\nu}=1.8$ eV, $z_d=1.08$, $z_i=1$, $\mu=8 \times 10^{20}$ m$^{-2}$, $\rho_d=10^{11}$m$^{-2}$ and $\rho_i+\rho_d=2 \times 10^{11}$m$^{-2}$. These parameters are corresponded to results of experiment [8] in which annealed pure nickel (99.96%) was irradiated inside nuclear reactor. Defect generation rate was $2 \times 10^{-8}$ dpa/s, temperature of the sample was 720K. It was established experimentally that at stress $\sigma=83$ MPa the fracture of dose dependence of creep (dip of creep rate) is watched in 900 s after the start of irradiation. When the stress is $\sigma=100$ MPa the fracture is watched in 1000 s after beginning of irradiation. When temperature of irradiated sample grows, the time of the fracture appearance (it means change of creep mechanism) decreases.

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4. Conclusions

The proposed model explains the experimentally observed behavior of the transient creep under irradiation. This non-monotonic behavior is connected with change of creep regime: in the beginning of the irradiation the thermal creep with vacancy regime is changed by transient creep with interstitial regime. After that the interstitial regime is again changed by vacancy regime and creep is developed through vacancy regime till stationary stage.

When creep regime changes, contribution of "glide-climb" mechanism is absent and dose dependence of transient creep has fractures or breaks. The analytical expression for rate of the transient creep allows obtaining dependence of peculiarities of creep on conditions of irradiation and properties of the irradiated material.

The measurement of the position of creep break as function of opportune parameter gives the precision method for determination of the material parameters under irradiation.
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