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On stability of spatial distributions of crystal structure defects in irradiated high burnup UO_2 fuel

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Abstract

Conditions of Kinoshita instability development of point defects and dislocation spatial distributions in the crystal structure of UO_2 fuel are studied. As a result of the instability development, spatially non-uniform regions with increased dislocation density are formed. Closed-form expressions of instability increment and spatial scale are derived. Parameters of the instability for irradiation conditions of high burnup UO_2 fuel are obtained by means of numerical simulation. Instability development time is shown to be inversely proportional to fission rate and it increases as dislocation density decreases. Calculated values of instability spatial scale and increment are in accordance with the size of fine grains and their formation rate in the peripheral zones of high burnup LWR fuel pellets.

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1. Introduction

In LWR fuel elements at average burnups over 40 MWd/kgU notable structural changes arise in the pellet outer zone [1,2]. Original grains with the size of about 6–10 μm subdivide into submicron grains (0.1–0.3 μm) and fuel porosity in that region grows up to 20%. The fuel structure formed is called rim-structure or high burnup structure. Such microstructure changes affect fuel physical properties: porosity growth influences the thermal conductivity in the high burnup structure region and it was shown experimentally that fracture toughness of the fuel increases after restructuring [3]. General phenomenology of restructuring is also described in [4].

The restructuring process is a complex process and several possible mechanisms are activated in parallel, several different diffusions and remote forces are at work. The kind of restructuring processes called 'polygonization' in [1] is the formation of polyhedral subgrains by arising dislocation walls experimentally observed in [5] and this possibility will be discussed further.

The problem of obtaining a criterion that would determine high burnup structure formation threshold is of special interest, because it would allow to determine the dependence of the start of restructuring on initial fuel microstructure and irradiation conditions, i.e. temperature, fission rate and grain size. Proposed correlation criteria (e.g. [6,7]) are

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based on experimental data, and they show a considerable spread with respect to new experimental data. Besides, it is not clear how to extrapolate them beyond the region of parameters at which the experiments were carried out.

One of the first works dealing with examination of a possible mechanism of fuel restructuring was Kinoshita's work [8]. The author suggests using the condition of development of instability in spatial distribution of lattice defects as starting criterion of forming high burnup structure. In [8], a system of non-linear rate equations describing the variation of point defect concentrations, dislocation density and concentration of fission gas atoms coupled with vacancy clusters was studied numerically. The instability was found to develop if, at least, bulk diffusion coefficient of uranium interstitials is doubled and diffusion coefficient of uranium interstitials determining their annihilation at grain boundaries and dislocations is kept the same. Instability was numerically shown not to develop in case of equality of these diffusion coefficients. However there was no physical explanation in [8] why such a difference of uranium interstitial diffusion coefficients can arise. Also in the literature there is no closed-form solution of the stability problem of homogenous distributions of defects in nuclear fuel under irradiation conditions.

Similar instability was considered in [9] to explain a cause for dislocation periodic pattern formation in irradiated metals and alloys. However, non-linear dependences of point defects fluxes to dislocations on their concentrations were not considered in [9] due to much smaller concentrations of point defects in irradiated metals. Also, expressions for instability scales were obtained without account of mutual point defects recombination.

The goal of the present paper is to determine a possible mechanism leading to the Kinoshita instability, as this is one of the possible mechanisms of high burnup structure formation. It was shown in [10] that under fuel irradiation conditions a peak in vacancy concentration forms near an edge dislocation thus enhancing uranium interstitial recombination. Such 'shielding' of dislocation reduces the uranium interstitial flux onto dislocation thus leading to the possibility of the Kinoshita instability development. However, computations in [10] were carried out without taking anisotropy of drift and dislocation motion into account. In the present paper a closed-form study of the stability problem is carried out and dependences of time and space scales of the instability upon parameters such as dislocation density, fission rate and temperature are found.

2. Stability analysis

Considering stability of spatial distributions of defects in irradiated fuel, let us take into account the following processes: point defects generation resulting from passage of fast fission fragments and interaction with the fuel crystal structure, diffusion transport of the point defects, their recombination, their annihilation at dislocation loops and increase of dislocation density, radiation-induced resolution of dislocation loops by slowing down fission fragments. There is no special account for processes involving gaseous fission products because it allowed us to obtain closed form solution for the instability scales and also because fission gases do not play explicit role in the instability mechanism proposed by Kinoshita. This mechanism will be discussed later. Under these assumptions, similar to [8], the system describing the variation with time of averaged point defect concentration and dislocation density can be written as follows:

$$\begin{aligned}
\frac{\partial c_{\mathrm{v}}}{\partial t} &= Q - \beta c_i c_{\mathrm{v}} - J_{\mathrm{v}} n_{\mathrm{d}} + D_{\mathrm{v}} \Delta c_{\mathrm{v}}, \\
\frac{\partial c_i}{\partial t} &= Q - \beta c_i c_{\mathrm{v}} - J_i n_{\mathrm{d}} + P n_{\mathrm{d}} + D_i \Delta c_i, \\
\frac{\partial n_{\mathrm{d}}}{\partial t} &= 2\pi c_{\mathrm{disl}} a_0^2 (J_i - J_{\mathrm{v}} - P).
\end{aligned} \tag{1}$$

Here Q is the source of uranium interstitials and vacancies, proportional to fission rate $\dot{F}$, $D_{i,\mathrm{v}}$ are their diffusion coefficients, $J_{i,\mathrm{v}}$ are their fluxes to unit length of dislocation, a_0 is lattice parameter, P is the number of interstitial atoms being knocked out from unit of dislocation length per unit time by fission fragments.

Diffusion coefficients involve an athermal component, related with intermixing inside tracks of fission products [11]. The radiation-induced component of vacancy diffusion coefficient can be assessed as: $D^{\mathrm{rad}} = r_{\mathrm{tr}}^2 \nu$, where r_{tr} is the track radius, and ν is intermixing frequency, i.e. frequency of fission fragments passage through the given lattice region. In turn, $\nu = 2V\dot{F}$, where V is track volume. One can see that $D^{\mathrm{rad}} \approx 2\pi l_{\mathrm{tr}} r_{\mathrm{tr}}^4 \dot{F}$. At temperatures of the high burnup structure formation (300–600 °C) the athermal component is larger than the thermal one for vacancies and much smaller than the thermal one for uranium interstitials [11,12].

Point defect recombination occurs with a coefficient β. If determined with quasistationary uranium interstitial flux to one vacancy, β equals: $\beta = D_i a_0 Z_v$, where Z_v is a 'number of sites of interaction of uranium interstitials with a vacancy', i.e. a dimensionless geometric parameter, which is a factor in corresponding formula for reaction constant.

Regions rich with the uranium interstitials form due to the interaction of slowing down fission fragments with the fuel crystal structure. Uranium interstitials combine into dislocation loops of small size as this state is optimal with respect to elastic energy. Then loops grow due to the uranium interstitial flux. However if during the time between two consequent passages of fission fragments through this region of fuel the loop radius does not become greater than the track radius the loop will be completely dissolved by slowing down fission fragment and the process concerned will repeat. The formation of dislocation loops of sufficiently large radius is possible only if the uranium interstitial concentration is sufficiently great.

In the beginning of fuel irradiation the vacancy and uranium interstitial concentrations increase until mutual recombination and annihilation mechanisms become significant. Then the concentration of uranium interstitials in the fuel decreases considerably, due to high mobility of interstitials, compared with vacancy mobility. At large timescales one can assume equilibrium between nucleation and resolution processes for small dislocation loops. For large dislocation loops equilibrium of fluxes of point defects and radiation-induced knock-out of uranium interstitials is established. Thus under quasistationary conditions we assume that the volumetric concentration of dislocations c_{disl}, i.e. their number per unit volume, as well as their average radius are constant. Under these assumptions, one can abstract from the process of dislocation loops nucleation which takes place at the initial stage of fuel irradiation and consider the volumetric concentration of the dislocation loops specified, e.g. taken from the experimental data.

The process of radiation-induced knock-out of uranium interstitials from dislocation loop edges is characterized by a parameter P, which is the number of interstitial atoms being knocked out from unit of dislocation length per unit time. It can be estimated as follows: the track cross-section is $S = 2r_{tr}l_{tr}$, the track intersects with Sn_d dislocation loops. From one site of intersection of loop edge with track approximately $N_1 = 0.5\pi(r_{tr}/a_0)^2$ atoms are knocked-out. Thus per unit time in unit volume $2\dot{F}N_1 Sn_d = (2\pi r_{tr}^3 l_{tr}\dot{F}/a_0^2)n_d$ atoms are knocked-out. Approximately half of them which is knocked out in front of the dislocation returns to it due to drift in the field of its strain, therefore the resolution parameter is $P \approx \pi r_{tr}^3 l_{tr}\dot{F}/a_0^2$. Later the ratio P/D_v which does not depend on fission rate $\dot{F}$ at low temperatures concerned will be denoted as p, and the value $2\pi c_{disl}a_0^2$ having the dimension of a reciprocal length will be denoted as q.

Fluxes to dislocation $J_{i,v}$ are convenient to characterize with the following dimensionless parameters:

$$\phi_{i,v} = \frac{J_{i,v}}{c_{i,v}D_{i,v}}, \quad \Phi_{i,v} = \frac{1}{D_{i,v}}\frac{\partial J_{i,v}}{\partial c_{i,v}}. \tag{2}$$

In the system of Eq. (1) defect annihilation at the grain boundaries is not taken into account, as well as vacancy recombination with gaseous and solid fission products, because it is not necessary for considering of the instability development mechanism, essence of which is 'unbalancing' of point defect fluxes to dislocation. In addition, under such a simplified formulation, the problem can be solved up to closed-form dependences.

Stationary spatially uniform solution of this system conforms to flux balance condition: $J_i = J_v + P$, which is equivalent to $c_i = (\phi_v D_v c_v + P)/(\phi_i D_i)$. The system of Eq. (1) does not impose any conditions on the dislocation density n_d, therefore one needs specifying it. Since averaged point defects concentrations depend on burnup, irradiation conditions and the grain size, these should be in accordance with those taken from the non-stationary problem solution with the same parameters.

Perturbations of steady-state solution of Eq. (1) can be found as

$$\begin{pmatrix} c_v \\ c_i \\ n_d \end{pmatrix} = \begin{pmatrix} c_v^0 \\ c_i^0 \\ n_d^0 \end{pmatrix} + \xi \exp(\Gamma t + ikx), \quad \xi = \begin{pmatrix} \tilde{c}_v \\ \tilde{c}_i \\ \tilde{n}_d \end{pmatrix}, \tag{3}$$

where Γ is an increment of the instability. According to Eq. (1) linearized with respect to the small perturbations one can obtain:

$A\xi = \Gamma\xi$, where

$$A = \begin{pmatrix} -\beta c_i - \Phi_v D_v n_d - k^2 D_v & -\beta c_v & -J_v \\ -\beta c_i & -\beta c_v - \Phi_i D_i n_d - k^2 D_i & -J_i + P \\ -q\Phi_v D_v & q\Phi_i D_i & 0 \end{pmatrix}. \tag{4}$$

Thus the problem is reduced to determination of eigenvalues of the matrix A. Characteristic equation is given by

$$(-\Gamma)^3 + (-\Gamma)^2 \mathrm{tr}A + (-\Gamma)\Sigma_2 A + \det A = 0, \quad (5)$$

where tr$A$ is the trace of the matrix A (sum of its diagonal elements), $\Sigma_2 A$ is the sum of all principal minors of second-order of the matrix A and det A is its determinant. After calculations using the condition of flux balance for stationary homogenous distributions we obtain:

$$\det A = J_v D_v D_i q k^2 (\Phi_v - \Phi_i), \quad (6)$$

$$\Sigma_2 A = k^4 D_i D_v + k^2(\beta(D_v c_v + D_i c_i) + n_d D_i D_v(\Phi_i + \Phi_v)) + \Phi_i \Phi_v D_i D_v n_d^2 + \beta n_d(\Phi_i D_i c_i + \Phi_v D_v c_v) + qJ_v(\Phi_i D_i - \Phi_v D_v), \quad (7)$$

$$\mathrm{tr}A = -(\beta c_v + D_i(\Phi_i n_d + k^2)). \quad (8)$$

As it will be shown later, to find a closed-form solution of unstable mode increment ($\Gamma > 0$) it is enough to use linear approximation in Γ in the characteristic equation (Eq. (5)). However, even without finding roots one can see that instability arises only if $\Phi_i < \Phi_v$. Indeed, Viet theorem implies that the sum of all Γ (Eq. (8)) that satisfy the equation is anyway negative, and the product of roots (Eq. (6)) will be positive (it corresponds to one positive and two negative roots) only if $\Phi_i < \Phi_v$. If $\Phi_i > \Phi_v$, sum and production of roots are negative, and their pairwise-sum (Eq. (7)) is positive, i.e. all roots are negative and instability does not develop. If $\Phi_i = \Phi_v$, here exist two negative and one zero root, thus instability does also not develop.

As a result the expression for increment is given by

$$\Gamma = (k^4 D_i D_v + k^2(\beta(D_v c_v + D_i c_i) + n_d D_i D_v(\Phi_i + \Phi_v)) + \Phi_i \Phi_v D_i D_v n_d^2 + \beta n_d(\Phi_i D_i c_i + \Phi_v D_v c_v) + qJ_v(\Phi_i D_i - \Phi_v D_v))^{-1} J_v D_v D_i q k^2 (\Phi_v - \Phi_i). \quad (9)$$

Its maximal value

$$\Gamma_{max} = (2k_{max}^2 D_i D_v + \beta(D_v c_v + D_i c_i) + n_d D_i D_v(\Phi_i + \Phi_v))^{-1} J_v D_v D_i q(\Phi_v - \Phi_i) \quad (10)$$

is reached as the wave number is equal to

$$k_{max} = (\Phi_i \Phi_v n_d^2 + \beta n_d(\Phi_i c_i / D_v + \Phi_v c_v / D_i) + qJ_v(\Phi_i / D_v - \Phi_v / D_i))^{0.25}. \quad (11)$$

Calculations with parameters in Table 1 were carried out. Fig. 1 shows dispersion curve calculated from Eqs. (5) to (8) as well as dispersion curve found using the linear approximation in Γ. It is seen that in considered region of parameters one can use linear approximation in Γ.

Calculations result in the following time of the instability development and corresponding instability wavelength: $\Gamma_{max}^{-1} \approx 7 \times 10^7$ s; $2\pi k_{max}^{-1} \approx 4 \times 10^{-8}$ m. The instability development time is of the order of years and the wavelength is of order of 0.1 μm, which is not far from size of fine grains observed in polygonized regions. Hence the magnitudes of instability scales have the same orders as experimentally observed values. It is important to note that regarded model based on averaging of concentration on the scale of mean inter-dislocation

Table 1
Calculation parameters of the instability problem

T	Temperature	330 °C
$\dot{F}$	Fission rate	10^{19} fission/(m^3 s)
Q	Source of point defects	$3 \times 10^4 \dot{F} = 3 \times 10^{23}$ (m^{-3}/s)
D_i	U-interstitial diffusion coefficient [12]	$7 \times 10^{-1} \exp(-22000/T) = 10^{-16}$ (m^2/s) ($T = 330$ °C)
D_v	Vacancy diffusion coefficient [11,12]	$10^{-7} \exp(-27800/T) + 10^{-40}\dot{F} = 10^{-21}$ (m^2/s)($T = 330$ °C)
a_0	Lattice parameter	5.4×10^{-10} m
Z_v	'Number of sites' of interstitial interaction with vacancy (in recombination)	0.6
P	Knock-out parameter	$7.84 \times 10^{-13}\dot{F} = 7.84 \times 10^6$ (m^{-1}/s)
n_d	Dislocation density	2×10^{14} m^{-2}
c_{disl}	Dislocation concentration	2.27×10^{21} m^{-3} (corresponds to loop radius 1.4×10^{-8} m)
ϕ_i	Parameters obtained by use of calculations for	2.90
ϕ_v	point defects behaviour near edge	10.77
Φ_i	dislocation (defined in Eq. (2))	2.06
Φ_v		3.73
Λ	Drift length [13,14]	1×10^{-9} m

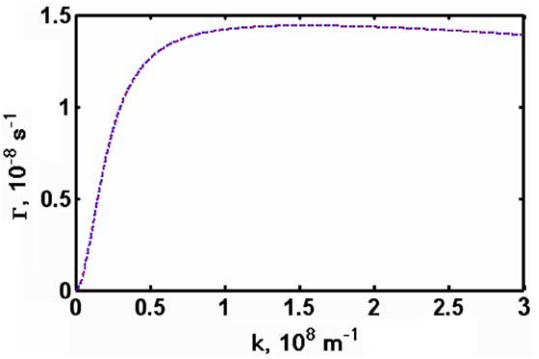

Fig. 1. Dispersion curve.

distance is valid only for perturbations with wavelength greater than this distance, i.e. there must be $k \leqslant 5 \times 10^7\ \mathrm{m}^{-1}$.

So, for instability development there must be $\Phi_i < \Phi_v$, which is in accordance with Kinoshita's calculations, [8]. The essence of the instability can be illustrated in the following way. Small growth of local dislocation density leads to growth of uranium interstitial and vacancy sink strengths and to diminishing of their local concentration. At that, diminishing of uranium interstitial local concentration hinders subsequent dislocation density growth, because they are formed by uranium interstitial atoms, and, on the contrary, diminishing of vacancy local concentration facilitates it. If, in addition, vacancy sink strength increases greater than uranium interstitial one, vacancy concentration will decrease faster than uranium interstitial concentration will increase. Thus dislocation density will increase further, which can lead to the instability development. In the following section it will be shown that one of the possible causes leading to such relation of point defect fluxes to dislocation is 'blocking' of uranium interstitial flux by peak in vacancy concentration. Near a dislocation there occurs a drift of uranium interstitials in the field of its strain, thus a region depleted with uranium interstitials exists. It leads to growth of vacancy concentration in this region, and a peak in vacancy concentration is formed. When uranium interstitials move through this peak, they recombine with vacancies and their annihilation rate at the dislocation is smaller than the annihilation rate under the conditions of the vacancy peak absence. Similar phenomenon happens when vacancies move through the region where there are less uranium interstitials than in average: vacancy sink strength increases.

Let us consider the condition of new dislocation loop formation. For very small loops of radius R_0^{disl} nucleated from uranium interstitial atoms clusters formed near fission spike regions diffusion transport predominates, therefore interstitial flux onto the dislocation loop equals: $J_i = 4\pi D_i R_{\mathrm{disl}} c_i$. Since an amount of uranium interstitial atoms in the dislocation loop is $N_i = \pi R_{\mathrm{disl}}^2 / a_0^2$ and $J_i = \dot{N}_i$, one can see that $\dot{R}_{\mathrm{disl}} = 2D_i c_i a_0^2$. Thus for the time between two consequent fission fragment interactions, the dislocation loop radius becomes: $R_{\mathrm{disl}} = 2D_i c_i a_0^2 / v$. Fission fragments will not dissolve the dislocation loop if $R_{\mathrm{disl}} > r_{\mathrm{tr}}$. Thus, the condition of increase of amount of small loops created from the uranium interstitial clusters in the fission fragments tracks has following form:

$$c_i > \frac{r_{\mathrm{tr}} v}{2D_i a_0^2} = \frac{P}{D_i}. \tag{12}$$

For the conditions given in Table 1, the threshold value is: $c_i^{\mathrm{thr}} = 7.84 \times 10^{22}\ \mathrm{m}^{-3}$, whereas calculated uranium interstitial concentration is: $c_i = 3.65 \times 10^{22}\ \mathrm{m}^{-3} < c_i^{\mathrm{thr}}$. Thus one can neglect the contribution of small dislocation loops to total dislocation loop density.

3. Numerical simulation of behaviour of point defects near a dislocation

To determine correctly fluxes of point defects to dislocations as well as their derivatives with respect to corresponding concentrations (i.e. values $\phi_{i,v}$, $\Phi_{i,v}$) it is necessary to solve the problem of behaviour of point defects near an edge dislocation.

Many processes take place near the dislocation core which are not considered 'in the average', particularly, drift in the field of strain and convective drift caused by dislocation motion. On the other hand, the problem of point defect behaviour near dislocations should be solved consistently with the averaged problem for the entire fuel grain. To simulate the behaviour of point defects near a dislocation core the following system of equations was used:

$$D_i \Delta c_i + D_i^{\mathrm{th}} \nabla c_i \cdot \nabla \frac{\Lambda \cos\theta}{r} + u \cdot \nabla c_i - z_i c_i - \beta c_i c_v + A\dot{F} + P n_d = 0. \tag{13}$$

$$D_v \Delta c_v - D_v^{\mathrm{th}} \nabla c_v \cdot \nabla \frac{\Lambda \cos\theta}{r} + u \cdot \nabla c_v - z_v c_v - \beta c_i c_v + A\dot{F} = 0. \tag{14}$$

$D^{\text{th}}_{i,\text{v}}$ are the thermal components of diffusion coefficients of uranium interstitials and vacancies, accordingly. At low temperature, appropriate to high burnup structure formation $D_i \approx D^{\text{th}}_i$, $D^{\text{th}}_{\text{v}} \approx 0$. The second terms of Eqs. (13) and (14) describe drift, and Λ is a drift length; $\Lambda = (G\Omega b)(1+\mu)/(3\pi kT(1-\mu))$ [13], where G and μ are shear modulus and Poisson's ratio of the fuel, Ω is a dilatation volume of the point defects, b is Burgers' vector of the dislocation. $z_{i,\text{v}}$ are virtual volumetric sinks of the point defects included to obtain variation of point defects fluxes to dislocation with respect to their concentrations. The last term describes the source of the point defects generated by tracks of fission products. A is a number of point defect pairs appearing on two tracks [11].

The velocity of dislocation edge motion is determined by difference of interstitial and vacancy fluxes and resolution parameter: $u = a_0^2(J_i - J_{\text{v}} - P)$. $J_{i,\text{v}}$ are the fluxes of point defects per unit of dislocation length: $J_{i,\text{v}} = a_0 D_{i,\text{v}} \int_0^{2\pi} (\partial c_{i,\text{v}}/\partial r)|_{r=a_0} \mathrm{d}\theta$.

The system of Eqs. (13) and (14) is solved inside the 2D ring domain, inner radius being the lattice parameter a_0, outer one being 'influence length' of the dislocation, which is defined as one-half of mean distance between dislocations; $R = 0.5 n_{\text{d}}^{-0.5}$.

Boundary conditions at the outer radius are chosen to make concordance with averaged problem: $c_{i,\text{v}}|_{r=R} = \bar{c}_{i,\text{v}}$, at the inner radius uranium interstitials and vacancies are considered to be absent.

Averaged problem is solved consistently with inner one

$$\begin{aligned} A\dot{F} - \beta \bar{c}_i \bar{c}_{\text{v}} - z_i \bar{c}_i - n_{\text{d}} J_i + P n_{\text{d}} = 0, \\ A\dot{F} - \beta \bar{c}_i \bar{c}_{\text{v}} - z_{\text{v}} \bar{c}_{\text{v}} - n_{\text{d}} J_{\text{v}} = 0. \end{aligned} \tag{15}$$

Derived system is quasistationary one. Without effective sinks (parameters $z_{i,\text{v}}$) its solution is in accordance with the flux balance condition: $J_i = J_{\text{v}} + P$. Effective sinks $z_{i,\text{v}}$ were included to slightly vary point defect concentrations $c_{i,\text{v}}$ and thus to obtain differential fluxes $\Phi_{i,\text{v}}$.

System of Eqs. (13)–(15) was solved numerically. The solving procedure meets a singularity near dislocation core caused by growth of interaction energy of dislocation and point defects. This complication was overcome in the following way. Since solutions of equation near a dislocation are periodic functions of angle, one can expand them into Fourier series. Then equation with two spatial variables becomes an infinite system of coupled 1D equations. To solve this system we took a finite number of angular harmonics until convergence was reached. Obtained finite system of 1D differential equations was solved using finite-difference method on non-uniform mesh. Concordance equations (by velocity and by averaged values) were solved using various iteration methods. In calculation from 5 to 15 angular harmonics were used to reach desired stability and accuracy of the scheme.

Calculations were carried out with the same values of input parameters (T, $\dot{F}$, n_{d}, Q, $D_{i,\text{v}}$, a_0, Z_{v}, P) as in the instability problem.

Calculations really show the presence of vacancy peak near a dislocation. Figs. 2 and 3 show its general view and contour plot of vacancy concentra-

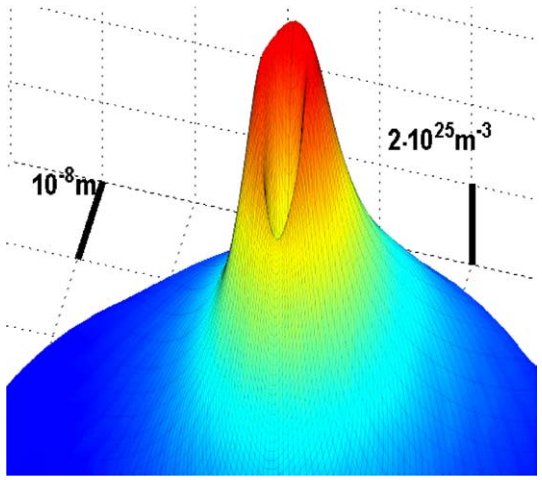

Fig. 2. Peak in the distribution of vacancies. $c_{\text{v}}^{\max} = 3.6 \times 10^{26}$ m^{-3}.

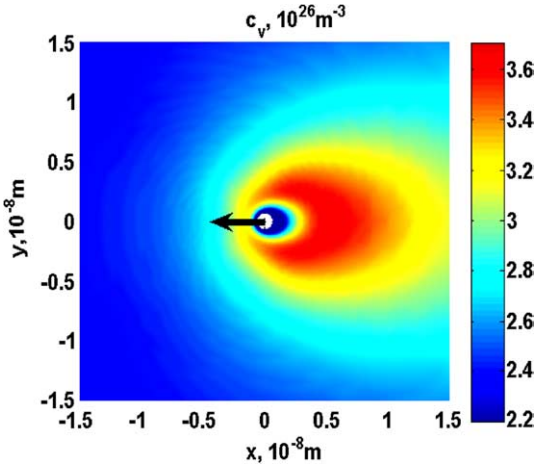

Fig. 3. Vacancy concentration contour plot.

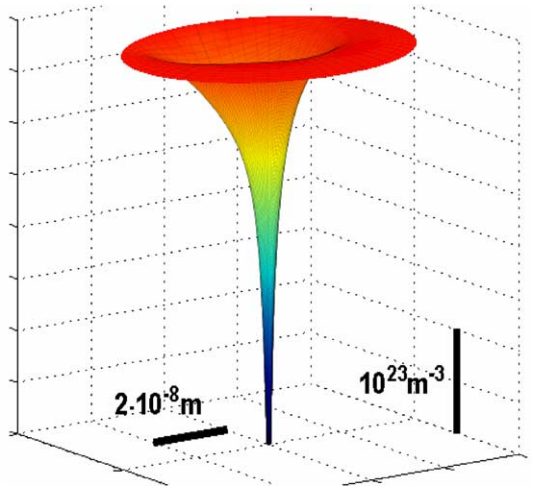

Fig. 4. Distribution of uranium interstitials near dislocation core.

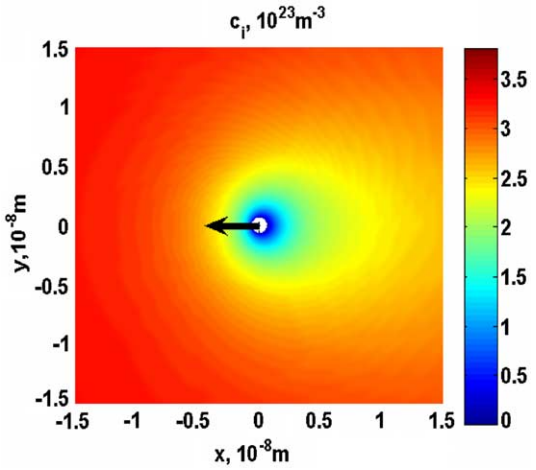

Fig. 5. Uranium interstitial concentration contour plot.

tion. One can see that, firstly, peak is anisotropic, and, secondly, is biased relative to dislocation core. Here, if uranium interstitial flux were greater than vacancy flux together with resolution, dislocation would move from the right to the left (direction of motion is indicated with an arrow), thus peak turn out to be in dislocation tail. Similar plots (Figs. 4 and 5) are presented for uranium interstitials.

Calculations were carried out for the instability increment using developed programs and analytical results of the previous section (Eq. (11)) at various values of parameters. Fig. 6 shows dependences of the increment on fission rate $\dot{F}$ and dislocation density. Resulting proportional dependence on $\dot{F}$ corresponds to burnup criterion determining polygonization start of UO_2 fuel, because instability

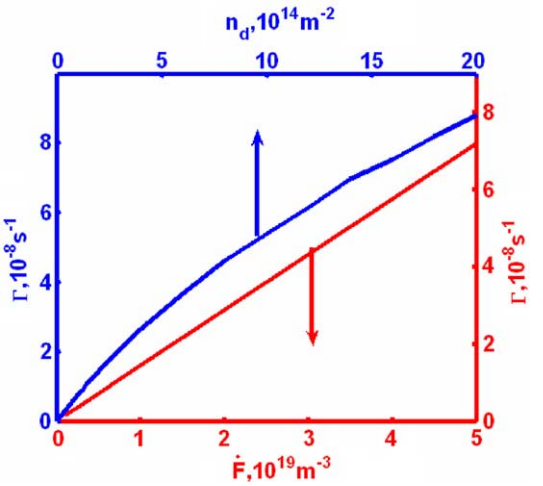

Fig. 6. Dependence of the instability increment on fission rate (lower line) and dislocation density (upper line).

development is determined by value $\dot{F}/\Gamma$, which does not depend on $\dot{F}$. The value $\dot{F}/\Gamma$ has a physical meaning similar to the threshold burnup. As dislocation density increases polygonization must start earlier, which is also in accordance with experimental observations. In this model instability increment is independent of temperature explicitly, as it can be seen from Eqs. (9) to (11), which do not involve the thermal diffusion coefficient D_i. However, as temperature increases the dislocation density decreases due to annealing (in this model the dislocation density is a free parameter), and it might prevent instability from development.

4. Conclusion

This paper deals with a study of one of the possible mechanisms of high burnup fuel polygonization, namely, development of instability in spatial distributions of crystal structure defects, proposed by Kinoshita [8]. The stability problem is examined analytically, formula of increment as well as requirement for instability development are derived. To obtain consistent values of parameters determining point defect fluxes to dislocation, a non-linear system of equations describing diffusion, anisotropic drift in the field of strain and convective drift caused by dislocation motion were numerically solved. Simulation showed that near the dislocations vacancy distribution peaks form and uranium interstitial recombination is enhanced in that region thus making Kinoshita's instability development possible. Calculations allowed to obtain consistently

instability increment dependence on various parameters, that proved to be in qualitative agreement with existing experimental observations. In future it is planned to further develop the model by accounting for gaseous fission products in UO_2 matrix and gas bubbles.

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References

[1] Hj. Matzke, M. Kinoshita, J. Nucl. Mater 247 (1997) 108.
[2] K. Nogita, K. Une, J. Nucl. Mater 226 (1995) 302.
[3] D. Baron, J. Spino, D. Papaioannou, Rim formation and fission gas behaviour: some structure remarks. In: International Seminar on Gas Behaviour in Water Reactor Fuels, France, Cadarache, 26–29 September, 2000.
[4] V.F. Chkuaseli, Hj. Matzke, ITU Report 1996 on the Modelling of Microstructure Transformations (RIM Effect) in High Burnup LWR Nuclear Fuels, private communications.
[5] T. Sonoda, M. Kinoshita, I.L.F. Ray, et al., Nucl. Insrum. and Meth. B 191 (2002) 622.
[6] T. Fujishiro, K. Yanagisawa, K. Ishijima, K. Shiba, J. Nucl. Mater 188 (1992) 162.
[7] R. Schenkel, J. Richter, D. Pel, R. Wellum (Eds.), Annual Report 1995 of Institute of Transuranium Elements, EUR 16368 EN, TUAR-95.
[8] M. Kinoshita, J. Nucl. Mater. 248 (1997) 185.
[9] C. Abromeit, H. Trinkaus, H. Wollenberger, Can. J. Phys. 68 (1990) 778.
[10] V.V. Likhanskii, O.V. Khoruzhii, A.A. Sorokin, Atomic Energy (in Russian) 96 (2) (2004) 113.
[11] Hj. Matzke, Radiat. Effects 75 (1983) 317.
[12] J.M. Griesmeuer, N.M. Ghoniem, J. Nucl. Mater. 80 (1979) 88.
[13] J. Hirth, I. Lothe. Theory of dislocations. M. Atomizdat, 1972 (in Russian).
[14] D.R. Olander. Fundamental Aspects of Nuclear Reactor Fuel Elements, TID-26711-P1, US Technical Information Center, Springfield, VA, 1976.