SECTION 1
PHYSICS OF RADIATION DAMAGES AND EFFECTS IN SOLIDS

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BIAS FOR PRISMATIC DISLOCATION LOOPS IN ZIRCONIUM. NUMERICAL ANALYSIS

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Using the analytical expression for the energy of elastic interaction of radiation point defects with a prismatic edge dislocation loop of zirconium (Burgers vector $b^D = 1/3\langle 11\bar{2}0\rangle$, $\{1\bar{1}\bar{2}0\}$ occurrence plane), the bias of loops of different nature (vacancy and interstitial) was calculated by the finite difference method. The toroidal geometry of the reservoir was used. It allowed one to calculate biases for loops of any size without any correction of the elastic field in its area of influence. In the dilatation center approximation the dependences of the loop bias on the loop radius were obtained. The principal possibility of coexistence of loops of different nature in the prismatic plane of zirconium is shown. A qualitative concept of the radiation growth (RG) mechanism was formulated within the framework of the classical elastic ideology.

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INTRODUCTION

The development of the domestic nuclear power industry at present is mainly based on the use of light-water reactors with the use of zirconium materials in the core that are low-absorbing in the thermal neutron spectrum [1, 2]. Ensuring the operational reliability, specified service life and safety of nuclear installations, taking into account economic and environmental factors, is feasible with compliance with very strict requirements for these materials, taking into account the degradation of their physical and mechanical properties under the influence of high temperatures, neutron irradiation and corrosion in the coolant. An example of such requirements is the high radiation [3] and mechanical [4] durability of the structural materials of fuel elements and fuel assemblies, in particular, their dimensional stability. This problem has many aspects. One of them is related to the radiation growth effect (RG). This term refers to the shape-change of crystalline solids under conditions of irradiation by energetic particles without the application of an external load. The volume of the material does not change, in contrast to the effect of radiation swelling. The RG phenomenon is particularly characteristic of anisotropic materials, in particular zirconium (hcp). So, the fuel cladding and fuel assemblies of all conventional nuclear reactors that generate power from the fission of uranium by thermal neutrons are made from zirconium alloys because of their low thermal neutron absorption cross-section. The stability of their dimensions, as well as the ability to predict changes in their shape, is very important to the designers and operators of such reactors, since deformation affects the operability and service life of the reactor core. Therefore, RG of zirconium and its alloys has been the subject of intensive research since the second half of the last century [5, 6]. It has been found that zirconium during growth expands in the $\langle a\rangle$-direction and shrinks along the $\langle c\rangle$-axis [7, 8]. Such its behavior is associated with the idea of Buckley S.N. [5, 6] that interstitial loops are formed predominantly on the prismatic planes of zirconium, {1010}, and vacancy loops on the basic (0001). The reason is the stresses caused by the thermal peaks of the collision cascades and the corresponding thermal expansion along the different zirconia axes. Although the physics of the RG mechanism has changed over the years, Buckley's general concept has remained the same: vacancy loops nucleating and growing on the base planes "eating" the crystal along the $\langle c\rangle$-axis, while growing interstitial loops, forming additional extra planes in the $\langle a\rangle$-direction, increase its size. Moreover, it turned out that dislocation loops of different nature (vacancy and interstitial) can coexist on prismatic planes [7, 8]. And that does not fit at all into the standard concept of the dislocation bias (EID – elastic interaction difference) [9], since it is believed [10] that the dislocation loop bias does not depend on its nature. Therefore, the joint coexistence of interstitial and vacancy loops, as well as the growth of basic vacancy loops within the standard elastic ideology seems inexplicable.

The most popular version of the cause of radiation growth of zirconium is anisotropic diffusion of radiation point defects (PD) between its planes (DAD theory – diffusional anisotropy difference) [9, 11]. Its main assumption is as follows $D_i^a / D_i^c > D_v^a / D_v^c$. Here D_m^a is the diffusion coefficient of PD of *m*-type in the basal plane of zirconium, D_m^c is the coefficient of diffusion in $\langle c\rangle$-direction (subscript v and i refer to vacancies and self-interstitial atoms (SIA) respectively). However, there is no experimental confirmation of this inequality to date. Moreover, numerical calculations [12] have shown that the inequality in the range of reactor temperatures (T < 800 K) is just the opposite

$D_i^a / D_i^c < D_v^a / D_v^c$. Therefore, a physical cause of RG associated only with anisotropic diffusion of radiation PD seems doubtful.

In [13, 14], within the framework of the classical elastic ideology (EID), the bias for basic zirconium loops (edge dislocation with the Burgers vector $\mathrm{b}^{\mathrm{D}} = 1/2[0001]$ and mixed with $\mathrm{b}^{\mathrm{DS}} = 1/6 < 20\bar{2}3 >$) of different nature was numerically calculated depending on their radius. It was shown that the existence of vacancy type loops is possible only in the presence of an uncompensated source of vacancies into the basal plane. It was suggested that such a source could be interstitial loops on prismatic planes. In order to verify it, in this work, a similar problem is solved for a prismatic edge dislocation loop (the Burgers vector $\mathrm{b}^{\mathrm{D}} = 1/3\langle 11\bar{2}0 \rangle$, the $\{11\bar{2}0\}\}$ occurrence plane [8]), using the analytical expression for the energy of its elastic interaction with the PD.

LOOP BIAS

The sink bias is determined by a relation of the form $B = 1 - Z_v / Z_i$. Here subscripts *v* and *i* correspond to vacancies and SIA respectively. If *B>0*, one says that the loop has a preference to SIA. The dimensionless quantity $Z_{v,i}$ is called the absorption efficiency of the PD by the sink. It appears as a result of calculating the PD diffusion flux to a specific sink. Under the assumption of diffusion isotropy of the medium ($D_{ij} = D\delta_{ij}$) the PD flux *J* to the dislocation loop is found by solving the following diffusion problem in its region of influence using the quasi-stationary approximation:

$$\begin{gathered} \omega\, div\, \boldsymbol{j}(\boldsymbol{r}) = 0; \quad \omega \boldsymbol{j}(\boldsymbol{r}) = -DC(\boldsymbol{r})\beta\, \nabla\mu(\boldsymbol{r}); \\ \beta \equiv 1/k_B T; \\ \beta\,\mu(\boldsymbol{r}) = \ln\left(\frac{C(\boldsymbol{r})}{C^e}\exp\left(\beta E(\boldsymbol{r})\right)\right); \\ J = -\iint_S \left[\boldsymbol{n}\, \boldsymbol{j}(\boldsymbol{r})\right] d\sigma. \end{gathered} \tag{1}$$

Here $C(\boldsymbol{r})$ is the concentration of migrating PD; $\boldsymbol{j}(\boldsymbol{r})$, $\mu(\boldsymbol{r})$ – their flux density and chemical potential, respectively; $E(\boldsymbol{r})$ – their interaction energy with the loop; C^e – equilibrium thermal concentration of PD in the crystal in the absence of a stress field E. The integral is taken over an arbitrary surface containing the loop with the outer normal $\boldsymbol{n}$. Equation (1) should be supplemented with boundary conditions. The inner surface $\mathrm{S_C}$ is usually chosen in the form of a torus containing a dislocation line. The torus minor radius r_c corresponds to the dislocation core radius. The boundary condition on it has the form:

$$C(\boldsymbol{r})\exp\left(E(\boldsymbol{r})\right)|_{S_C} = 0. \tag{2}$$

The condition has the traditional form and corresponds to the value of the PD chemical potential at the dislocation core $\mu|_{S_C} = 0$, when we neglect the linear tension of the loop and the effect of coalescence of loops of the same nature during annealing. Outer surface S_{ext} following the authors [10] we choose in the form torus coaxial with $\mathrm{S_C}$ with generating circle radius $\mathrm{R_{ext}}$ which corresponds to the radius of the loop influence region. By analogy with (2) we formulate it for the chemical potential in the form: $\beta\,\mu|_{S_{ext}} = \ln(\bar{C}/C^e)$. Here $\bar{C}$ is the average PD concentration in an effective medium that simulates the influence of all sinks. This is the standard form of the PD chemical potential in an effective medium, where the influence of a particular sink is neutralized by the others. Then:

$$C(\boldsymbol{r})\exp\left(E(\boldsymbol{r})\right)|_{S_{ext}} = \bar{C}. \tag{3}$$

The central element of system (1)-(3) is the interaction energy of PD with the loop. According to [13], in zirconium in the case of a dilatation center, it has the form:

$$E(\boldsymbol{r}) = -\Delta V\, P\, Spu_{ij}(\boldsymbol{r}),$$

$$P = \frac{2C_{13}{}^2 - (C_{11} + C_{12})\, C_{33}}{4C_{13} - 2C_{33} - (C_{11} + C_{12})}. \tag{4}$$

Here C_{ij} is the crystal elastic moduli, ΔV - the change in the volume of the finite crystal, associated with PD; $u_{ij}(\boldsymbol{r})$ – deformation field caused by the loop in the point where the PD is situated. In contrast to the basic loop (axial symmetry); there is only one option for calculating the deformation field $u_{ij}(\boldsymbol{r})$ – through the tensor Green's function $\mathrm{G_{ij}}$ (TGF) equations of equilibrium of a given elastic medium, which allows calculating the displacements created by a loop with any Burgers vector

$$u_i(\boldsymbol{r}) = C_{jklm} b_m^D \int_{S_D} n_l^D \frac{\partial G_{ij}(\boldsymbol{r} - \boldsymbol{r}')}{\partial x_k} dS'. \tag{5}$$

Here C_{jklm} is the tensor of elastic moduli of the medium, modeling crystal; $b_m{}^D$ is the *m* – component of the dislocation Burgers vector; $n_l{}^D$ is *l* – component of the normal vector to an arbitrary surface S_D, based on the dislocation line; $\boldsymbol{r}$ is the observation point coordinate; $\boldsymbol{r}'$ is the surface point coordinate S_D. TGF is calculated by the method Lifshitz-Rosenzweig [15]. For a-loop, the direction of the Burgers vector $\mathbf{b^D} = 1/3\langle 11\bar{2}0 \rangle$ and the normal $\boldsymbol{n}$ to the plane of occurrence $\{11\bar{2}0\}$ of the loop coincide, so it is natural to choose the axis *"x"* of the Cartesian coordinate system in the same direction. As a result, for the vacancy a-loop we have [12]:

$$E(\boldsymbol{r}) = \Delta V\, P \frac{b^D}{4\pi}\left\{\int_{S_D} \frac{d^2 r'}{|\boldsymbol{r} - \boldsymbol{r}'|^3} Q(\tau_3^2) + (C_{11} - C_{12}) \int_{S_D} \frac{d^2 r'}{|\boldsymbol{r} - \boldsymbol{r}'|^3} \tau_1^2 \left[3Y(\tau_3^2) + 2\tau_3^2 \frac{dY}{d\tau_3^2}\right]\right\}; \tag{6}$$

$$Q(\tau_3^2) = (1 - 3\tau_3^2)\left[C_{12}Y(\tau_3^2) + C_{13}\Psi(\tau_3^2)\right] + 2\tau_3^2(1 - \tau_3^2)\frac{d}{d\tau_3^2}\left[C_{12}Y(\tau_3^2) + C_{13}\Psi(\tau_3^2)\right] - (C_{11} - C_{12})Y(\tau_3^2);$$

$$\Psi(\tau_3^2)=V(\tau_3^2)+W(\tau_3^2);\qquad Y(\tau_3^2)=\mathrm{K}(\tau_3^2)+V(\tau_3^2);$$
$$\tau_1=x/|\boldsymbol{r}-\boldsymbol{r}'|;\qquad \tau_3=(z-z')/|\boldsymbol{r}-\boldsymbol{r}'|.$$

The functions $\mathrm{K}(\tau_3^2)$, $W(\tau_3^2)$, $V(\tau_3^2)$ are quite complicated. Their explicit expressions are given in [16]. It is important that they all depend on only one variable τ_3^2.

CALCULATION PROCEDURE

Consider a circular vacancy loop of radius R, lying in the zirconium prismatic plane ($x'=0$) of a cylindrical system coordinates (*r, φ, x*). All calculations are performed using dimensionless coordinates $r\to r/b^D$; $r^2=y^2+z^2$; $y=r\cos\varphi$; $z=r\sin\varphi$; $x\to x/b^D$; $|\boldsymbol{r}-\boldsymbol{r}'|^2=x^2+r^2-2rr'\cos(\varphi-\varphi')+r'^2$; $(z-z')=r\sin\varphi-r'\sin\varphi'$. The integration in (6) is carried out over the area of the loop. Figs. 1 and 2 for loop with radius $R=100$ in plane $x = 40$ illustrate dependency of the dimensionless energy $E/\Delta V\,P$ (6) on the azimuth angle φ for two regions: inner $r=50$ (Fig. 1) and external $r=150$ (Fig. 2). Wherein experimental values of the elastic moduli of zirconium according to [17] (Mbar) look like: C_{11} = 1.554; C_{12} = 0.672; C_{13} = 0.646; C_{33} = 1.725; C_{55} = C_{44} = 0.363.

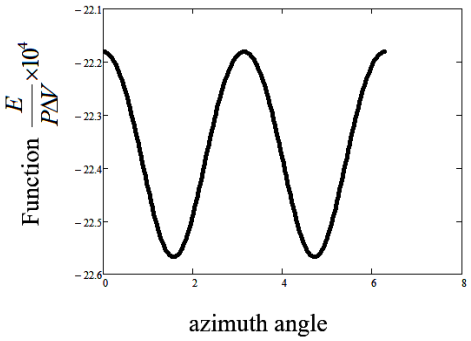


Fig. 1. Dependence of the interaction energy $E(\varphi)/\Delta V\,P$ of the vacancy loop and SIA on the azimuth angle φ for R = 100; x = 40 and the inner region of the loop r = 50

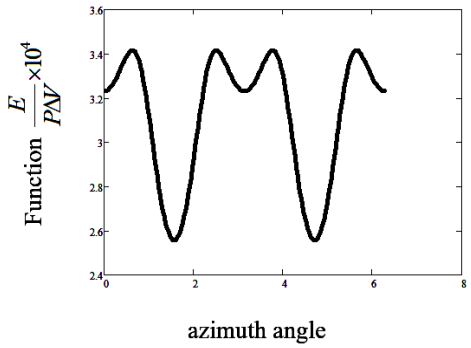


Fig. 2. Dependence of the interaction energy $E(\varphi)/\Delta V\,P$ of the vacancy loop and SIA on the azimuth angle φ for R = 100; x = 40 and outer region of the loop r = 50

Note that in (4) for zirconium, the value $P>0$. Therefore, for SIA ($\Delta V>0$), the inner region of the vacancy loop must be the attraction region ($E(\boldsymbol{r})/\Delta V\,P<0$), and the outer region, the repulsion region ($E(\boldsymbol{r})/\Delta V\,P>0$). It is this behavior of energy that takes place in Figs. 1, 2. For a vacancy ($\Delta V<0$), the illustration will be reversed. We note a very weak dependence on the azimuth angle φ, which does not change the nature of the interaction (sign) in each region. The interaction changes sign only when passing from the inner region of the loop ($r=50$) to the outer one ($r=150$). However, this dependence greatly complicates the calculations for numerical calculations. Therefore, as in [14], we will eliminate it by averaging the right side of (6) over the azimuthal angle φ, making the problem isotropic in the "*yz*" plane. And one more note. In the expression for energy (6), the dependence on the variable "*x*" is quadratic, i.e. replacement $x\to -x$ doesn't change anything. Therefore, as in the case of the basic loop, $\mathrm{b}^{\mathrm{D}}=1/2[0001]$ numerical calculations can be carried out only in one part of the half-space $x>0$.

In terms of a variable $\psi(r,x)=C(r,x)\exp E(r,x)/\bar{C}$ the diffusion problem (1)–(3) in dimensionless cylindrical coordinates taking into account isotropy after averaging has the form:

$$\frac{\partial^2\psi}{\partial r^2}+\frac{\partial^2\psi}{\partial x^2}+\left(\frac{1}{r}-\frac{\partial E}{\partial r}\right)\frac{\partial\psi}{\partial r}-\frac{\partial E}{\partial x}\frac{\partial\psi}{\partial x}=0 \qquad (7)$$

with boundary conditions $\psi(r,x)=0$ on the inner toroidal surface

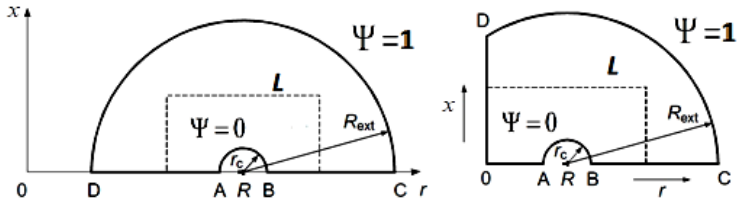
$$R-r_c\le r\le R+r_c, \qquad (8)$$

$\psi(r,x)=1$ on the outer toroidal surface $R-R_{ext}\le r\le R+R_{ext}$ for $R>R_{ext}$; $0\le r\le R+R_{ext}$ for $R<R_{ext}$.

Then for the flux and absorption efficiency $Z_{v,i}$ we have:

$$J=2\pi R\frac{D\bar{C}}{\omega}Z\left(r_c,R,R_{ext}\right),$$
$$Z\left(r_c,R,R_{ext}\right)=\frac{1}{2\pi R}\iint_S\exp\left(-E(r,x)\right)\left[\boldsymbol{n}\nabla\psi(r,x)\right]d\sigma. \qquad (9)$$

The diffusion problem (7), (8) was solved numerically by the finite difference method. Fig. 3 shows a cross section of a toroidal reservoir containing a loop [14], taking into account the reflection symmetry in the plane $x=0$ and symmetry (после усреднения по φ) about rotation around the x-axis. The specified symmetry imposes additional boundary conditions: $\partial\psi/\partial x=0$ on DA, BC, OA, corresponding to zero flux through the plane $x=0$, and $\partial\psi/\partial r=0$ on DO (axis of symmetry). An arbitrary inner surface *S* in (9) is chosen for the convenience of calculations in the form of a rectangle of rotation. In Fig. 3, this is the contour *L*.



a *b*

Fig. 3. Coordinate system for a toroidal reservoir: a – $R>R_{ext}$, b – $R<R_{ext}$

RESULTS

Fig. 4 shows the dependences of the bias of the prismatic loops of zirconium B on their radius (in b^D) for three values of the outer size of the toroidal reservoir: $R_{ext} = 200\, b^D$, which corresponds to the dislocation density $\rho \approx 8\cdot10^9$ cm^{-2} (see Fig. 4,a), $R_{ext} = 120\, b^D$ ($\rho \approx 2\cdot10^{10}$ cm^{-2}, see Fig. 4,b) and $R_{ext} = 60\, b^D$ ($\rho \approx 8.4\cdot10^{10}$ cm^{-2}, see Fig. 4,c). Curves with a maximums refer to interstitial (●) loops, with a minimum to vacancy (■).

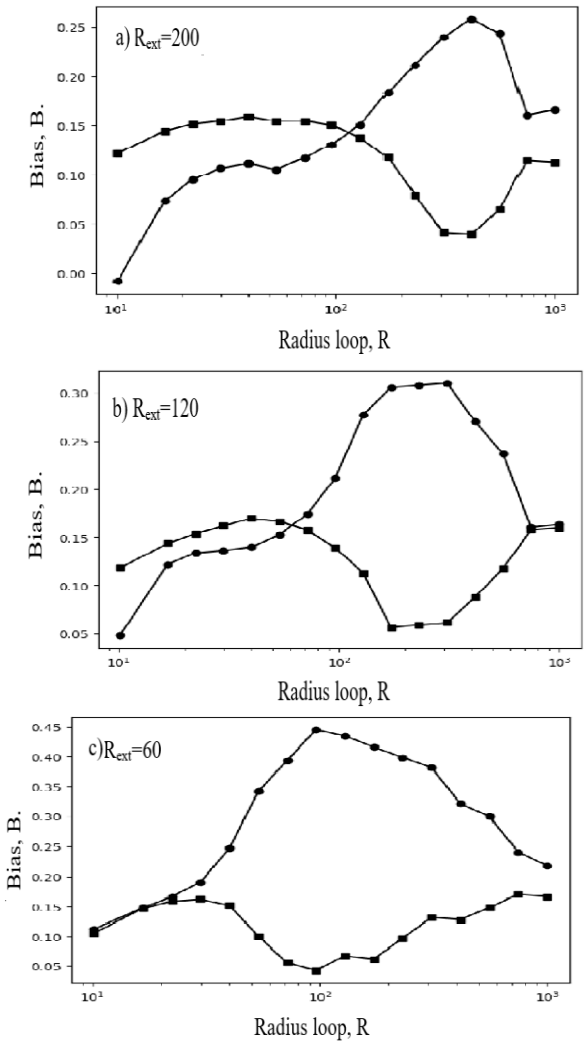


Fig. 4. Dependences of the bias of the prismatic loops of zirconium, B, on their radius for three values of the outer size of the toroidal reservoir:
a – $R_{ext} = 200\, b^D$; b – $R_{ext} = 120\, b^D$; c – $R_{ext} = 60\, b^D$.
"●" – refer to interstitial loop; "■" – to vacancy

First, it should be noted that prismatic dislocation loops in zirconia are biased sinks, which absorb SIAs more efficiently than vacancies, since $B>0$ for both types of loops (see Fig. 4). The bias depends on the loop radius and the sink density. And this is consistent with the general conclusions [10]. However, in [10] there is an important conclusion that the bias does not depend on the nature of the loop. In our case, this is not the case.

Secondly, and this is the most important, there is always a region in space of the size, in which the bias of interstitial loop B_{int} (●) is higher, than bias vacansion one B_{vac} (■). Those, in this region, SIAs are mainly absorbed by interstitial loops. The remaining in excess vacancies can be absorbed by the existing vacancy loops or migrate to the basal plane and contribute to the nucleation and further growth of vacancy basis loops. In our opinion, this makes it possible in principle to explain two experimentally observed points: the joint coexistence of loops of both types in the prismatic plane of zirconium, as well as the nucleation and subsequent growth of vacancy loops in the basal plane. Note that the lower boundary of this region $B_{int}(R^*) = B_{vac}(R^*)$ moves towards smaller sizes with an increase in the total dislocation density ρ (or a decrease in R_{ext}). So, at $\rho \approx 2\cdot10^{10}$ cm^{-2} (see Fig. 4,b) $R^* \approx 60b^D$ or ≈ 20 nm. And these are quite real loops visible in a microscope. In the region $R < R^*$ the existence of loops within the framework of EID is difficult to explain, since here $B_{int} < B_{vac}$. Those SIAs should be predominantly absorbed by vacancy loops and dissolve them. Excess vacancies should, accordingly, dissolve the emerging interstitial loops. Therefore, this area is the subject of research in the theory of nucliation. Thus, the following picture emerges. At a low dislocation density (the initial stage of irradiation), the loops in the prismatic plane cannot grow due to their large value R^* (see Fig. 4,a). As the dislocation density increases, it moves towards lower values. There is a real possibility of diffusion growth of emerging interstitial loops. And they, in turn, stimulate the nucleation and growth of vacancy loops, first on the prismatic and then on the basal planes of zirconium. However, here we must also include the bias of the basis loops and only then formulate a qualitative possible RR mechanism based on the classical elastic ideology. In conclusion, we note that understanding the physics of the RR mechanism may be useful for predicting the behavior of fuel cladding during long-term storage of spent fuel in dry reservoir.

SUMMARY

1. Prismatic loops in zirconia are biased sinks, which absorb SIAs more efficiently than vacancies, since for both types of loops (see Fig. 4) $B > 0$.

2. The principal possibility of coexistence of loops of different nature in the prismatic plane of zirconium is shown.

3. A qualitative concept of the RG mechanism was formulated within the framework of the classical elastic ideology.

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ФАКТОР ПЕРЕВАГИ ПРИЗМАТИЧНИХ ДИСЛОКАЦІЙНИХ ПЕТЕЛЬ У ЦИРКОНІЇ. ЧИСЕЛЬНИЙ АНАЛІЗ

О.Г. Троценко, А.В. Бабіч, П.М. Остапчук

Використовуючи аналітичний вираз для енергії пружної взаємодії радіаційних точкових дефектів із призматичною крайовою дислокаційною петлею цирконію (вектор Бюргерса $b^D = 1/3\langle 11\bar{2}0\rangle$, площина залягання $\{11\bar{2}0\}$) методом кінцевих різниць пораховано фактор переваги петель різної природи (вакансіонної та міжвузлової). Використовувалася тороїдальна геометрія резервуара, що дозволяє провести розрахунки для петлі будь-якого розміру без будь-якої корекції пружного поля в її області впливу. У наближенні центру дилатації отримані залежності фактора переваги петель від їхнього радіусу. Показано важливу можливість спільного співіснування в призматичній площині цирконію петель різної природи. Сформульовано якісну концепцію механізму радіаційного зростання (РЗ) у рамках класичної пружної ідеології.