Modeling of Processes in Austenitic Steel Produced under Irradiation in Fast Reactors and Possibilities of Model Practical Application

A.V. Kozlov[1], I.A. Portnykh[1]

[1]Joint Stock Company "Institute of Nuclear Materials" (INM), Zarechny, Russia

E-mail contact of main author: kozlov_av@irmatom.ru

Abstract. Descriptive models of different stages of structural changes in austenitic steels, radiation-induced swelling in particular, as well as the effect of structural changes on physical and mechanical properties of steels are given. Their application to predict changes in structure and material properties for claddings from BN-600 core is shown.

Key words: modeling of processes in fast reactor materials, radiation-induced swelling, physical and mechanical properties, prediction.

1. Introduction

Nowadays austenitic stainless steels are used as a cladding material for BN-600 and BN-800 reactors. Examinations carried out at the end of fuel element service life give the information about cladding state. On the basis of the examination it is necessary: 1 – to determine residual life and 2 – to find the way for its extension.

Extrapolation methods are generally used for the first aspect. The results obtained for fuel elements of different fuel assemblies at attaining different damage doses and fuel burn-ups are used. As a rule the results are limited by linear extrapolation. The prediction accuracy is not high for several reasons. The initial state of claddings from different lots (and casts) is not the same; therefore there is some error even when processes of properties changes depend linearly on irradiation parameters (for example, damage dose). Moreover, dose and temperature dependence of some processes, radiation-induced swelling in particular, is quite nonlinear. Therefore linear extrapolation is unacceptable. Extrapolation used for the second aspect almost gives no results as the characteristics to be correlated are not defined.

At INM a technical description of the processes occurring in metal materials under irradiation has been developed for a long time. The key concept is in description of point defect (vacancies and interstitials) formation and further microstructural changes, which are determined by the formation intensity, migration and interaction with other microstructural elements (impurity atoms, dislocations, grain boundaries and other sinks). A machine for quantitative description of point defect migration and concentration has been designed and is used for austenitic steels.

Based on the developed theoretical concepts different stages of structural changes, radiation-induced swelling in particular, as well as the effect of structural changes on physical and mechanical properties, have been modeled. These models are used to predict changes in material structure and properties of claddings operated in BN-600 reactor core.

The paper aims to show developed at INM models of changes in austenitic steel structure and properties under irradiation in fast reactors and to demonstrate their application for BN-600 reactor claddings.

2. Principles of Point Defect Migration Model

The given migration model is based on the principles of static thermodynamics of solids [1]:

- point defect migration is accomplished by their jumps to one of the nearby positions,

- probability of the jump in a certain direction in case of equal positions point defect is determined by the following equation $\exp(-E_m / kT)/6$ (where E_m is energy of point defect migration, k is Boltzmann constant, T is temperature, *K*),

- if the jump is between positions with different energies, probability of ω_{12} jump from E_1 -energy position to E_2-energy position is expressed as follows

$$\omega_{12} = A \cdot \exp((E_1 - E_2)/kT) \cdot \exp(-E_m / kT), \tag{1}$$

where *A* is normalization constant defined if total probability of the jump to all the nearby positions is equal to 1.

In uniform matrix (equal probability of jumps in all directions) time $t = \tau'$ (hereinafter migration time), required for distancing from the initial position s, can be expressed [1] as follows

$$\tau' = \left(\frac{s}{a}\right)^2 \cdot \frac{6}{\nu} \cdot \exp\left(\frac{E_m}{kT}\right) \quad , \tag{2}$$

where ν is Debye frequency representing atom fluctuation,

a is distance between nearby positions of a point defect, here defined as a lattice parameter.

Point defect migrates along uniform and isotropic crystal matrix with nonuniformity and anisotropy developed only near the sink. This approximation replaces smoothly changing nonuniform strain field by uniform matrix with potential drops at sinks. Probability of point defect emission and absorption by the sink from adjacent area is defined regarding relation (2) and normalization conditions.

The given approximations are enough to describe point defect migration to sinks: grain boundaries, dislocation and recombination.

3. Principles of Radiation Defect Formation and Evolution Model

Under neutron irradiation point defects are formed in displacement cascades and during formation of single Frenkel pairs as well. Relative part of point defect formed these ways depends on neutron irradiation spectrum. Fraction of point defects left in displacement cascades is ~0.2 [2]. Relative part of point defects formed as single Frenkel pairs is up to 50 % of total defect number for thermal reactors, and only about 10 % for fast reactors [3].

During operation of structural systems inside BN-600 reactor core temperature of structural materials is in the range between 370 and 650 °C (643-723 K). At these temperatures dissociation of generated vacancy clusters is rather fast and is in direct proportion to the temperature. At standard displacement rate of ~1×10^{-6}dpa/s, after a rather short period of time since irradiation has started, steady-state cluster concentration is settled, so the number of vacancies in them is the same. Intensity of point defects entering crystal matrix is expressed by displacement rate, cascade efficiency, correlation of the number of point defects generated in cascades and single Frenkel pairs, coefficient of interstitial cluster formation in cascades [3].

Using the equation for the rate of point defects entering matrix and description of the intensity of their transition to all sinks, and regarding thermal point defect generation, the equations for vacancy and interstitial atom concentration in irradiated materials are obtained.

It is found it takes a short period of time in claddings made of austenitic steels to settle steady-state vacancy and interstitial concentration, which slowly varies with varied sink characteristics for point defects. *Table I* gives calculated time to settle steady-state vacancy and interstitial concentration in ChS-68 steel, used as a cladding material during BN-600 reactor operation [4].

TABLE I. TIME TO ATTAIN QUASI-EQUILIBRIUM VACANCY AND INTERSTITIAL CONCENTRATION IN COLD WORKED CHS-68 STEEL UNDER NEUTRON IRRADIATION WITH POINT DEFECT GENERATION RATE OF 1×10-6 DPA/S.

Time to attain steady-state concentration, s	Irradiation temperature, K				
	573	673	773	783	973
for interstitials	1.4×10^{-5}	5.8×10^{-6}	3.0×10^{-6}	1.8×10^{-6}	1.2×10^{-6}
for vacancies	1.7×10^{3}	5.1×10^{1}	3.4×10^{0}	4.3×10^{-1}	8.2×10^{-2}

Hereinafter the results for ChS-68 claddings, which composition is given in *Table II*, are used to demonstrate application of the designed device.

TABLE II. CHEMICAL COMPOSITION OF ChS-68 CLADDINGS [5].

Alloy	Element, wt. %								
	C	Cr	Ni	Mo	Mn	Si	Ti	P	B
ChS-68	0.07	16.5	15.0	2.3	1.7	0.36	0.36	-	0.003

It is clear the time to settle steady-state concentration of point defects in austenitic cladding steels is negligibly small, as opposed to operating time (~2 years). Therefore steady-state concentration of point defects is settled from the beginning of irradiation. For intertitials it is by several orders of magnitude smaller than for vacancies.

Calculated values of steady-state vacancy and interstitial concentration in ChS-68 at the initial stage are given in *Table III*.

The results of the effect of irradiation with significantly higher displacement rate on the material are used to predict irradiation effect during operation of structural elements. It enables prediction of structural material behaviour when high damage dose is attained. However, there is no clear evidence that the effect of irradiation to similar damage doses at different displacement rates is the same. Moreover, there is experimental evidence of the opposite. Therefore, an irradiation temperature shift is introduced for simulating process, though it has no theoretical base, and certain empirical references are used.

TABLE III. VALUES OF STEADY-STATE VACANCY AND INTERSTITIAL CONCENTRATION AT THE INCUBATION SWELLING STAGE IN COLD-WORKED CHS-68 STEEL UNDER IRRADIATION IN REACTOR AT RADIATION DAMAGE RATE OF 1×10-6 DPA/S [4].

Defect type	Temperature, K			
	573	673	773	873
Vacancies	4.7×10^{-7}	1.3×10^{-8}	1.4×10^{-8}	1.0×10^{-7}
Interstitials	1.2×10^{-15}	5.4×10^{-16}	4.0×10^{-16}	1.7×10^{-15}

The developed model enables defining steady-state point defect concentrations that influence all the processes of structural evolution at different displacement rates. *Fig. 1* shows dependence of the calculated steady-state vacancy concentrations in ChS-68 steel irradiated at different displacement rates on irradiation temperature [4]. The range of displacement rates includes different irradiation conditions: from vessel materials for thermal reactors to irradiation in heavy ion colliders. With irradiation temperature increase steady-state vacancy and interstitial concentration decreases to values close to thermally equilibrium concentration, and then they increase together. The temperature, when steady-state and thermally equilibrium vacancy concentrations become almost equal, is taken as maximum swelling temperature – T_{sh}.

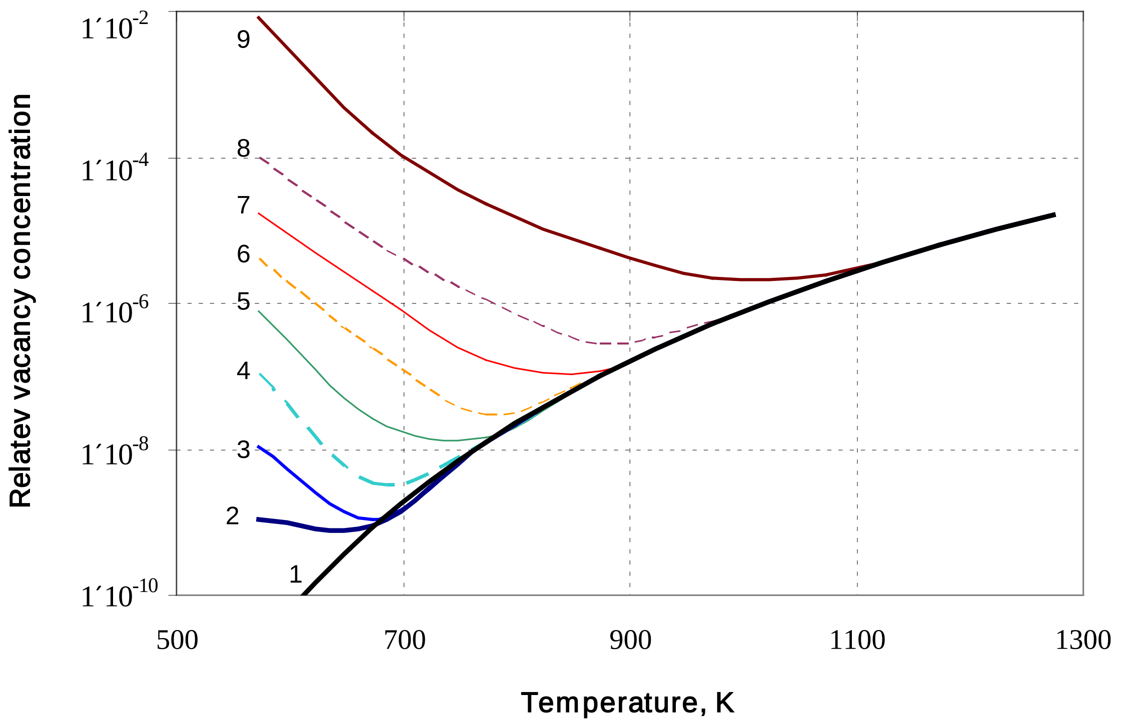
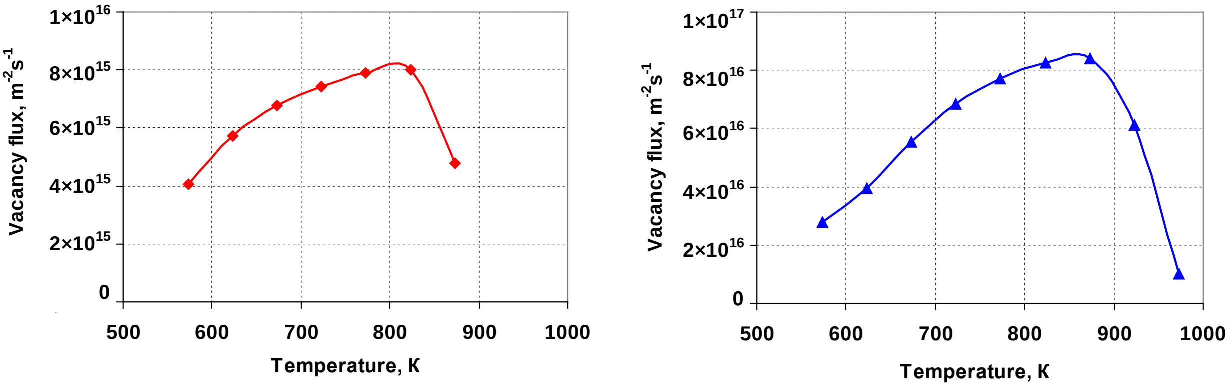


FIG. 1. Irradiation temperature dependence of steady-state vacancy concentration in ChS-68 steel at different displacement rates: 1 – thermally equilibrium concentration, 2 – 10^{-10}dpa/s, 3 – 10^{-9}dpa/s, 4 – 10^{-8}dpa/s, 5 – 10^{-7}dpa/s, 6 – 10^{-6}dpa/s, 7 – 10^{-5}dpa/s, 8 – 10^{-4}dpa/s, 9 – 10^{-2}dpa/s.

With increasing displacement rate T_{sh} moves to high-temperature area. Maximum swelling temperature increases as well. It helps to estimate temperature shift during irradiation simulation with higher displacement rate.

Swelling description in terms of the applied model is a nontrivial task as far as this is a multiple-stage process, with each stage requiring a vast number of parameters to be used. A simplified estimation can be obtained by using the relation for unbalanced vacancy flux – difference in vacancy and interstitial flux to the sink, and vacancies vaporized from the sink. The calculations show the maximum temperature depending on displacement rate. *Fig. 2* gives the dependence for ChS-68 steel under irradiation at displacement rates of $1{\cdot}10^{-5}$ and $1{\cdot}10^{-6}$ dpa/s. Displacement rate increase causes maximum temperature increase by an order of magnitude, up to ~40 K.



a b

Fig. 2. Temperature dependence of unbalanced density of vacancy flux into the void in ChS-68 claddings under neutron irradiation at displacement rate:
a – $1{\cdot}10^{-6}$ dpa/s, b – $1{\cdot}10^{-5}$ dpa/s.

Crystal matrix supersaturation with vacancies and interstitials induces development of a number of processes causing structural changes. Interstitial atoms form dislocation interstitial loops. Vacancy supersaturation promotes vacancy voids to generate and grow. However it is not a trivial process. Using analysis of growth conditions for a small void carried out at INM, an equation enabling calculation of critical diameter of vacancy void nucleus has been obtained. Solution of an equation shows that vacancy void smaller than critical size will dissolve. Critical diameter value for vacancy void nucleus depends on structural and physical characteristics of material, temperature, vacancy and interstitial concentration [6]. The latter depends on displacement rate as well. In order to grow vacancy void has to exceed estimated critical size.

The number of vacancies left in subcascades is not enough to form a critical vacancy void nucleus. It is found that formation of gas vacancy bubbles under neutron irradiation is a dominant mechanism of void nuclei formation. Transmutation reactions resulting in helium and hydrogen generation are gas suppliers in steel. Characteristics of helium and hydrogen mobility and interaction with vacancies and dislocations show that helium, combining with excess vacancies into clusters soon after its generation, plays a dominant role. Migrating along the crystal helium-vacancy clusters go to dislocations and other sinks, where they form helium-vacancy bubbles growing equilibrium. Their growth is maintained by introducing helium with production rate by 6 times less than vacancy generation rate [7].

Reaching critical size of helium-vacancy void nucleus (less than critical size of vacancy void nucleus) a void grows due to unbalanced vacancy flow introduced into it. This point indicates the end of the incubation stage and the beginning of the unsteady-state swelling stage. neutron Irradiation temperature dependence of critical diameter of helium-vacancy void nucleus for ChS-68 steel at displacement rate of $1{\times}10^{-6}$dpa/s is given in *Fig. 3*.

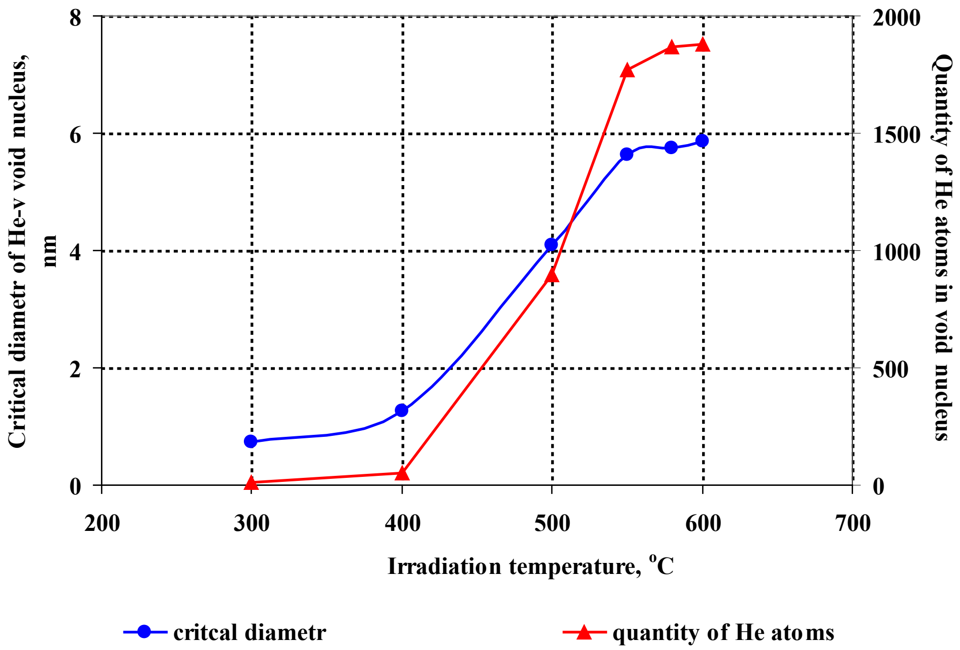


FIG. 3. Irradiation temperature dependence of critical diameter of helium-vacancy void nucleus and helium concentration in it [6].

Reaching critical size of void nuclei they start growing intensely due to unbalanced vacancy flow introduced into them. Void-size distribution bar charts show that along with maximum of small 2-3 nm voids the second maximum of larger voids appears. In the course of time (dose) this maximum moves to larger sizes and grows (in relative units), *Fig. 4*.

Void size and integrated surface area increase along with vacancy flow into voids (proportional to their surface area). Swelling rate increases. However, as the swelling increases, so does the probability to merge (void coalescence, if voids roved with experimental observations. Every void coalescence act causes reduction of their surface area.

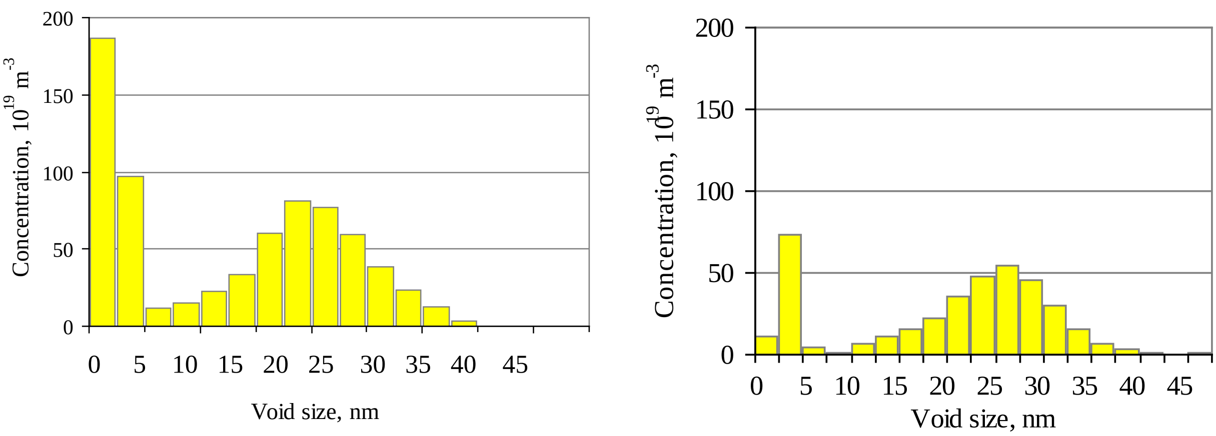


FIG. 4. Bar chart of void distribution in cladding specimens from areas of low-temperature irradiation 443±3 °C (716±3 K):
a – D=69 dpa, swelling of 2.2 %, b – D=76 dpa, swelling of 2.9 %) [8].

As a result of two competing processes – void surface growing due to introduction of vacancies into them, and reduction of their integrated surface area due to coalescence – a dynamic equilibrium is settled with constant integrated surface area. *Fig. 5* gives experimentally made curve of swelling value dependence of integrated surface area for different irradiation temperatures [9], saturated at swelling of ~9 % (as it is theoretically shown).

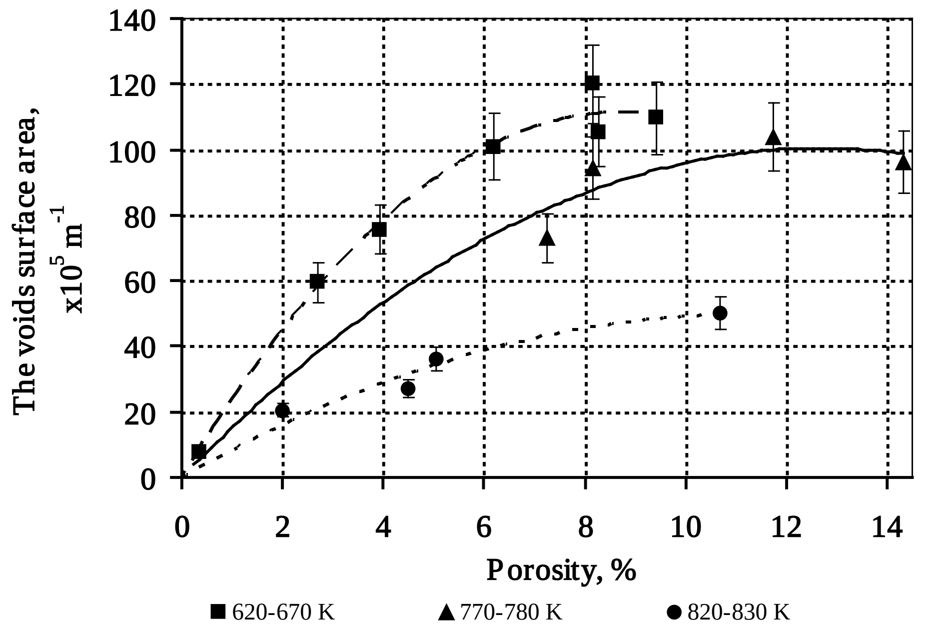


FIG. 5. Dependence of integral area of void surface on porosity value in ChS-68 steel at different irradiation temperatures in the range between 410 and 560 °C.

The steady-state swelling stage (with constant rate) begins at saturation of integrated void surface area. To calculate this rate it is necessary to know steady-state vacancy and interstitial concentration. As opposed to the beginning of irradiation, the rate changes significantly due to expansion in the number of sinks for point defects. Voids are the main sinks now. Boundaries of second phase precipitates, generated in vast numbers, have an effect on the process as well. The estimation of steady-state vacancy and interstitial concentrations regardless of second phases shows that at the stage of swelling with constant rate steady-state concentration of point defects decreases almost by an order of magnitude, as opposed to the beginning of irradiation (*see Fig. 6*).

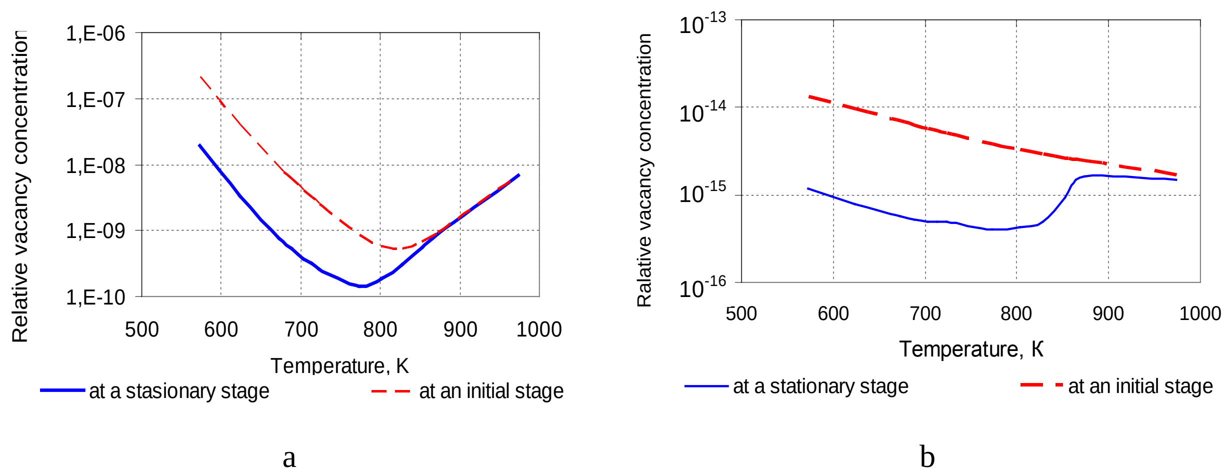


FIG. 6. Irradiation temperature dependence of point defect concentration in ChS-68 steel at the steady-state swelling and initial stages: a – for vacancies; b – for interstitial atoms [10].

Using these values irradiation temperature dependence of dose swelling rate of ChS-68 steel for displacement rate of 1×10^{-6}dpa/s has been obtained (*see Fig. 7*).

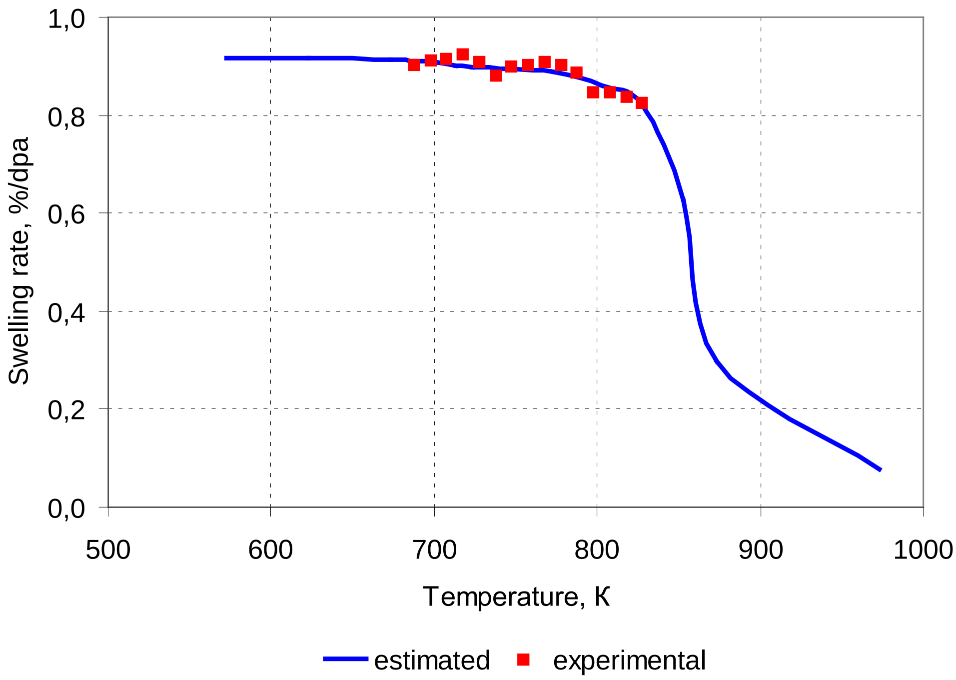


FIG. 7. Irradiation temperature dependence of dose swelling rate of ChS-68 steel for displacement rate of 1×10^{-6}dpa/s [10].

It appears that in temperature range between 573 and 828 K swelling rate changes slightly and is from 0.92 to 0.82 %/dpa. This explains an assumption based on experimental observations that a steady-state swelling rate for austenitic steels is ~1%/dpa and does not depend on temperature. The obtained estimation results adjust value for steady-state swelling rate and describe its dependence on structural characteristics of irradiated material. Further irradiation temperature increase causes sharp decrease of steady-state swelling rate.

4. Conclusion

Processes in austenitic steels under neutron irradiation have been modeled, including:

- a model of point defect migration,

- a model of point defect formation and evolution under neutron irradiation,

- a model of vacancy void nuclei growth,

- a model of gas-vacancy void nuclei formation in austenitic steels under neutron irradiation and their growth to critical size,

- a model of the incubation swelling stage,

- a model of void coalescence,

- a model of the steady-state swelling stage with constant swelling rate.

Using the developed models and basing on the obtained experimental results, the following is found:

- steady-state concentration of point defects is set in austenitic steels under short-term (from hundreds of seconds to centiseconds for vacancies, and 10^{-5}-10^{-6} seconds for interstitial atoms) irradiation in fast reactors,

- steady-state vacancy and interstitial concentrations for ChS-68 steel are estimated for different irradiation temperatures and displacement rates,

- formation of gas-vacancy bubbles and their growth to critical size is a dominant mechanism of void formation in austenitic steels under irradiation,

- critical size of gas vacancy void nucleus is estimated for ChS-68 and EK-164 steels,

- nuclei growth to critical size finishes the incubation swelling stage and marks the beginning of the unsteady-state swelling stage with swelling at increasing rate,

- integrated void surface area increases due to void coalescence and reaches saturation at swelling of ~9 %, thereby causing the beginning of the steady-state swelling stage,

- steady-state swelling rate is estimated for ChS-68 steel under neutron irradiation,

- dependence of steady-state swelling rate on irradiation temperature is established,

- in temperature range between 573 and 828 K dose swelling rate changes slightly from 0.92 to 0.82 %/dpa, further temperature increase causes its sharp decrease,

- dependence of steady-state swelling rate on steel structural characteristics is established.

5. References

[1] ZHIRIFALKO, L. Statistical physics of solids. Moscow. Mir, 1975, 432 p. (in Russian).

[2] DEVYATKO, Yu.N., PLYASOV, A.A., ROGOZHKIN S.V. Efficiency of defect generation under cascade-forming irradiation. Izvestiya RAN, Physics series, 2006, Vol. 70, No. 8, 1231-1234 (in Russian).

[3] KOZLOV, A.V. Radiation defects in austenitic steels under neutron irradiation and their effect on physical and mechanical properties. Izvestia visshikh uchebnikh zavedeniy, Yadernaya energetika, 2011, No. 1, 196-210 (in Russian).

[4] KOZLOV, A.V. Dependence of point defect concentration in ChS-68 austenitic steel on their generation rate and irradiation temperature. Phys. Met. Metallogr., 2009, Vol. 107, No. 6, 574-581 (in Russian).

[5] TSELISHCHEV, A.V., AGEEV, V.S., BUDANOV, Yu.P. et al. Development of structural steel for fuel pins and assemblies of fast sodium reactor. Atomnaya Energiya, 2010, Vol. 108, No 4, 217-22 (in Russian).

[6] KOZLOV, A.V., PORTNYKH, I.A., BLOKHIN, A.I., BLOKHIN, D.A., DEMIN, N.A. The dependence of critical diameter of void nuclei in ChS68 austenitic steel on temperature of neutron irradiation in the model of formation of helium-vacancy bubbles. Inorganic Materials Applied Research, 2013, Vol. 4, No. 3, 183-188.

[7] KOZLOV, A.V., PORTNYKH, I.A. Migration and concentration on dislocations of transmutation helium in austenitic steels under neutron irradiation. Phys. Met. Metallogr., 2016, Vol. 117, No. 4, 399-406.

[8] PORTNYKH, I.A., PANCHENKO, V.L. Characteristics of radiation porosity and structural phase state of 07C-16Cr-19Mo-2Mo-2Mn-Ti-Si-V-P-B steel after irradiation at temperatures 440 and 600 °C to damage doses of 36...94 dpa. Phys. Met. Metallogr., 2016, Vol. 117, No 6, 611- 623.

[9] PORTNYKH, I.A., KOZLOV, A.V. Correlation of radiation-induced swelling rate with radiation void growth and coalescence. Problems of Atomic Science and Technology, Materials Science and New Materials, 2008, Issue 2(71), 3-13 (in Russian).

[10] KOZLOV, A.V., PORTNYKH, I.A. Dependence of steady-state swelling rate for claddings made of ChS-68 steel on neutron irradiation characteristics. Phys. Met. Metallogr. 2016, Vol. 117, No 8, 871-874.