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Temperature effect on characteristics of void population formed in the austenitic steel under neutron irradiation up to high damage dose

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Abstract

Radiation-induced porosity in fuel pin cladding of the BN-600 reactor fabricated of cold-worked austenitic steel 16Cr–15Ni–2Mo–2Mn irradiated to different damage dose 20–90 dpa at 410–600 °C has been examined by transmission electron microscopy. Formation and growth of various types of voids were shown to occur according to their both duration and mechanism of nucleation. Dependencies of average diameters and concentration of all void types on neutron irradiation damage dose were plotted for various temperature ranges. The change of void population with increasing dose at various temperature ranges was analyzed based on point defect kinetic. The contribution of different types of voids to swelling was examined.

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1. Introduction

The study of radiation-induced swelling is needed both for the physics of radiation damage and in applied engineering. Empirical formulas do not often give predictions applicable to new experimental data. The causes may arise from formation and growth of voids depending on different factors, and from experimental data which are insufficiently detailed. The intensity of void formation and growth depends on both their nucleation mechanism and characteristics of point defect diffusion generated by irradiation.

The purpose of this work was to obtain quantitative characterization of radiation-induced void populations in austenitic steel and to find out dependencies of their formation and growth on temperature and neutron irradiation dose.

2. Material and test method

Fuel pin cladding fabricated of cold worked 16Cr–15Ni–2Mo–2Mn has been examined after operation in the BN-600 reactor to different damage doses. The chemical composition of the steel is listed in Table 1. Cladding of irradiated fuel elements with the fuel removed were sectioned and submitted to transmission electron microscopy. Each set of cut specimens was selected such that they corresponded to certain swelling temperature ranges, i.e., maximum swelling 450–480 °C, average swelling 500–510 °C and low swelling (low-temperature 410–420 °C and high-temperature 550–560 and 590–600 °C).

Microstructural examinations were performed by transmission electron microscopy methods on a JEM-2000 EX under an accelerating voltage of 200 kV. Histograms of void size distribution were plotted as a sum of single log–normal distributions. Each type of void corresponded to a single distribution determined by a χ^2 criterion [1]. The parameters of the void size distributions were used to plot the dose dependencies of average size and concentration of each void type. The

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Table 1
Chemical composition of ChS-68 according to TU 14-3-1511-87 requirements

Element	C	Cr	Ni	Mo	Mn	Si	Ti	V	B	N	Co	P	S
wt%	0.05–0.08	15.5–17.0	14.0–15.5	1.9–2.5	1.3–2.0	0.3–0.6	0.2–0.5	0.10–0.30	0.001–0.003	<0.02	<0.02	<0.02	<0.012

test technique has been described in detail by Portnykh et al. [2].

3. Results

Voids were shown to fall into three main types in all specimens. Each type corresponds to its own log–normal distribution. Below we use a terminology from a previous paper [2]. The voids of a-type – large voids are not associated with precipitates, b-type voids – average voids are mainly associated with radiation-induced phases; c-type voids – small voids are not associated with precipitates. One of the typical histograms and corresponding size distribution are shown at Fig. 1.

Fig. 2 demonstrates dependencies of the average size of voids and their concentration on damage dose for different irradiation temperature ranges.

One can see that in all cases, the average size of voids increased with dose. Void concentration varied less systematically. The concentration of all types of voids grows up to some dose and then begins to decrease. This dose depends on irradiation temperature and type of voids. The concentration of a-type voids begins to decrease earlier, especially at high irradiation temperatures.

The characteristic feature of voids at the higher irradiation temperature range 580–595 °C is that voids are spaced non-uniformly. Areas practically free of voids can be seen as well as areas with a lot of large and small voids.

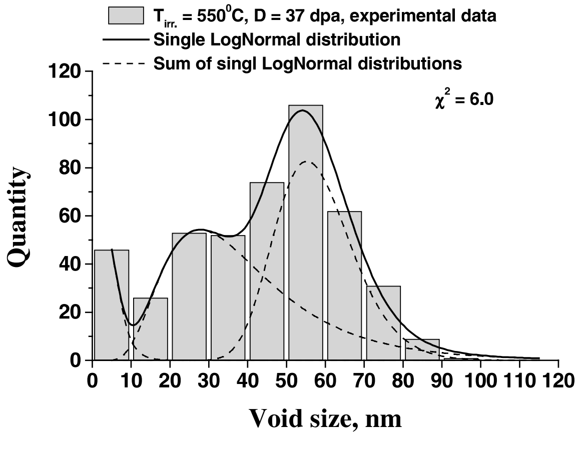

Fig. 1. Histogram of size distribution of voids in the specimen irradiated to 37 dpa at 550 °C.

4. Discussion

For void nucleation, it is necessary to reach a definite critical vacancy concentration depending on temperature and nucleation size of a void, according to the results of statistical–thermodynamic analysis given in [3]. Some period precedes the onset of vacancy void formation, and corresponding to a swelling incubation dose. Moreover the incubation dose is different for various void types. Void formation begins earlier at larger nucleation rate and lower damage dose [3]. Homogeneous void formation may be exhibited as void growth on vacancy complexes (three-vacancies or divacancies) formed randomly. Nucleus size is small and void formation from them requires more time to accumulate a higher concentration of vacancies in the matrix.

A variety of different types of voids depends on their formation time, nucleation process and growth kinetic [2]. Realization conditions of void growth are determined by initial structure and material composition as well as by neutron irradiation temperature. Voids of the a-type are formed heterogeneously. The voids are observed especially at an early irradiation stage and they are seldom associated with precipitates. The presence of voids of a-type depends on dislocation structure. Formation of b-type voids is mainly associated with radiation-induced phases. Voids of this type are smaller than a-type voids and the majority of them adjoin G-phase precipitates in the steel. The composition of G-phases is: 57Ni–21Si–7Fe–7Cr–5Mo–5Mn–4Ti, 42Ni–17Si–17Ti–9Fe–8Mo–4Cr–3Mn. Their formation mechanism may be connected with migration of 'vacancy-impurity' complexes [4]. Both vacancies forming a void and impurity atoms (Si, Ni) necessary for G-phase formation are simultaneously delivered during agglomerations of the complexes. Voids of c-type probably form due to realization of homogeneous mechanism inside a cascade region (a vacancy cluster) where vacancy concentration is higher than that in the matrix and formation of a vacancy complex is more probable. The scheme of various types of void formation is shown in Fig. 3.

The dynamics of growth of voids is determined by an integral flux of point defects into a void (I_+) which is the

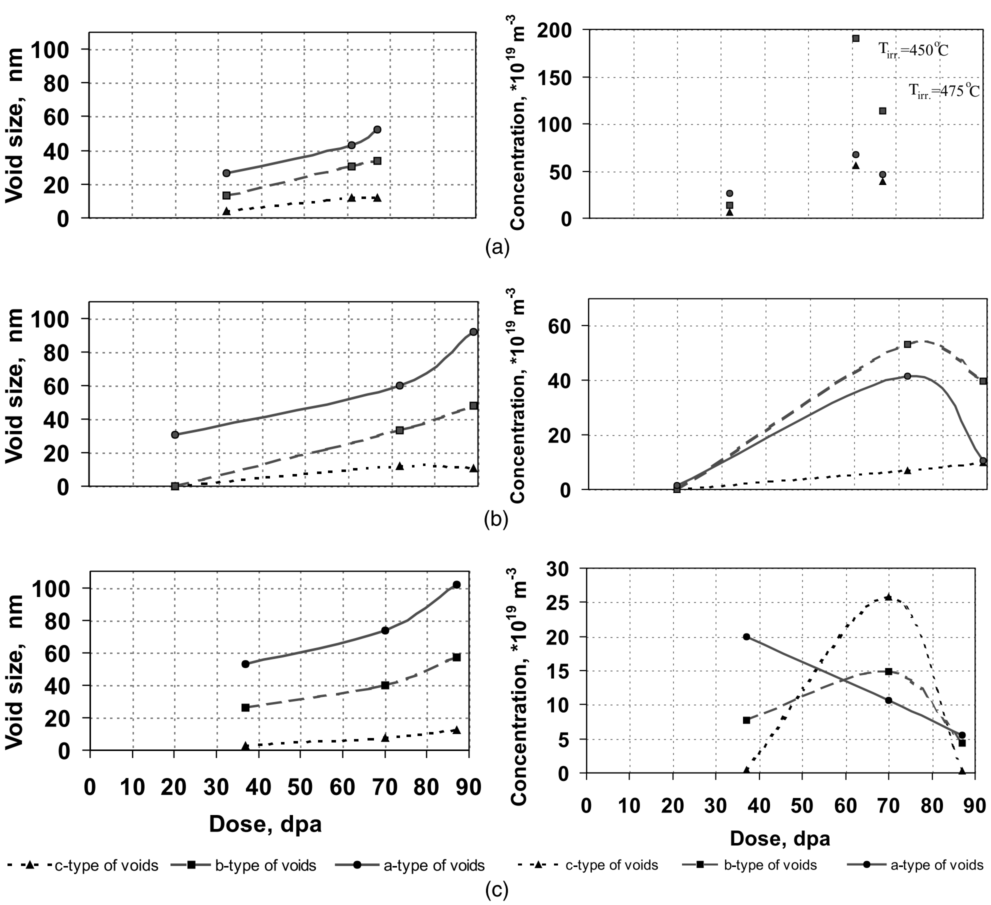

Fig. 2. Dependencies of average size of different types of voids and their concentration from damage doses at irradiation temperature ranges 450–480 °C (a) 500–510 °C (b) and 550–560 °C (c).

difference between a flux of vacancies absorbed by the void (I_{v+}), and both a flux of vacancies evaporated from the void (I_{v-}) and interstitials absorbed by the void (I_{i+}).

$$I_+ = I_{v+} - I_{v-} - I_{i+}. \qquad (1)$$

We have obtained the formula for growth rate (v_n) of a void radius using the expressions [3].

$$v_n = dr_n/dt = I_+ \Omega dt = \Omega \beta c_v \exp(-E_{mv}/kT), \qquad (2)$$

$$\beta = c_v - 0.2 \exp\left(\frac{U}{kT}\right) - c_i \exp\left(\frac{E_{mv} - E_{mi}}{kT}\right), \qquad (3)$$

where $U = 2\Omega\tau/r - E_v$; E_v is the energy of vacancy formation; r_n is void radius; E_{mv}, E_{mi} are migration energy of vacancies and interstitials respectively; c_v, c_i concentration of vacancies and interstitials in a matrix, in relative units; k is the Boltzmann constant; T is the temperature in Kelvin; U is the energy change during absorption of a vacancy by a void; Ω is the atomic volume.

One can see that the growth rate of a large void is higher than a small one. That is why a value U decreases with increasing radius, therefore it decreases the second term in the expression (2). The value β increases. The larger the void size and the higher vacancy concentration, the lower the dependence of the expression (2) on radius. This is in a good agreement with experimental data.

Since at some dose, void concentration is reduced and the growth rate of large voids is sharply increased, this may be connected with both an exhaustion of nucleation (the formation rate of new voids of the given type decreases) and coalescence of voids, for which the probability is increased with increases in void concentration and size. Let us consider how coalescence of voids influences change in their concentration. Void coalescence is taken to be a process by which a void boundary during its growth touches upon another void, so a coalesced void is formed. When a large void (a-type) absorbs small voids (e.g. c-types), it leads to a decreasing concentration of the second type. When voids of a-type

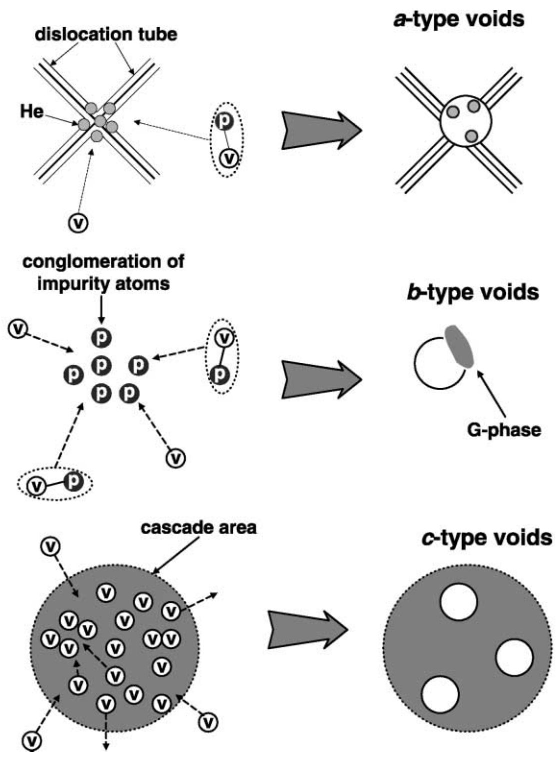

Fig. 3. The scheme of void formation of various types.

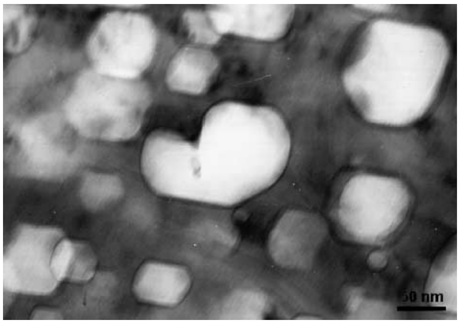

Fig. 4. Coalescence of two voids in the specimen irradiated to 72 dpa at 510 °C.

coalesce among themselves their concentration is decreased and simultaneously larger voids are formed. An example of coalescence is shown in Fig. 4.

To approximate this, we have used an approach which considers voids of one and the same type to have an identical average size, increasing in time due to mainly vacancy absorption. Moreover we have considered that voids are spaced homogeneously.

Voids of i-type are increased when they coalesce with j-type voids. The amount of voids of j-type coalesced with i-type voids per a time unit due to an increase of the i-type has the following expression:

$$\dot{n}_{\mathrm{ji}} = \mathrm{d}n_{\mathrm{ji}}/\mathrm{d}t = C_{\mathrm{j}}C_{\mathrm{i}}4\pi(r_{\mathrm{j}} + r_{\mathrm{i}})^2\,\mathrm{d}r/\mathrm{d}t, \tag{4}$$

where C_{i}, C_{j} are the concentrations of i-type and j-type voids considerably.

Using this approach, equations for the rate of reduction of different types of void concentration due to coalescence were obtained. The void concentration rate reduction for specimens irradiated to 70 dpa was calculated and compared with experimental data (Table 2). The concentration of a-type voids decreases due to coalescence after 70 dpa, according to these calculated results, at irradiation temperatures 450–480 °C. The concentration continues to grow in the temperature range 500–510 °C at 70 dpa. This corresponds to experimental data (see Table 2).

For practical application, it is worth knowing what type of voids mainly contribute to swelling. Fig. 5 exhibits histograms of the relative contribution of each type of voids to swelling. One can see that a-type voids provide the main contribution to swelling at low doses (to ≃40 dpa). The relative contribution of b-type voids is increased with increasing dose. The contribution of c-type voids is small at temperatures higher than 500 °C and it becomes appreciable at ≃60 dpa at lower temperatures.

5. Conclusions

The nucleation and growth of voids, distinguished by their formation mechanism, have been observed in

Table 2

Calculation of reduction rate of void concentration of different type due to coalescence and experimental values of intensity of their formation, $\mathrm{m}^{-3}\,\mathrm{s}^{-1}$, at irradiation to 70 dpa

Void type	450–480 °C		500–515 °C	
	Rate of concentration reduction due to coalescence	Intensity of formation	Rate of concentration reduction due to coalescence	Intensity of formation
a	8.23E+18	8.00E+18	3.63E+18	9.00E+18
b	1.29E+19	2.80E+19	5.07E+18	1.10E+19
c	1.28E+18	3.00E+18	2.17E+17	1.60E+18

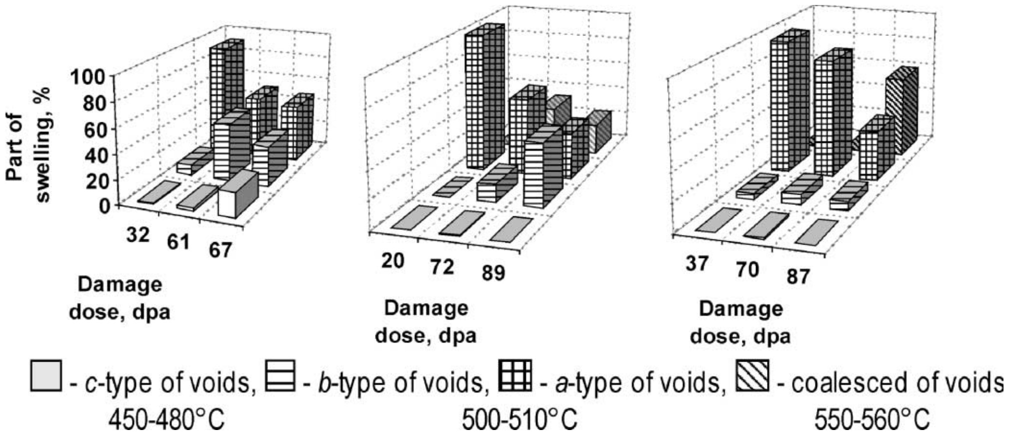

Fig. 5. A fraction of swelling brought by voids of various types for different temperature ranges.

Fe–16Cr–15Ni–2Mo–2Mn (20% c.w.) under neutron irradiation at 410–600 °C.

The largest a-type voids are associated with the characteristics of the dislocation structure. The formation of b-type voids is mainly associated with radiation-induced phases. Voids of c-type are formed due to a homogeneous mechanism.

The concentration of all types of voids increased with increasing of damage dose up to some threshold value. When void nucleation is exhausted and the concentration and size of voids becomes considerable their concentration begins to decrease. Smaller doses are necessary to reduce void concentration of the larger voids.

The average size of all types of voids increases with dose, and the growth of large voids is faster than small ones. This agrees with the equation for void growth rate obtained within the framework of statistical thermodynamics.

Voids of a- and b-types provide the greatest contribution to swelling. Moreover the larger the damage dose, the larger the contribution of the b-type voids. Contribution of voids of c-type to swelling is small at irradiation temperatures higher than 500 °C.

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