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Research Article

A study into the structure and physical properties of the Cr18Ni9-grade steel following long-term irradiation as part of the BN-600 reactor internals*

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Abstract

The microstructures and physical properties of the austenitic Cr18Ni9-grade steel after 22 and 33 years of operation as part of the reactor internals were tested for assessing the conditions of the BN-600 reactor non-replaceable components (internals) and the potential of their subsequent use in predicting the reactor ultimate life. The paper presents histograms of the porosity distribution depending on the void size, in samples taken from portions that were subjected to neutron irradiation with displacement rates ranging from 1.0×10^{-9} to 4.3×10^{-8} dpa/s at temperatures from 370 to 440 °C. The elasticity characteristics were measured by resonance-type ultrasonic technique for the samples taken from the same portions of material. It was demonstrated that swelling calculated using the histograms of the porosity distribution depending on the void size has the maximum value at ~415 °C and after 33 years of irradiation reaches values of ~3%. Long-term variations of Young's modulus demonstrate non-monotonous dependence on the damage dose. The maximum relative variation of Young's modulus after 22 and 33 years of operation does not exceed 2% and 6%, respectively, of the values corresponding to the initial state. It was shown that along with the irradiation-induced swelling the changes in the physical properties are also affected in the process of irradiation by other structural changes and, in particular, by the formation of secondary phases. As shown by the results of the studies, operation of the BN-600 reactor internals made of Cr18Ni9-grade steel can be extended beyond 33 years of service. The comparison of the results obtained for the material after 22 and 33 years of operation contains information required for describing subsequent changes of the structure and properties of the Cr18Ni9 internals. The obtained results can be used for forecasting the reactor ultimate life within the framework of existing and developed models.

Keywords

Internals, austenitic steel, neutron irradiation, porosity characteristics, irradiation-induced swelling, elasticity characteristics

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Introduction

Austenitic steels of Cr18Ni9 type tending to the formation of alpha-phase caused by radiation deformation (Kursevich et al. 2012) are used as the materials of elements of fast and thermal nuclear reactor internals. A series of radiation induced processes resulting in the radiation swelling and transformations of phase composition of steels which causes degradation of physical and mechanical properties develop under the effects of reactor irradiation (Margolin et al. 2009) thus restricting the reactor ultimate life.

The purpose of the present study was to assess the conditions of non-replaceable elements (reactor internals) of the BN-600 reactor made of Cr18Ni9 after long-term operation and to obtain results for their subsequent use in forecasting the ultimate reactor life.

Investigated material and research methodologies

Investigation was performed using samples made of Cr18Ni9 prepared from fragments of BN-600 reactor internals after long-term irradiation. Chemical composition of Cr18Ni9 steel is presented in Table 1 (Kozlov et al. 2009). Two batches of samples were investigated: the first batch was irradiated during 22 years and the second one was irradiated during 33 years. The rates both of generation of displacements per atom and of irradiation temperatures were determined by calculation methods without experimental verification; uncertainties of their determination were not evaluated. Radiation porosity characteristics were quantitatively determined according to the results of electron microscopy studies using experimentally plotted distributions of voids according to their sizes. In particular, average void sizes, void concentrations, specific integral surface area of the voids and swelling were determined in the process using the methods described in (Portnykh and Panchenko 2016). Besides the above elasticity characteristics of the material measured by resonance-type ultrasonic technique were determined for the same samples.

Irradiation parameters are presented for the investigated samples in Table 2 (Technical Certificate of the Beloyarsk NPP 2016). The range of neutron irradiation doses was equal for samples in the first batch from less than 1dpa to 21 dpa (displacements per atom) at temperatures from 370 °C to 425 °C, and that for samples in the second batch varied from less than 1 dpa to 33 dpa at temperatures from 370 °C to 440 °C. It was assumed that conditions of samples irradiated during 33 years can be regarded as subsequent evolution of conditions of samples after 22 years of irradiation (Technical Certificate of the Beloyarsk NPP 2015). The following circumstances should be taken into consideration in such analysis:

- Irradiation parameters for samples from different batches are not completely identical; in particular, the rates of generation displacements per atom differed by up to 1.5 times at close irradiation temperatures;
- Calculated irradiation parameters correspond to really observed ones with some uncertainty;
- Original conditions of the material are similar within the accuracy of initial non-homogeneity of the material.

Table 1. Chemical composition of Cr18Ni9 steel (Kozlov et al. 2009).

Element	C	Mn	Si	P	S	Ni	Cr	Cu	Fe
Concentration, weight %	0.09	1.36	0.38	0.025	0.02	8.75	17.66	0.21	The rest

Table 2. Irradiation characteristics of the investigated samples.

Batch	Sample No.	Irradiation temperature, °C	Rate of displacement per atoms, $\times10^{-8}$ dpa/s	Damage dose, dpa
22 years	22-1	368	0.4	1.7
	22-2	380	2.1	9.9
	22-3	394	2.6	12.4
	22-4	414	3.4	16.2
	22-5	426	4.3	20.5
33 years	33-1	368	0.1	>1
	33-2	374	1,6	12.8
	33-3	377	3.1	24.4
	33-4	415	4.1	33.1
	33-5	433	3.0	23.6
	33-6	442	1.6	12.6

Experimental results

It was established by transmission electron microscopy studies (TEM) that voids are observed in all irradiated samples of both batches. Pictures of small-size voids formed in the samples irradiated at temperature of ~370 °C to damage doses of ~1 dpa after 22 years and 33 years of irradiation are presented in Figure 1a, b. Pictures of voids formed in the samples irradiated at temperature of ~415 °C to damage doses of ~1b dpa after 22 years of irradiation and of ~33 dpa after 33 years of irradiation are shown in Figure 1c, d. High concentrations of small and large size voids uniformly distributed over the sample volume are observed. Pictures of voids formed in the samples irradiated at temperature of ~425 °C to damage doses of ~22 dpa after 22 years of irradiation and at temperature of ~430 °C to damage dose of ~24 dpa after 33 years of irradiation are given in Figure 1e, f. The observed voids are uniformly distributed over the sample volume with concentrations smaller than those for the preceding temperature range.

Typical histograms of voids distribution according to void sizes (Portnykh and Kozlov 2002) in the investigated samples are presented in Figure 2. The histograms have two, in some cases three maxima: the first pronounced maximum with average void size of 2–3 nm and the second diffuse maximum within the range of void sizes from 10 to 20–50 nm. Conventionally voids were divided into two groups: “small voids” associated with the first

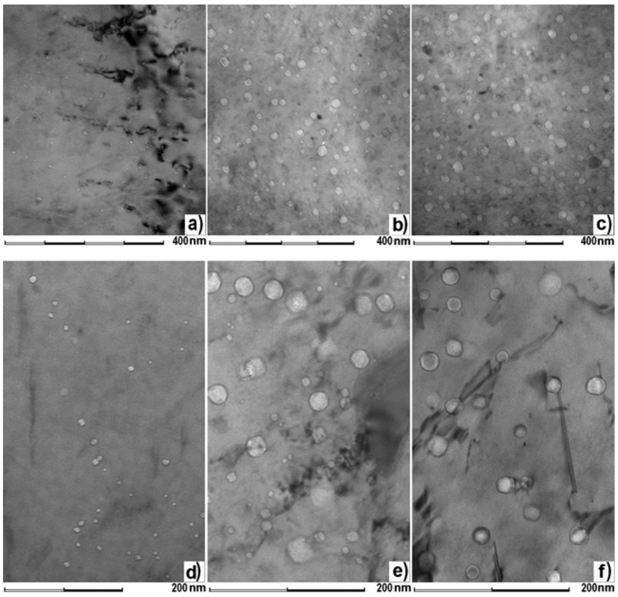

Figure 1. Vacancy pores in the samples after irradiation during 22 yearsт (a,b,c) and 33 yearsт (d,e,f) to damaging doses of ~1 dpa ate temperature of ~370 °C (a,d), to damaging doses of ~16 dpa (b) and ~33 dpa (f) at temperature of ~415 °C, to damaging dose of ~21 dpa (c) at ~425 °C and to damaging dose of ~24 dpa (f) at ~430 °C.

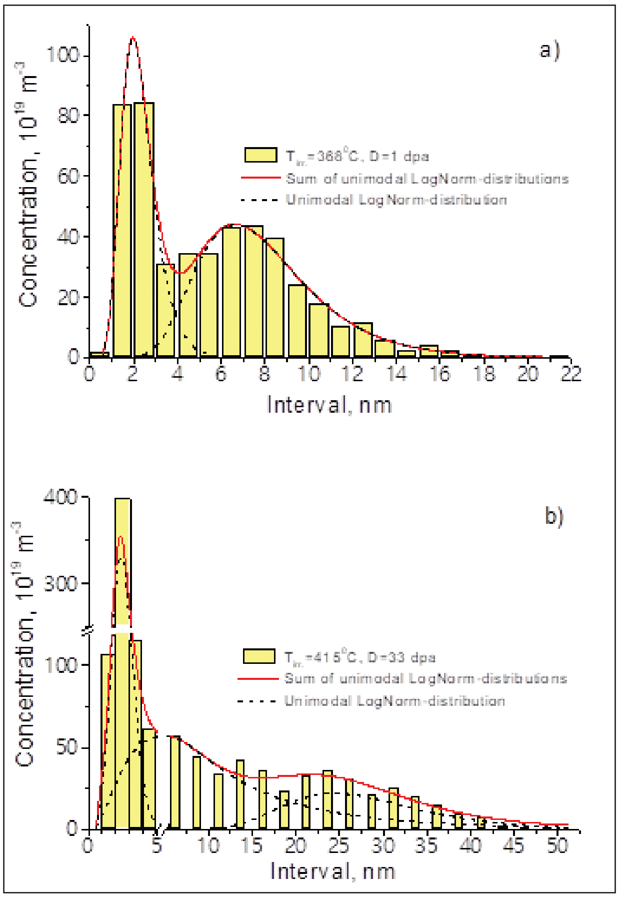

Figure 2. Histograms of void distributions according to void sizes for samples irradiated at ~370 °C to damage dose >1 dpa (a) and at ~415 °C to damage dose of ~33 dpa (b).

maximum and "large voids" including all remaining void sizes. Since the main contribution in radiation swelling is made by large-size voids (more than 5 nm), quantitative characteristics of large-size voids were examined and, accordingly, the value of swelling of samples was estimated based on them.

Porosity characteristics were determined using histograms of void distribution according to void sizes for samples belonging to the first and second batches including the following: average void size, concentration, void surface area and swelling. Temperature dependences of the average void size and concentrations of large-size voids in Cr18Ni9 steel after irradiation in the BN-600 reactor during 22 years and 33 years are presented in Figure 3. It is evident that average void size grows with increasing irradiation temperature and for material irradiated during 33 years at temperatures in excess of 370 °C average void size is larger than that for material irradiated during 22 years. Concentration of voids is maximum within the temperature interval of 380–420 °C. Concentration of voids decreases when temperature goes beyond this interval on both sides. Increased duration of irradiation (from 22 years to 33 years) results in the decrease of void concentration, at the same time void size increases. This effect is probably caused by the coalescence of voids (Kozlov and Portnykh 2008) and by the fact that practically no new voids are formed during this phase.

Specific integral void surface areas also reach maximum within the temperature range of 380–420 °C (Fig. 4a) and the value of integral void surface area increases with duration of operation increased from 22 years to 33 years. Behavior of swelling of Cr18Ni9 material is similar with maximum observed at temperature of ~415 °C (Fig. 4b). At lower temperatures (≤ 370 °C) and at higher temperatures (≥ 440 °C) swelling is estimated to be at the level of uncertainty of its determination, which does not allow detecting differences between the values of swelling for samples after irradiation during 22 years and 33 years.

Modification of structure of the material under irradiation results in the change of its physical characteristics one of which is the Young's modulus of the material (Kozlov et al. 2004b, Balachov et al. 2004). Dependence of variation of Young's modulus for Cr18Ni9 steel after irradiation in the BN-600 reactor during 22 years and 33 years is presented in Figure 5. Value of Young's modulus measured on non-irradiated samples manufactured from thick steel plate taken from the same melt as the material under investigation is also provided in the diagrame. Symbols with indicated measurement error bars correspond to the average values with their standard deviations obtained for four samples under identical irradiation conditions. It is evident that Young's modulus decreases with increased damage dose after 22 years and 33 years of irradiation. Notably, the values of Young's modulus for the material irradiated during 22 years are lower than those of the material irradiated during 33 years for the same values of radiation dose.

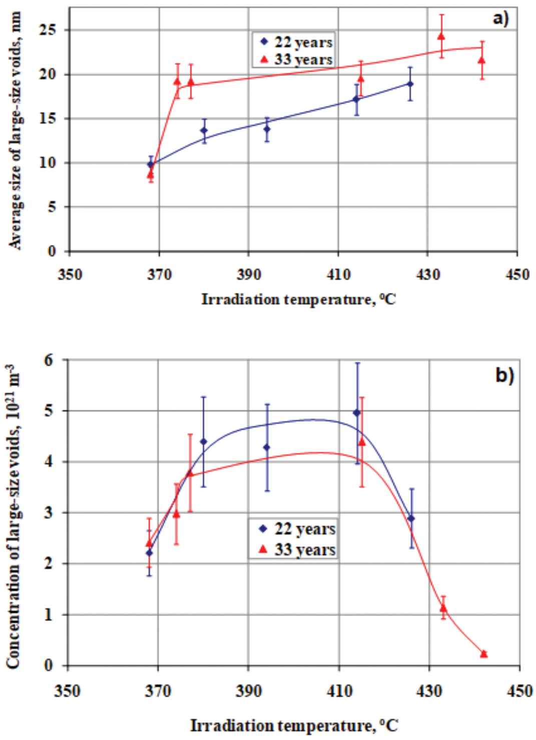

Figure 3. Temperature dependences of the average pore size (a) and concentration (b) of large-size pores in Cr18Ni9 steel after irradiation in the BN-600 reactor during 22 years and 33 years.

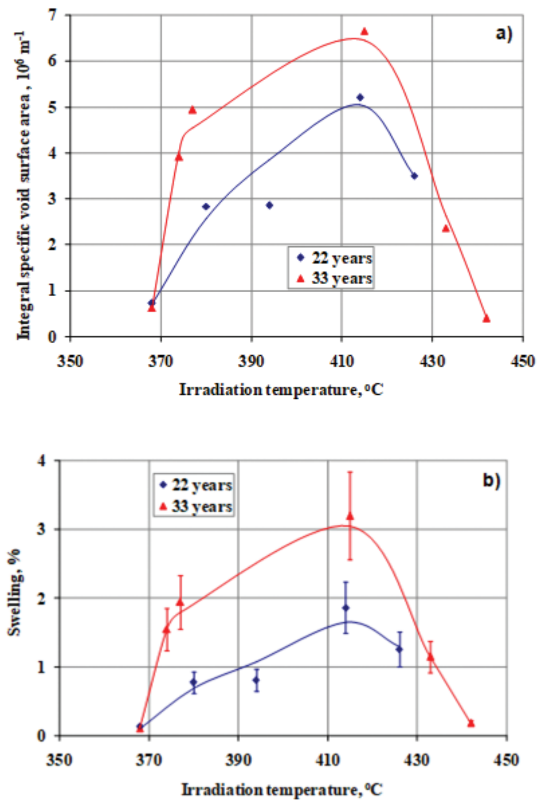

Figure 4. Temperature dependences of integral specific void surface area (a) and swelling (b) for large-size voids in Cr18Ni9 steel after irradiation in the BN-600 reactor during 22 years and 33 years.

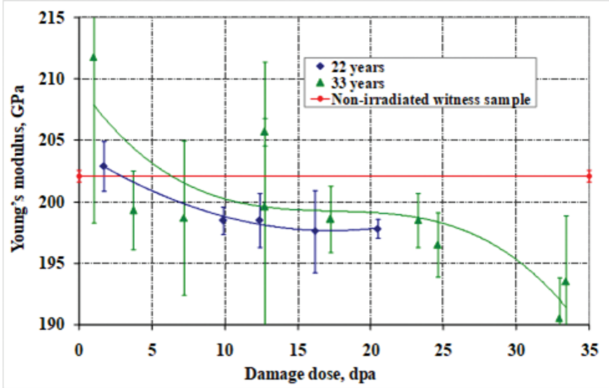

Figure 5. Variation of Young's modulus for Cr18Ni9 steel after irradiation in the BN-600 reactor during 22 years and 33 years depending on the damage radiation dose.

Discussion

The implemented studies demonstrated that already for doses equal to several units of dpa radiation swelling of Cr18Ni9 steel takes place, and it reaches about 3% with dose increasing to ~30 dpa. Other structural changes occur and along with mentioned above result in the change of elasticity characteristics. As it was demonstrated in (Kozlov et al. 2004a, Mosin et al. 2008), in the case when no other structural changes take place or when radiation swelling produces dominating effect on physical and mechanical properties of Young's modulus caused by swelling can be calculated using the following formula:

$$\frac{\Delta E}{E_0} = \frac{1}{(1+S)^2} - 1, \qquad (1)$$

where ΔE is the absolute variation of Young's modulus; E_0 is the Young's modulus in the initial state; S is the radiation swelling expressed in fractions of unity.

Variation of Young's modulus dependences on different irradiation doses both calculated using formula (1) and in accordance with the values of swelling obtained for irradiated samples by electron microscopy technique were plotted (Fig. 6). Dependences obtained for the material after 22 years of irradiation are shown in Figure 6a. It is clear that relative changes of Young's modulus calculated according to the values of swelling are lower than the measured one. Similar dependences were obtained for the material after 33 years of irradiation (Fig. 6b). Discrepancy between the relative change of Young's modulus calculated according to swelling data and the measured values is also observed. This proves that under long-term irradiation to small values (< 2 %) of swelling the latter is not the dominating factor influencing variation of mechanical properties of the material (Povstyanko et al. 2004, Ershova et al. 2008).

Such variations of mechanical properties include formation of secondary phases developing with simultaneous variation of composition of the crystalline matrix (Margolin et al. 2010). Precipitates of carbides is accompanied with depletion of interstitial impurities in the solid

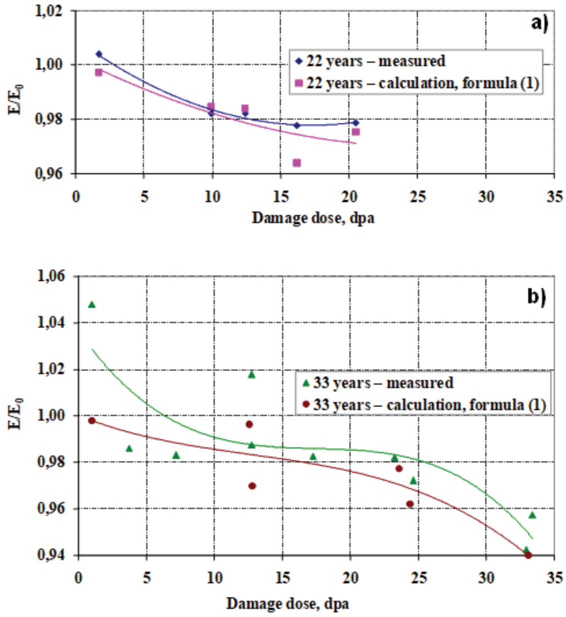

Figure 6. Relative variation of value of Young's modulus for Cr18NIi9 steel after irradiation in the BN-600 reactor during 22 years and 33 years (a.b) and the value of Young's modulus theoretically calculated according to the formula (1) using the value of swelling of samples depending on the damagie dose.

solution which should result in the decrease of Young's modulus (Lifshitz et al. 1980). On the other hand, carbide particles have Young's modulus higher than that for crystalline matrix (Neumark 1967, Poplavsky 2014). Obtaining numerical estimation of the effect on Young's modulus caused by secondary phases appears to be impossible because of the absence of quantitative information on them and on their elasticity characteristics.

One more factor capable to lead in wide scattering of properties among samples and as the reason in scattering of average values of obtained characteristics may be the non-uniformity of the material of the metal pipe, since the samples were cut from real metal-intensive industrial structures where it is more difficult to maintain the conditions of uniformity of structure and composition in the process of manufacturing than in fabricating dedicated samples from laboratory melts. Scattering of structure and properties inside the same batch of samples can be associated with gradients of temperatures and stresses over the section in different parts of a thick-wall structure with factual values which may differ from the values obtained in model thermal physics calculations of γ-heating and heat removal from parts of pipe wall during its operation in the reactor.

Conclusion

Investigation of physical properties and radiation porosity of Cr18Ni9 steel formed under neutron irradiation with displacement rates ranging from 1.0×10^{-9} dpa/s to 4.3×10^{-8} dpa/s at temperatures of 370–440°C during 22 years and 33 years demonstrated the following.

Radiation voids were formed in the all investigated samples with two maxima observed in the histograms of void distributions: "small-size" voids with sizes less than 5 nm and "large-size" voids with sizes in excess of 5 nm with main contribution in radiation swelling made by the latter.

The average size of "large-size" voids increases at temperatures in excess of 370 °C with growing irradiation temperature and it is larger for the material irradiated during 33 years than for the material irradiated during 22 years.

Concentrations of "large-size" voids reach maximum within temperature interval of 380–420 °C; increase of duration of irradiation from 22 years to 33 years results in certain decrease of void concentration.

Integral specific surface area for "large-size" voids is maximum within temperature range of 380–420 °C with values of integral void surface area increasing with duration of irradiation increased from 22 years to 33 years.

Swelling of Cr18Ni9 steel material has maximum at temperature of ~415 °C and reaches values of ~3% after 33 years of irradiation.

Variation of Young's modulus over the long time period demonstrates non-monotonous dependence on the damage dose; maximum relative variation of Young's modulus after 22 years of operation does not exceed 2%, and that after 33 years of operation does not exceed 6% of respective values in the initial state.

Along with radiation swelling, other structural changes are produced significant influence on the change of physical properties under the existing irradiation conditions such as the formation of secondary phases and the composition changes of crystalline matrix.

According to the results of investigation, BN-600 reactor internals made of Cr18Ni9 steel still retain operability after 33 years of operation.

Comparison of results obtained on the material after 22 and 33 years of irradiation contain information required for the description of subsequent variations of structure and properties of elements of reactor internals made of Cr18Ni9 steel. The obtained results can be used in forecasting the ultimate life of the reactor within the framework of already existing or newly developed models.

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