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Microstructure evolution and degradation mechanisms of reactor internal steel irradiated with heavy ions

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ABSTRACT

Structure evolution and degradation mechanisms during irradiation of 18Cr–10Ni–Ti steel (material of VVER-1000 reactor internals are investigated). Using accelerator irradiations with Cr^{3+} and Ar^{+} ions allowed studying effects of dose rate, different initial structure state and implanted ions on features of structure evolution and main mechanisms of degradation including low temperature swelling and embrittlement of the 18Cr–10Ni–Ti steel. It is shown that differences in dose rate at most irradiation temperatures mainly exert their influence on the duration of the swelling transient regime. Calculations of possible transmutation products during irradiation of this steel in a VVER-1000 spectrum were performed. It is shown that gaseous atoms (He and H), which are generated simultaneously with radiation defects, stabilize the elements of radiation microstructure and influence the swelling. The nature of deformation under different temperatures of irradiation and of mechanical testing is investigated. It is shown that the temperature sensitivity of swelling behaviour in the investigated steel, with different initial structures can be connected with the dynamic behaviour of point defect sinks.

1. Introduction

The materials of pressure vessel internals (PVI) during operation can degrade mainly due to processes of low temperature swelling and embrittlement. These phenomena are very complicated and can be influenced by various factors. Evolution of structure-phase conditions will determine the level of swelling and the degree of degradation [1].

In the present study the influence of dose rate on the swelling of 18Cr–10Ni–Ti steel is investigated. The 18Cr–10Ni–Ti steel is used in the states of the former Soviet Union for nuclear applications where AISI 304 would be used in Western countries. Structure-phase variations, in steel of different initial structure states, after irradiation with ions of chromium are analyzed. The relation of microstructure evolution, produced under various regimes of irradiation, with mechanisms of deformation and failure of steel were studied. Mechanisms of void development under damage production and simultaneous implantation of inert gas and hydrogen ions are also investigated.

2. Experimental

18Cr–10Ni–Ti steel samples with different initial conditions such as: solution annealed (SA) and cold worked (CW) are investigated. The nominal composition of the steel was, in weight percent, 0.08 C, 0.01 O, 0.2 Ti, 18.2 Cr, 10.4 Ni, 0.3 Si, 1.2 Mn, 0.03 P, 0.03 N with the balance Fe.

The conditions of treatments and irradiation of steels are summarized in Table 1. Irradiation with chromium ions was carried out on the ESUVI accelerator of NSC KIPT. Irradiations of specimens with argon and deuterium ions were carried out on the ESU-2 accelerator. The substitution of deuterium for proton allows the use of nuclear reactions to determine the depth distribution and concentration of hydrogen isotopes.

Specimens were produced as microscopic discs of 3 mm diameter or plate tensile specimens with a total length of 55 mm (the length of the effective part being 10 mm), a width of 3 mm, and a thickness of 0.3 mm. Specimens for electron-microscopic investigation were thinned from two sides. The layer situated at a depth of 150–200 nm from the irradiated surface was selected for analyses [2]. Final electropolishing for TEM investigation was performed with an electrolyte containing 530 ml of $C_3H_8O_3$, phosphoric acid H_3PO_4 and 80 ml H_2O.

Some of the irradiated foils were tensile deformed at room temperature and at 573 K (in vacuum) with a deformation rate of

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Table 1
Experimental conditions.

	Initial state		Irradiation	Deformation		
1	3 mm discs, SA 1323 K, 30 min 3 mm discs, 5.5% CW	Previous electropolishing	2 MeV Cr^{3+}, D = 5–100 dpa, $k = 10^{-2}$ dpa s^{-1}, T_{irr} = 823–923 K		Final electropolishing	Electron-microscopic study
2	Tensile specimen, SA 1323 K, 30 min		2 MeV Cr^{3+}, D = 5–100 dpa, $k = 10^{-2}, 10^{-3}$ dpa s^{-1}, T_{irr} = 573–908 K	ε= 7%, $\dot{\varepsilon} = 3\times10^{-3} s^{-1}$ T = 293, 573 K		
3	3 mm discs, SA 1323 K, 30 min		2 MeV Cr^{3+}, D = 5–100 dpa, $k = 10^{-2}, 10^{-3}$ dpa s^{-1}, T_{irr} = 723–923 K			
4	3 mm discs, SA 1323 K, 30 min		1.4 MeV Ar^+, $\Phi_{Ar} = 3–12\times10^{16} cm^{-2}$; 5 keV D^+, $\Phi_D = 5\times10^{16} cm^{-2}$; $j = 5.5\times10^{12} cm^{-2} s^{-1}$ T_{irr} = 873 K			

D = damage dose, k = dose rate, T_{irr} = irradiation temperature, $\phi\phi$ = fluence, j = flux, T = deformation temperature, ε = total elongation, $\dot{\varepsilon}$ = strain rate, SA = solution annealed and CW = cold worked.

$3\times10^{-3}\ s^{-1}$ up to 7%. For electron-microscopic study, the foils-discs were cut from the deformed area of these specimens. The electron-microscopic study was performed on an electron-microscope JEM-100CX.

During the study of specimen structure, density of dislocation, size, concentration and volume fraction of voids (swelling) were determined. The density of dislocations was calculated by the secant method. The value of swelling S was determined by the formula

$$S = 100 \cdot \frac{\pi}{6} \sum_{i=1}^{n} d_i^3 \approx 100 \cdot \frac{\pi}{6} \cdot \bar{d}^3 \cdot N,$$

where S is the swelling (%), $\bar{d}$ the average cavity diameter and N their number density.

3. Results of investigation

3.1. Structure of solution annealed and cold worked (5.5%) steel 18Cr–10Ni–Ti

Evolution of specimens structure from 18Cr–10Ni–Ti steel in different initial conditions – solid solution and deformed by 5.5% (which are typical for VVER – internals' materials) are investigated.

A dislocation network which consists mainly of perfect dislocation, forms in the steel solid solution for doses causing more than 5 dpa. Isolated dislocation loops are also observed, an insignificant number of which have a stacking fault. For doses causing more than 25 dpa, dislocation loops are practically absent. Under an irradiation temperature of 623 K, the main elements of dislocation structure are the dislocation loops with stacking faults and perfect prismatic dislocation loops. At a temperature of 863 K, the separate segments of dislocation network and dislocation loops are also observed. The size of loops is larger and the concentration is lower, in comparison with these parameters at an irradiation temperature of 623 K. In the temperature range 873–908 K, a perfect dislocation network is observed with a complete absence of dislocation loops.

It was stated for cold worked (5.5%) 18Cr–10Ni–Ti steel, that the dislocation structure for irradiation doses causing up to 5 dpa consists of primary dislocation and radiation-induced dislocation loops. The number of loops with stacking fault is considerably higher than in solid solution annealed steel irradiated in the same conditions. Under a dose corresponding to 20 dpa the isolated dislocation loops are observed, but, on the whole, the dislocation network is already formed. The density of dislocations in the solution annealed steel also decreases insignificantly with increase of temperature.

The temperature dependence of swelling for solution annealed steel has the characteristic bell-like appearance with a maximum at 888 K (Fig. 1). The abrupt decrease of swelling in the narrow range 863–883 K is observed below the maximum.

Swelling (porosity nucleation) is already observed at 5 dpa (sub-figure in Fig. 1). The incubation period is practically absent but the transient period is very large. In this paper the transient period (D_0) is thought to be the point of intersection with the axis of abscissa of the line at the angle corresponding to the swelling rate in the stationary region. The steady state swelling is fixed under a dose corresponding to ~25 dpa and the rate of swelling does not exceed 0.36% dpa^{-1}. Porosity at doses corresponding to less

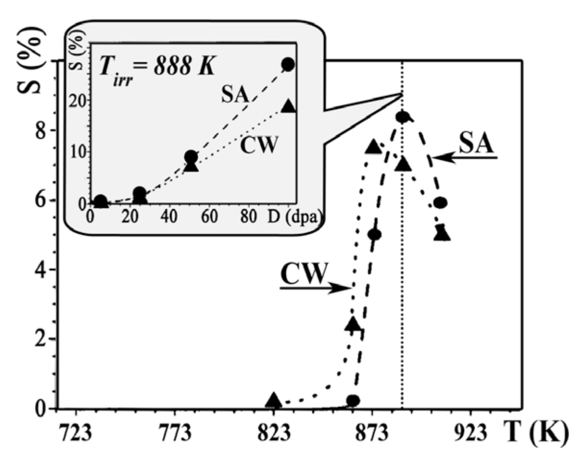

Fig. 1. Temperature (dose = 50 dpa) and dose (T_{irr} = 888 K) swelling dependencies for 18Cr–10Ni–Ti steel with different initial states (● – SA at 1323 K, 30 min, ▲ – 5.5% CW).

than 50 dpa is inhomogeneous. The void distribution is rather heterogeneous – both boundary areas free from voids and area with increased porosity are observed. Voids have the exact crystallographic cut.

The temperature dependence of swelling of cold worked (5.5%) steel also has a bell-like appearance (Fig. 1) but the maximum is situated at 878 K. After the abrupt decrease of swelling in the range 873–863 K its gentle decrease is observed down to 823 K. Swelling of the deformed 18Cr–10Ni–Ti steel reaches a stationary level at a dose corresponding to ~40 dpa. The rate of swelling does not exceed 0.23% per dpa in the stationary region. Porosity for doses corresponding to less that 50 dpa is inhomogeneous.

3.2. Investigation of micro-mechanisms of plastic deformation and failure of 18Cr–10Ni–Ti steel

Study of the microstructure of a specimen irradiated at 578 K and 7% post-deformed at room temperature had shown that the main mechanism of deformation is twinning (see also [3]). Deformation at 573 K of specimens irradiated to 5 and 10 dpa causes localized plastic flow. Narrow bands (~50–60 nm) completely free from dislocation loops are observed. In the literature these bands are named ‘dislocation channels’ [4]. The important part of the material volume remains completely undeformed.

The stress that is localized on the grain boundary during dislocation channeling reaches 900–1000 MPa. It is much higher than the value of 640–650 MPa corresponding to the grain boundary strength of this steel.

Irradiation at 908 K with a dose rate causing 1×10^{-2} dpa s^{-1} up to 100 dpa causes the formation of a defect structure that is characterized by a high value of porosity ($\geqslant$20%) and by the contribution of voids to the process of steel failure.

So at a temperature lower than the maximum for cold-deformed steel swelling the swelling resistance of austenite steel is higher under similar conditions of irradiation (damage dose and temperature). At temperatures higher than that for the maximum swelling of cold-deformed steel, the situation is reversed: the cold-deformed steel swelling resistance is higher than that of austenite. As it is seen from the dose dependence of swelling in the high temperature range, it occurs at the expense of a higher swelling rate in the stationary region for austenite steel. It is important to note that the initial difference in the dislocation density did not influence the incubation period or the transient stage.

3.3. Influence of dose rate on void swelling

The temperature dependency of steel swelling (Fig. 2) has the characteristic bell-like appearance and reveals a displacement by 25 K to higher temperatures during the variation of dose rate from 10^{-3} to 10^{-2} dpa s^{-1}. It must be noted that the behaviour of the swelling curve in the rising area (low temperature range of swelling) is more extended at a dose rate of 10^{-3} dpa s^{-1} in comparison with 10^{-2} dpa s^{-1}. The influence of dose rate is manifested in the duration of the transient period of swelling and is more defined at lower temperatures. Irradiation at 888 K leads to a considerably lower influence of dose rate (see Fig. 2 (a) and (b)).

Comparison of typical dose rates of 10^{-3}–10^{-2} dpa s^{-1} used in the presented investigation and 10^{-9}–10^{-8} dpa s^{-1} which are typical for VVER reactor internals allows concluding that the damage level corresponding to the start of swelling will not exceed 20 dpa. This assumption confirms the results which are obtained in experiments on the BOR-60 reactor [5].

It is generally suggested that the decrease of dose rate in the high temperature range may influence only the duration of the transient period of swelling [6]. Fig. 3 shows that the dose rate influences both the duration of the transient period and the rate of steady state swelling. The duration of the transient period grows with the increase of dose rate. As to the rate of swelling, the reverse trend is observed – with the increase of dose rate the rate of swelling decreases. The shaded areas indicate the ranges of transient period and swelling rate values.

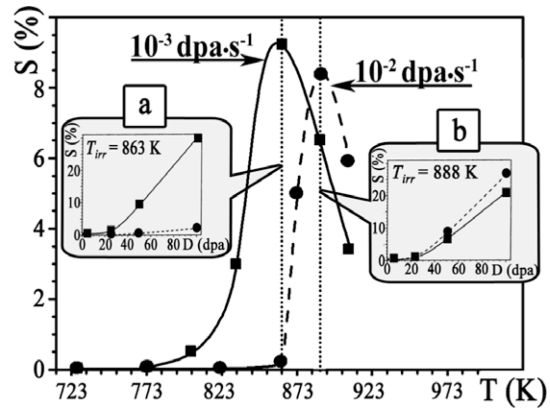

Fig. 2. Temperature dependence of swelling of solution annealed stainless steel 18Cr–10Ni–Ti (dose = 50 dpa.) The dose rates are: 1×10^{-3} dpa s^{-1} (■) and 1×10^{-2} dpa s^{-1} (●); (a) and (b) – dose dependence of swelling at T_{irr} = 863 and T_{irr} = 888 K, respectively.

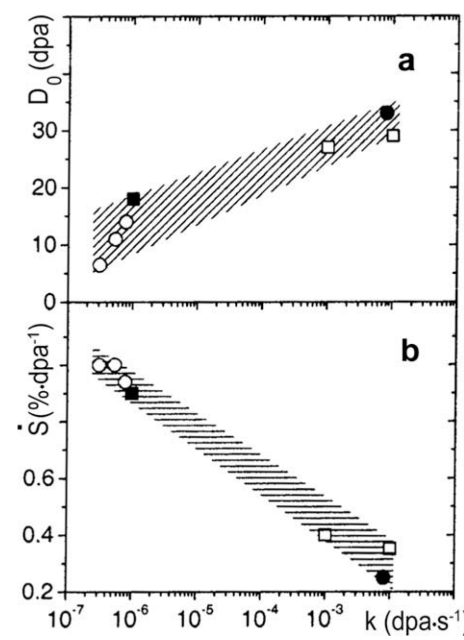

Fig. 3. Influence of dose rate (k) on the value of transient period (D_0) (a) and the rate of swelling (S) (b) in the maximum of temperature of steel swelling: (□) – 18Cr–10Ni–Ti T_{irr} = 888 and 863 K for 10^{-2} and 10^{-3} dpa s^{-1}, respectively (present paper); (■) – 18Cr–10Ni–Ti $T_{irr} \approx 753$ K (BOR-60) [5]; (○) – Fe–15Cr–16Ni $T_{irr} \approx 683$ K (FFTF) [7]; (●) 16Cr–15Ni–3 M-B T_{irr} = 923 K (heavy ions) [8].

Therefore, a decrease of dose rate leads to a shift of the beginning of swelling to lower temperatures of irradiation. This is very important for materials for reactor internals, which have a very complicated construction, and are in service at comparatively low temperature under low dose rate.

3.4. Stabilization of the radiation microstructure elements by helium and hydrogen

In radiation environments, the potential interaction of helium, hydrogen, and radiation defects is of current interest, especially with respect to their potential participation in life-limiting phenomena such as void swelling. It is known that the generation rates of helium and hydrogen can be rather large. There are various sources of these two gases in fission neutron spectra for different components of the alloy.

Computation of the processes of transmutation of element isotopes entering in the composition of steel 18Cr–10Ni–Ti showed that fission neutrons ($E_n > 0.1$ MeV) form continuously atoms of hydrogen in (n,p) nuclear reactions on practically all isotopes of 18Cr–10Ni–Ti steel without variation of element composition, because the daughter nuclei-products are short-lived and their β-decay leads to the parent isotope. The rate of hydrogen production is $\approx 2.3 \times 10^{-6}$ appm s^{-1}. The impact of the two step reaction: ^{58}Ni(n,γ) ^{59}Ni(n,p) 59Co depends on the concentration of nickel in the steel and on the effect of moderation on the neutron spectrum (0.4 eV < E_n < 1 MeV).

The predominating sources of hydrogen penetration into the steel of pressure vessel internals are the processes that occur in the medium of moderator coolant. The rate of hydrogen production from corrosion reactions contributes $\sim 1 \times 10^{-4}$ appm s^{-1} [9].

Most helium in reactor core materials is produced in (n,γ) threshold nuclear reactions by fission neutrons ($E_n > 0.1$ MeV) on practically all isotopes of elements, composing the 18Cr–10Ni–Ti steel and also on isotopes of impurity elements and reaches, during 40 years of operation, 625 appm.

Hydrogen and helium are produced simultaneously in transmutation nuclear reactions of all isotopes of elements composing the 18Cr–10Ni–Ti steel, including impurities. The performed calculations showed that generation of atoms of hydrogen in nuclear reactions with fast neutrons ($E_n > 0.1$ MeV) exceeds by approximately five times the generation of helium atoms.

For the investigation of mechanisms of porosity development under damage production and simultaneous implantation of atoms of helium and hydrogen, 18Cr–10Ni–Ti steel was irradiated by high-energy ions of argon and deuterium. It is known [10] that inert gases neon and argon may be used instead of helium due to the character of their influence on gas porosity development.

An increase of the irradiation dose by ions of argon at a temperature of 873 K, causes the simultaneous increase of inert gas concentration and of damage level. Under on irradiation dose by argon of 3×10^{16} cm^{-2}, fine 3 nm bubbles with density 2.5×10^{17} cm^{-2} form, and the swelling was 0.35%. With on increase of irradiation dose by argon to 1.2×10^{17} cm^{-2}, the formation of large 5 nm bubbles is observed, with a decreased concentration ($\rho = 1.2 \times 10^{17}$ cm^{-3}) but with an approximately doubled swelling ($S = 0.78\%$).

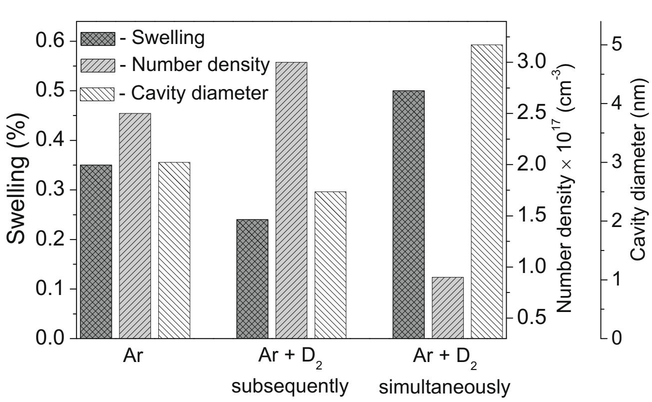

Fig. 4. Parameters of porosity in 18Cr–10Ni–Ti steel irradiated by different ions at $T_{irr} = 873$ K.

Irradiation of steel by high-energy ions of argon at $T_{irr} = 873$ K with subsequent or simultaneous ion implantation of deuterium at 873 K causes the change of parameters of previously formed void structures. Parameters of porosity (swelling, number density and cavity diameter) in 18Cr–10Ni–Ti steel irradiated by different ions are presented in Fig. 4. After subsequent ion implantation of deuterium a decrease of void sizes and an increase of their density is observed. After simultaneous production of damage and the implantation of inert gas and hydrogen (deuterium), the mean size of voids increases to 5 nm and their density decreases.

Swelling in the case of damage production at a level of 50 dpa with simultaneous implantation of inert gas and hydrogen reaches 0.5%, i.e., approximately 1.7 times higher than that with implantation of only one inert gas and more than two times higher than the swelling that is observed after subsequent implantation of deuterium.

4. Conclusions

Features of structure evolution and degradation mechanisms of irradiated 18Cr–10Ni–Ti steel, used for VVER reactor internals, were studied.

A decrease of dose rate causes swelling to start at lower temperatures. Dose rate influences on the rate of steel swelling and on the duration of transient period have been established.

It has been shown that at temperatures lower that the maximum of swelling of solution annealed 18Cr–10Ni–Ti steel the swelling resistance of solution annealed steel is higher than for cold worked (5.5%) steel.

The change of deformation mechanism connected with increase of temperature and irradiation dose has been described, this change consists in going from twinning to dislocation channeling which may cause the embrittlement and subsequent failure of baffle material.

The evolution of porosity in 18Cr–10Ni–Ti stainless steel shows that the production of damage and the simultaneous implantation of inert gas and deuterium causes increased of material swelling.

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