THE EVOLUTION OF THE MICROSTRUCTURE OF Cr16-Ni19 STEEL UNDER IRRADIATION IN THE LOW ENRICHMENT ZONE OF A FAST NEUTRON REACTOR. THE EFFECT OF NEUTRON IRRADIATION CONDITIONS ON THE STRUCTURAL AND PHASE STATE

© 2025 V. L. Panchenko*, I. A. Portnykh, A. E. Ustinov

JSC "Institute of Nuclear Materials", Zarechny, Sverdlovsk region, Russia

**e-mail:* panchenko_vl@irmatom.ru

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Abstract. Microstructural studies of samples made from various sections of fuel element shells were carried out after irradiation in the low enrichment zone of a fast neutron reactor with a sodium coolant to damaging doses of over 100 sna. At different sites, the rate of generation of atomic displacements varied from 0.5×10–8 to 1.6×10–6 sleep/s, the irradiation temperature ranged from 370 to 630°C. The structural and phase state of the shell samples is investigated, the evolution of the composition and morphology of the secretions of the second phases and the austenitic matrix is shown.

Keywords: *fuel rods, neutron irradiation, austenitic steel type Cr16–Ni19, microstructure, radiation-induced segregation, phase composition*

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INTRODUCTION

During operation in fast neutron reactors, the core material is exposed to extreme temperatures and high-flux neutron irradiation. The maximum impact is experienced by the fuel element cladding material, the service life of which directly affects the economic efficiency of the reactor. The main factor limiting the service life of fuel elements is radiation swelling of austenitic steel cladding [1]. Improvement of the resistance of Cr16–Ni19 steel in the cold-worked state (c.d.), used as fuel element cladding material, to pore formation is achieved by improving the cladding manufacturing technology [2, 3].

In addition to the generation of vacancies, which form pores, there is a redistribution of steel alloying elements, leading to the evolution of the structural-phase state, which determines the physical-mechanical and corrosion properties of steel. Therefore, to understand the processes leading to changes in the properties of structural materials, to forecast the service life of products made from them, increased attention should be paid to studying the evolution of the microstructure formed under various steel irradiation conditions (temperature, rate of atomic displacement generation, time).

Previously, studies were conducted on Cr16-Ni19 type steel used in claddings manufactured according to earlier technology, after irradiation in the BN-600 reactor to a damaging dose of ~ 95 dpa [4].

The aim of the work is to obtain and systematize experimental data on the structural-phase state of austenitic steel type Cr16-Ni19 c.w. in claddings manufactured using modern technology, after irradiation in the low enrichment zone of the BN-600 reactor in the temperature range from 370 to 630 °C.

MATERIALS AND METHODS

The studies were conducted on samples of claddings from four fuel elements made of steel type Cr16-Ni19 c.w., which had been operated in the BN-600 reactor in the low enrichment zone for 750 effective days. Samples were cut from different areas along the height of the fuel elements, which differed in irradiation conditions - temperature and dose accumulation rate.

All studied samples were conditionally divided into four groups according to the irradiation temperature: the cladding section in the area of the lower gas cavity of the fuel elements, located below the active part (containing fuel) of the fuel element - ~ 370°C, low-temperature section (420-455)°C, medium-temperature section (485-550)°C and high-temperature section (570-630)°C. In each temperature range, there were samples that slightly differed in the rate of atomic displacement generation. In addition, a sample of fuel element cladding made of steel type Cr16-Ni19 c.w. in unirradiated state, manufactured using the same technology, was studied.

For electron microscopy studies, segments ~3 × 3 mm were cut from the fuel rod claddings. Then the segment was mechanically thinned on sandpaper to a thickness of (0.10 – 0.12) mm. Thin f oils for microscopy were prepared on a TwinJet (Fischione) dual-jet electropolishing unit in an electrolyte cooled to 16 °C with composition – (90% CH_3COOH + 10 % $HClO_4$) at a voltage of (30 – 35) V. After electropolishing, the samples were subjected to additional cleaning with low-energy Ar^+ ion beams on a TEMMill 1050 (Fischione) unit at an accelerating voltage of 300 V on the ion gun.

The studies were carried out on a Talos F200X G2 (ThermoFisher electron microscope Scientific) high-resolution transmission scanning with a resolution of 0.1 nm in transmission (TEM - Transmitted Electron Microscopy) and scanning (STEM - Scanning Transmitted Electron Microscopy) modes. The microscope makes it possible to obtain images with atomic resolution and perform direct measurements of interplanar distances in the crystal lattice.

The microscope is equipped with an integrated Super-X EDS (Energy-Dispersive Spectrometer , ThermoFisher Scientific) system for analyzing the elemental composition of the material by detecting characteristic X-ray radiation generated by an electron beam, with the ability to register the spectrum of elements in the range from B5 to Am95 with a detection limit of ~ 0.1 wt. %. Since the material irradiated by fast neutrons acquired its own induced activity, in the spectrum of which the peaks of manganese and chromium predominate, when conducting a quantitative analysis of the composition of the matrix and precipitates of secondary phases, a correction was made for chromium, while manganese was excluded from the quantitative assessment. The use of local elemental analysis together with fast Fourier transform (FT) in processing crystal lattice images obtained in high-resolution mode (HRTEM) allows identification of even single precipitates of finely dispersed secondary phases.

To determine the local thickness of the foil when conducting quantitative assessments of the concentration of various microstructure objects and elemental composition of fine dispersion precipitates, Electron Energy Loss Spectroscopy (EELS, Gatan) was used - measuring the ratio of the integral intensity of the spectrum to the intensity of the zero-loss peak [5, 6].

3. RESULTS

Initial (unirradiated) state

The Cr16-Ni19 c.w. type steel before irradiation was in an austenitized state, with an average grain size of ~ 20 μm, and a dislocation density of ~ $3.4 \cdot 10^{14}$ m^{-2}. The dislocations formed a cellular structure with an average cell size of ~ 0.2 μm (Fig. 1a).

Fig. 1. Microstructure of Cr16-Ni19 c.w. steel in the initial (unirradiated) state: a - cellular dislocation structure; b-d - segregations of Cr, Mo, Ti at the grain boundary (marked by black arrows); e - distribution profile of Cr, Ni, Mo, Ti across the grain boundary (dashed line).

The main alloying elements are relatively uniformly distributed throughout the material volume; of the second phases, only primary carbonitrides of the (Ti,Nb,Mo)(C,N) type were observed. On one of the grain boundaries, whose surface was oriented perpendicular to the image plane, segregation of chromium, molybdenum, and titanium was registered, which substitute Fe and Ni (Fig. 1b-d).

Area of gas cavity, 370°C

In the structure of the material irradiated outside the active zone at a temperature of ~ 370°C under conditions of low atomic displacement generation rate G ~ $(0.5\text{-}1.1) \cdot 10^{-8}$ dpa/s, there was a transformation of the dislocation structure - instead of cells, a relatively uniform dislocation network was observed (Fig. 2a), and interstitial Frank dislocation loops with stacking faults (SF) inside [7] were formed, lying in the close-packed planes {111} of the austenitic FCC matrix (Fig. 2b). The dislocation density in samples from various fuel elements was ~ $(5\text{-}10) \cdot 10^{14}$ m^{-2}, the concentration of Frank loops was ~ $(2\text{-}7) \cdot 10^{21}$ m^{-3}, and the average size of the loops was (20-36) nm.

Fig. 2. Dislocation structure of Cr16-Ni19 c.w. steel after irradiation, T_{irr} ~ 370°C, G ~ $1.1 \cdot 10^{-8}$ dpa/s: a - relatively uniform dislocation network; b - banded contrast on Frank loops with stacking fault, dark-field STEM image.

No new phases were detected, only primary carbonitrides were identified. Diffusion of point defects generated during irradiation led to the formation of radiation-induced segregation (RIS) of Ni and Si both at grain boundaries (Fig. 3a, c, e) and at dislocations and Frank loops (Fig. 3b, d, f).

Fig. 3. RIS at grain boundaries and dislocations in Cr16-Ni19 c.w. steel after irradiation at temperature ~ 370°C. a, c, e - respectively, a section of grain boundary (dark-field STEM image), mixed map and distribution profile of Cr, Mo, Ni, Si across the boundary (boundary position is marked by a dashed line on the diagram), G ~ $1.1 \cdot 10^{-8}$ dpa/s; b, d, f - respectively, forest of dislocations (bright-field STEM image), mixed map and distribution profile of Si, Ni, Cr across the Frank loop and dislocation (dashed and dotted lines on the diagram) in the grain volume , G ~ $0.6 \cdot 10^{-8}$ dpa/s.

X-ray spectral microanalysis (XSMA) showed that as a result of RIS, the composition of the austenitic matrix changed - the proportion of Si decreased by almost 2 times (ΔC Si ~ (-45)%), the proportion of Ni decreased slightly (ΔC Ni ~ (-7)%) , and the relative content of Cr and Fe, respectively, increased by ~ 5% and ~ 2%.

Low-temperature range (420-455)°C

In the low-temperature range, the atomic displacement generation rate was $(1.0\text{-}1.4)\cdot 10^{-6}$ dpa/s. In various samples under TEM examination, a relatively uniformly distributed network (forest) of dislocations with a density of ~ $(4\text{-}6)\cdot 10^{14}$ m^{-2} and Frank loops with an average size of (20-30) nm and a concentration of $(5\text{-}13)\cdot 10^{21}$ m^{-3} were observed.

In the bulk of grains, intermetallic FCC phases based on Ni and Si were observed: G (a =(1.115–1.145) nm) and γ' (a =0.35 nm), as well as complex τ-carbides of the $M_{23}C_6$ type, where M=(Cr, Mo, V). Typically, G -phase forms in conjunction with vacancy pores and has similar dimensions (20–40) nm. Fine (10–15) nm γ'-phase forms on dislocations, where segregation of Ni and Si takes place. The particles of γ'-phase, having a lattice parameter close to that of austenite, are coherent with the matrix, therefore giving weak contrast in TEM; their presence can be determined by "coffee bean" type contrast under certain diffraction conditions (Fig. 4a) and by EPMA results (Fig. 5a–c). Complex FCC carbides have dimensions of (20–40) nm and are enriched with molybdenum and vanadium (Fig. 5a, d–f).

Fig. 4. Morphology of intermetallics in the grain bulk of Cr16–Ni19 c.w. steel after irradiation, $T_{irrad.}$ ~ 450°C, G ~ $1.3\cdot 10^{-6}$ dpa/s: a – "coffee bean" type contrast on coherent precipitates of γ'-phase; b – precipitates of G phase on vacancy pores; c, d – direct resolution of austenite lattice and corresponding FT with interpretation; e, f – direct resolution of lattice on γ' particle and corresponding FT with interpretation, forbidden reflections for FCC lattice are visible (in parentheses); g, h – direct resolution of lattice in the vicinity of interphase boundary and corresponding FT with interpretation, planes {022} of *G-phase* are parallel to {111} planes of austenite.

Fig. 5. Secondary phase precipitates and RIS in the grain bulk of Cr16–Ni19 c.w. steel after irradiation, T_{irrad} ~ 450°C, G ~ $1.3\cdot 10^{-6}$ dpa/s: a – grain area with secondary phase precipitates, bright-field STEM image; b, c – particles of γ'- and G -phase on Ni and Si distribution maps; d, e, f – τ-carbides ($M_{23}C_6$) on Cr, Mo and V distribution maps.

At grain boundaries (Fig. 6), τ-carbides form less frequently (Fig. 6b), while predominantly G -phase precipitates are observed (Fig. 6c, d), with non-uniform distribution of precipitates along grain boundaries. On boundary sections not occupied by second phase precipitates, the element distribution maps clearly show a thin segregation layer of nickel enriched with silicon (Fig. 6c, d). Formation of G -phase precipitates and τ-carbides is observed at the boundaries of some primary carbonitrides (Fig. 7, 8).

Fig. 6. Second phase precipitates and RIS at grain boundary in cold-worked Cr16-Ni19 steel after irradiation, T_{irr} ~ 450°C, G ~ $1.3\cdot 10^{-6}$ dpa/s: a – intergranular boundary section with second phase precipitates, dark-field STEM image; b – grain boundary τ-carbide particle on the Cr distribution map; c, d – G-phase particles and thin RIS line along the intergranular boundary on Ni and Si distribution maps; e, f – RIS of Ti and P in second phase precipitates.

Fig. 7. Formation of second phases and segregations at the periphery of primary carbonitride in cold-worked Cr16-Ni19 steel after irradiation, T_{irr} ~ 450°C, G ~ $1.4\cdot 10^{-6}$ dpa/s: a – dark-field STEM image; b–h – distribution maps of Ti , Cr, Ni, Mo, Si, V, P respectively on the analyzed foil section.

Fig. 8. Identification of phases formed on the primary carbide (Fig. 7) in cold-worked Cr16-Ni19 steel, by direct resolution of the crystal lattice, T_{irr} ~ 450°C, G ~ $1.3 \cdot 10^{-6}$ dpa/s: a, b – direct lattice resolution in the vicinity of the interface between primary carbide and intermetallic compound, and corresponding FT with interpretation; c, d – direct lattice resolution on $M_{23}C_6$ carbide and corresponding FT with interpretation.

EPMA showed (Fig. 5, 6, 7) that with increasing irradiation temperature, the composition of the austenitic matrix changes significantly due to the formation of new phases – ΔC Si ~ (–70)%, ΔC Ni ~ (–20)%, the relative content of Cr and Fe increased by approximately 8 and 6%, respectively. Additionally, molybdenum and vanadium affiliated with primary and secondary carbides, titanium forming RIS together with nickel and silicon and enriching intermetallic and carbide phases, as well as phosphorus, whose RIS is predominantly observed in G-phase at grain boundaries and at the periphery of primary carbides, are removed from the solid solution.

Medium-temperature range (485–550)°C

In the medium-temperature irradiation range, the rate of atomic displacement generation in the samples was ~ $(1.5–1.6) \cdot 10^{-6}$ dpa/s. Increased temperature and irradiation dose stimulated local grain boundary migration – they changed from straight to curved (Fig. 9a, b). Increased dislocation mobility led to polygonization effects, where an area with low-angle misorientation forms near the grain boundary, bounded by a dislocation wall separating the depleted boundary area from an area with high dislocation density (Fig. 9b). The dislocation density in various samples was ~ $(1–5) \cdot 10^{14}$ m^{-2}. Frank loops with stacking faults are uniformly distributed in the material, with average loop size of ~ (28–46) nm, and concentration of ~ $(1–4) \cdot 10^{21}$ m^{-3}.

Fig. 9. Microstructure of Cr16-Ni19 c.w. type steel, characteristic for the medium-temperature irradiation range, T_{irr} ~ (490–550)°C, G ~ $(1.5–1.6) \cdot 10^{-6}$ dpa/s: a – grain boundary bends caused by local migration; b – boundary area with low-angle misorientation and reduced dislocation density bounded on the right by a dislocation wall; c – grain boundary with a chain of second phase precipitates; d – particles of τ-carbides and G-phase with prismatic shape.

The phase composition of steel in the medium-temperature irradiation range practically does not differ from the composition after low-temperature irradiation. The occupancy of grain boundaries with second phase precipitates increases, but remains heterogeneous, with boundaries both highly filled (Fig. 9c) and free from precipitates. Among the grain boundary precipitates, the proportion of τ-carbides increases compared to the G-phase (Fig. 9d).

Fig. 10. Phosphides in the grain volume of Cr16-Ni19 steel, T_{irr} ~ 490°C, G ~ $1.5 \cdot 10^{-6}$ dpa/s: a, b – bright-field STEM image and phosphorus distribution map in the analyzed area; c, d - HR TEM and corresponding FFT with interpretation on one of the M_2P type phosphide particles (HCP, a ~0.587 nm, c ~0.346 nm).

A new phase discovered in the medium-temperature range is rod-shaped phosphides with a size of (10-80) nm (Fig. 10), usually enriched with Si and Ti. The phosphide particles have a characteristic twin structure, which leads to the appearance of streaks in the diffraction pattern or in atomic resolution FFT images (Fig. 10c, d). The concentration of phosphides in the steel structure after medium-temperature irradiation is insignificant.

Near the upper boundary of the medium-temperature range, EPMA methods reveal fine dispersed (2-5 nm) particles based on nickel and titanium. It was not possible to identify the phase by diffraction methods, but based on the estimated ratio of elements in the particles, it can be assumed that this is an intermetallic compound of the Ni_3Ti type.

With increasing irradiation temperature, the morphology of the second phase precipitate particles changes. Compared to the low-temperature range, there is an increase in their average size and a decrease in concentration, with characteristic appearance of conglomerates consisting of 2-3 particles of heterogeneous phases such as carbide-intermetallic, with common boundaries. This does not refer to previously observed conglomerates of second phases at lower irradiation temperatures that nucleate at the periphery of primary carbonitrides. In the medium-temperature range, conglomerates are formed by newly formed complex carbides of the $M_{23}C_6$ (τ), M_6C (η) type and *G*-phase (Fig. 11). It is noted that with increasing irradiation temperature, the probability of conglomerate formation increases.

Fig. 11. Morphology of carbide-intermetallic particle conglomerates in the structure of Cr16-Ni19 steel from the medium-temperature range, T_{irr} ~ 525°C, G ~ $1.6\cdot10^{-6}$ dpa/s: a, b - bright-field STEM image and corresponding composite map of Cr and Ni distribution; c-d - HR TEM on conglomerate particles marked by arrow (b), and corresponding diffraction patterns with identification of carbide (e) and intermetallic (f) lattice reflections.

It is also noted that with increasing irradiation temperature, carbide and intermetallic particles more often acquire prismatic shape with noticeable faceting (Fig. 9d). Furthermore, if at lower temperatures *G*-phase was localized directly on pores, and carbides were not associated with pores, then with increasing irradiation temperature, strict separation of localization is no longer observed.

According to EPMA data, in the medium-temperature irradiation range, the composition of the austenitic matrix continues to change. The nickel content in the matrix decreases by 30-40% and amounts to about 11-13 at.%, silicon depletion reaches ~ 80%, and as a result, the relative content of Cr and Fe increases by approximately 10-15% and 8-10%, respectively.

High-temperature range (570–630)°C

In the high-temperature irradiation range, the atomic displacement generation rate in the samples was from $1.5\cdot10^{-6}$ dpa/s in the lower part to $0.6\cdot10^{-6}$ dpa/s in the upper part of the range. The increase in irradiation temperature led to further evolution of the dislocation structure – a secondary cellular structure was formed, caused by polygonization processes, with the formation of low-angle dislocation boundaries in the grain volume (Fig. 12). The dislocation density in various samples was ~ $(1–3)\cdot10^{14}$ m^{-2}. Frank loops with stacking faults are uniformly distributed in the material, the average loop size is ~ (30–60) nm, concentration – ~ $(0.4–0.7)\cdot10^{21}$ m^{-3}.

Fig. 12. Cellular dislocation structure of Cr16–Ni19 cold-worked steel, characteristic of the high-temperature irradiation range, $T_{irr.}$ ~ 590°C, G ~ $0.6\cdot10^{-6}$ dpa/s: a – bright-field STEM image of a triple grain junction, polygonization dislocation walls are visible; b – nickel distribution map in the studied area, RIS at grain boundaries, dislocations and low-angle dislocation boundaries.

In the lower part of the high-temperature range, $M_{23}C_6$ (τ) and M_6C (η) type carbide precipitates predominate in the grain volume and at the boundaries; most particles are conglomerates of carbide-intermetallic in various combinations of two or three alternating phases ηG *G*, τ– *G*, Fig. 13a–e. The average size of second phase particles is larger compared to the size of particles observed in the previous temperature range, and the concentration of particles decreases with increasing irradiation temperature, while a heterogeneous distribution of intragranular precipitates is observed throughout the material volume. In addition to the main relatively large phases τ, η and *G,* as well as rod-shaped phosphides (Fig. 13), coherent fine-dispersed ~ (5–10) nm particles based on nickel and titanium, most likely, cubic γ'–phase Ni_3Ti (*a* ~ 0.35 nm), Fig. 13 and [8].

Fig. 13. Morphology of precipitates in Cr16-Ni19 c.w. type steel, characteristic of the lower edge of the high-temperature irradiation range, $T_{irr.}$ ~ 570°C, *G* ~ $1.3\cdot10^{-6}$ dpa/s: a - bright-field STEM image, τ-carbide precipitates, M_2P phosphides and Frank loops (SF) are marked; b-d - distribution maps of Cr, Ni, Ti. Fine particles of Ni_3Ti and *G-phase* in conglomerates with τ-carbides are marked; e, f - element distribution profiles of Cr, Ni, Si, Mo, Ti, P through a three-layer conglomerate τ + G + τ and phosphide M_2P, enriched with Si and Ti.

At irradiation temperatures ~ 590°C and above, precipitation of fine ~ (5-10) nm TiC carbide precipitates occurs in the grain volume, coherently oriented in the austenitic matrix (Fig. 14). The local concentration of carbide particles correlates with the local dislocation density, therefore the highest concentration is observed in dislocation clusters, for example, along the boundaries of the cellular structure.

Fig. 14. Fine TiC precipitates in Cr16-Ni19 c.w. type steel, T_{irr} ~ 630 °C, *G* ~ $0.6\cdot10^{-6}$ dpa/s: a - morphology of fine carbides; b - element distribution profile of Ti, C, Ni, Si through a secondary titanium carbide particle; c, d - HR TEM and corresponding FFT with interpretation, obtained on a coherent carbide particle. The identified reflections belong to the zone axis [-1 -1 0] of the austenitic matrix, the corresponding reflections of TiC with lattice parameter *a* ~ 0.433 nm are circled.

In the upper part of the high-temperature range, the formation of the HCP Laves phase of the Fe_2Mo (*a* ~ 0.475 nm, *c* ~ 0.773 nm) type occurs. The Laves phase can already appear at an irradiation temperature of ~ 590°C, forming primarily on precipitates of primary carbonitrides.

Fig. 15. Laves phase in Cr16-Ni19 cold worked steel in the range of maximum irradiation temperatures, T_{irr} ~ (625-630) °C, *G* ~ $(0.6\text{-}0.8)\cdot10^{-6}$ dpa/s: a, b - morphology and DP with interpretation of high-resolution image of the Laves phase (λ) formed on the periphery of the primary carbonitride M(C,N) particle; c, d - STEM image in the vicinity of the triple junction of grains and the corresponding composite map of Cr and Mo distribution, grain boundary precipitates of the Laves phase and τ-carbide and intragranular precipitates of the Laves phase on primary carbonitrides are marked; e, f - distribution profiles of Fe, Mo, Cr, Ni, Si across the cross-section through the particles of the Laves phase (e) and τ-carbide (f).

Most often, the Laves phase forms as petals growing from the carbonitride surface into the matrix (Fig. 15a). The Laves phase has a specific twinned structure [9], which makes it easy to identify by morphological features. The presence of fine twins in the Laves phase structure is also evidenced by the streaks in the diffraction pattern (Fig. 15b).

After irradiation at a temperature of ~ 630°C, the Laves phase intensively precipitates at grain boundaries, forming extended layers (Fig. 15c, d). Along with the Laves phase, carbides of the $M_{23}C_6$ type are found at grain boundaries and on primary carbonitrides (Fig. 15d). Both τ-carbides and the Laves phase have similar content of molybdenum and silicon in their composition, but they differ significantly in chromium and iron content (Fig. 15e, f).

At the irradiation temperature of ~ 630°C, *G and* γ'-phases are practically not found in the grain volume, only secondary fine dispersed TiC carbides and M_2P phosphides of needle (rod-like) shape, usually enriched with titanium and silicon, are observed.

DISCUSSION OF RESULTS

Analysis of the results shows that irradiation of Cr16-Ni19 cold-worked steel leads to significant changes in the material microstructure. The dislocation structure undergoes significant reorganization at low irradiation doses ~ 0.1 dpa, in samples with atomic displacement generation rates $G \sim 5 \cdot 10^{-9}$ dpa/s and operating temperature ~ 370°C, the structural state of the cladding material is characterized by a relatively uniform distribution of dislocations, i.e., there is no cellular structure typical for the distribution of dislocations in non-irradiated cold-worked state.

With increasing irradiation temperature, the dislocation structure evolves from a uniformly distributed dislocation network to the formation of dislocation walls and the formation of a secondary cellular structure. Initially, locally disoriented areas are observed near grain boundaries due to polygonization processes, which intensify with increasing temperature, encompassing the entire grain volume and forming a subgrain structure. The average density of intragranular dislocations monotonically decreases with increasing irradiation temperature (Fig. 16a).

Fig. 16. Dependencies of dislocation density (a), average size and concentration of Frank loops (b) on irradiation temperature in Cr16-Ni19 cold-worked steel.

After irradiation, radiation-induced Frank dislocation loops with stacking faults are always present in the cladding material. With increasing temperature, the average size of the loops monotonically increases, while the concentration decreases, reaching a plateau in the temperature range of (520-530)°C (Fig. 16b). Grain boundaries that are initially equilibrium (straight) begin to migrate with increasing irradiation temperature, forming local bends.

At all irradiation temperatures, segregation of alloying elements Ni and Si is observed at grain boundaries and dislocations. With increasing irradiation temperature, phase formation processes, both radiation-stimulated and radiation-induced, are added to the segregation processes. Radiation-stimulated phases include those that can form in an unirradiated alloy during prolonged aging, for example, at higher temperature or longer exposure; such phases include complex carbides of the $M_{23}C_6$ and M_6C type, Laves phase (AB_2), phosphides of the Fe_2P type, secondary fine-dispersed monocarbides [10, 11]. Radiation-induced phases include those that are not detected in the unirradiated state, for example, intermetallic *G* -phase and γ'-phase, fine-dispersed titanium nickelides (Ni_aTi_b). Table 1 shows the phases detected at various irradiation temperatures both within grains and along the boundaries of grains and primary carbonitride precipitates.

With increasing irradiation temperature, the size of intragranular G-phase precipitates *G* -phase and complex FCC carbides increases, while their concentration decreases, and the non-uniformity of their distribution throughout the material volume increases. At temperatures above 510°C, these phases form as conglomerates with common boundaries.

Table 1. Secondary Phase Precipitation in Cr16-Ni19 Cold-Worked Steel at Different Irradiation Parameters

Temperature Range, °C	G, $\cdot 10^{-6}$ dpa/s	Secondary Phase Precipitates					
		intragranular		grain boundary		on primary M(C, N)	
		type	size, nm	type	Grain boundary coverage	type	thickness, nm
420–455	1.1–1.4	*G*	20–40	predominant *G*	non-uniform	*G*	up to 30
		$M_{23}C_6$		$M_{23}C_6$		$M_{23}C_6$	
		γ′	10–15				
485–550	1.5–1.6	*G*	30–50	*G*	non-uniform	*G*	30
		$M_{23}C_6$	70–120				
		M_6C	up to 30	predominant $M_{23}C_6$		$M_{23}C_6$	
		γ′	20–30				
		M_xP	10–50				
570–600	1.6–1.2	*G*	50	*G*	High, especially large precipitates at triple grain junctions	$M_{23}C_6$	not determined
		$M_{23}C_6$	up to 200				
		M_6C	up to 200	predominant $M_{23}C_6$			
		γ′	10–25				
		M_xP	20–80				
		Ni_3Ti	10–20				
		TiC	5				
590–630	0.8–0.6	M_xP	20–80	predominant Fe_2Mo	High	Fe_2Mo	up to 150
		TiC	5	$M_{23}C_6$			

The size of the Si-based γ'-phase precipitates increases with the irradiation temperature from 420°C to 520°C, where it reaches its maximum; further increase in irradiation temperature leads to a slight decrease in size. With increasing irradiation temperature, this phase becomes more enriched with Ti. At temperatures above 540°C, fine dispersed phases of titanium nickelide (Ni_3Ti) and titanium carbide (TiC) are observed. At irradiation temperatures above 485°C in the grain bodies of Cr16-Ni19 cold-worked steel, phosphide formation is observed, the size of which increases with rising irradiation temperature, and phosphides become enriched with Si and Ti.

G-phase precipitates and complex FCC carbides form along grain boundaries; with increasing irradiation temperature, the grain boundary coverage by precipitates increases, and $M_{23}C_6$ carbides begin to predominate, reaching particularly large sizes (more than 200 nm) at triple grain junctions at temperatures above 540°C. With increasing irradiation temperature, carbides become enriched with Si, Ti, and P. At temperatures above 590°C, Laves phase precipitates are observed at grain boundaries, which become predominant at a temperature of ~ 630°C.

At irradiation temperatures above 420°C, G *G*-phase and complex FCC carbide precipitates are observed at the boundaries of some primary carbonitrides; with increasing irradiation temperature, the number of primary carbonitrides with second-phase precipitates at their boundaries increases. At temperatures above 590°C, Laves phase precipitates form at the boundaries of primary carbonitrides.

Fig. 17. Relative change in concentration of main elements in the austenitic matrix composition depending on irradiation temperature in fuel rod cladding samples #1 (a) and #2 (b) made of Cr16-Ni19 cold-worked steel.

Figure 17 shows the change in concentration of main elements in the austenitic matrix composition depending on irradiation temperature, using the example of two fuel rod claddings irradiated to similar maximum damage doses but with somewhat different temperature distributions along the cladding height. It has been established that with increasing temperature and dose accumulation rate, due to solid solution decomposition, significant depletion of Ni and Si occurs in the austenitic matrix, with a corresponding increase in the proportion of Cr and Fe.

The minimum values of the relative change in nickel concentration occur in the temperature range (550–570) ° °C for one cladding and (570–600) ° °C for the other. For silicon, the concentration dependence on temperature on both claddings has an oscillating character. In the upper part of the claddings, where the temperature reaches maximum values and the dose accumulation rate decreases, the volume fraction of radiation-induced phase precipitates is noticeably lower than in the previous temperature range, so the depletion of the solid solution in silicon and nickel occurs to a lesser extent.

CONCLUSION

Data were obtained on the microstructure of the fuel rod cladding material made of Cr16–Ni19 cold-worked steel after irradiation in the BN-600 reactor at different temperatures and atomic displacement generation rates.

The fuel rod cladding material made of Cr16–Ni19 cold-worked steel undergoes significant structural and phase changes during operation, caused by diffusion processes accelerated by irradiation. It has been established that even at relatively low operating temperatures of the fuel rod cladding in the gas plenum area (~ 370 ° °C) and minimum damaging doses (≤ (0.1–0.3) dpa), the evolution of the initial dislocation structure occurs, and the process of segregation of the austenitic matrix solid solution is observed – the most mobile elements Ni and Si segregate on dislocations, grain boundaries, and on austenite – M(C, N) interphase boundaries.

At higher temperatures and damaging doses during operation, more significant phase transformations occur, resulting in the appearance of complex FCC carbide precipitates (M 23 C 6 , M 6 C) and intermetallics (G *phase,* Laves), fine precipitates of γ′, Ni 3 Ti, M 2 P , MC.

As a result of these processes, the physical and chemical properties of the fuel rod cladding material change, the cladding swells, and mechanical properties degrade.

CONFLICT OF INTERESTS

The authors of this work declare that they have no conflict of interest.

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