Radiation thermal processes in Cr13Mo2NbVB steel – the material of the fuel assembly shell in reactor BN-350 under mechanical tests

A S Larionov[1], A S Dikov[1], V P Poltavtseva[1], S B Kislitsin[1],
M V Kuimova[2] and A V Chernyavskii[2]

[1] Institute of Nuclear Physics, Almaty, Republic of Kazakhstan

[2] National Research Tomsk Polytechnic University, Tomsk, Russia

E-mail: poltavtseva@inp.kz

Abstract. Regularities of changes of structural-phase state and mechanical properties of steel 13Mo2NbVB – the material of the fuel assembly shell in reactor BN-350 after various mechanical tests at 350°C are experimentally studied. The formation of microprecipitations FeMo, enriched or depleted with molybdenum was found in the short-time mechanical tests, which is the cause of thermal hardening of irradiated Cr13Mo2NbVB steel and its destruction by the ductile-brittle mechanism. On the basis of long-time creep tests it was shown that the material of the spent fuel assembly shell has sufficient resource for long-time storage in the temperature and force conditions simulating long-time storage of spent nuclear fuel.

1. Introduction

The majority of countries using nuclear energy, due to the limited capacity at the present for the reprocessing of spent nuclear fuel (SNF), have chosen direct disposal [1]. This period of exposure depends on the fuel burn-up (at higher burn-up the holding period can last for decades). At the second stage SNF may be directed to the ultimate disposal [1-3] or temporary dry storage for a long period (10^4-10^5 years), during which occurs the reduction of activity, heat input and the radioactivity of removed waste to a safe level [4].

The experience of recent decades shows that storage of spent fuel in dry conditions is becoming more popular, especially for new storages on off-site new nuclear power stations.

In order to justify the concept of dry storage of spent nuclear reactor designs, we proposed the studies of structural materials of the fuel assembly shells to long-time strength in the temperature and force conditions simulating long-time storage of spent nuclear fuel, and forecasting on their basis the behavior of the material during long-time storage. Conducted studies of austenitic and ferritic steels of different fuel assembly shells on fast neutrons [5, 6] have shown the prospectivity of such direction.

The article presents the experimental results of studies of changes in mechanical properties and structure of ferritic Cr13Mo2NbVB steel – material of the fuel assembly shell in reactor on fast neutrons BN-350 after various mechanical tests at a temperature of 350°C.

2. Material and experimental methods

As an object for the research we chose ferritic Cr13Mo2NbVB steel – the material of hexagonal fuel assembly shell of reactor fuel assembly (RFA) of reactor on fast neutrons BN-350 after prolonged exposure to fast neutron irradiation. At the mark of "500 mm" from the center of the core at conditions: T ~ 365°C; dose ~ 6.3 dpa; velocity of dose rate ~ 2×10^{-8} dpa/s.

According to the certification at the initial state we have Cr13Mo2NbVB steel with body-centered cubic lattice (a = 0.2866 nm) as a solid solution of Cr and Mo in iron, with carbide (Nb (V) C) hardening. Elemental composition (wt.%): Cr: – 13.2; Mo – 1.5; C – 0.11; Mn – 0.6; Nb – 0.5; V – 0.2; Si – 0.5; Fe – all the rest.

Due to the high activity of the irradiated material of hexagonal fuel assembly shell of spent RFA, the size of flat specimens for mechanical testing was $0{,}020 \times 0{,}002 \times 0{,}0003$ m^3. The samples of the irradiated and non-irradiated material were cut by the method of spark cutting from the sheet of face of the fuel assembly shell transversely with respect to its axis direction. The surface areas of the samples near the break before the investigation of the structure were subjected to mechanical grinding and polishing, chemical etching in the electrolyte: 70% C_2H_5OH; 17% HF; 13% HCl, the etching time is 20 minutes.

Long-time mechanical strength tests at various applied strains of duration 1500-1800 hours and short-time mechanical testing (at a rate of 0.5 mm/min) were performed at 20 and 350°C on a universal testing machine LR5K Plus in air environment. The studies of changes in the structure and the hardening/softening of materials after mechanical testing were performed by X-ray diffraction, optical and scanning electron microscopy and microhardness measurement with the use of diffractometer D8 ADVANCE, microscopes AxioObserver and JSM-7500F (JEOL), microhardness PMT-3M.

3. Experimental results

Figure 1 shows the results of the calculation with the steps of 1 hour creep rate on the basis of the test chart on the long-time strength at 350°C Cr13Mo2NbVB steel – the material of the fuel assembly shell in reactor BN-350. The choice of test temperature and applied strain 166 MPa was due to the temperature and force conditions typical for storages of long dry storage of spent nuclear fuel.

Clearly, the creep rate increases twofold with the increase of applied strain from 166 to 642 MPa. Regardless of the magnitude of the applied strain, the curves of the creep rate, depending on the duration of the test, three stages are observed, the duration of which is reduced with the increase of applied strain. In the studied range of test duration the destruction of samples did not happen and there were no visible traces of external damage.

The degree of reduction of the creep rate on the linear portion of the first stage is 2.5 times higher for higher applied strain (figure 1, curve 2). With further increase in the duration of the test at this stage, it decelerates in ~ 3 and ~ 5 times, respectively, for low (figure 1, curve 1) and high (figure 1, curve 2) of the applied strain.

In the second stage it is characteristic the increase in the creep rate of 1.4 and 2 times respectively, for high and low applied strain, while in the third stage creep rate increases two times, regardless of its magnitude. However, with further increase in the duration of the test, we noticed a sharp decrease in the rate of creep, with the exception of the second stage in the case of low applied strain. A distinctive feature of this stage is the presence of a time interval of about 400 hours at a constant rate of creep. It is important to emphasize that regardless of the applied strain, the creep rate is compared with the duration of almost 1,500 hours.

The obtained results of long-time creep tests at 350°C, which is only slightly lower than the temperature of irradiation of the material of the fuel assembly shell, indicate the presence of at least two types of the most effective stoppers of glide dislocations of radiation origin when exposed to high and low applied strain, respectively. It can be concluded that the Cr13Mo2NbVB steel – material of the fuel assembly shell has sufficient resources for long-time storage in the temperature and force conditions simulating long-time storage of spent nuclear fuel.

To determine the mechanical properties of Cr13Mo2NbVB steel were held short-time mechanical tests (SMT) at a temperatures of 20 and 350°C. Stress-strain diagrams at 20°C exposed and unexposed Cr13Mo2NbVB steel are presented in figure 2.

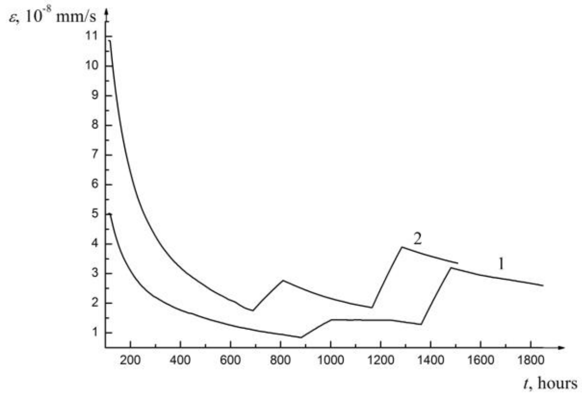

Figure 1. The dependence of creep rate Cr13Mo2NbVB steel at 350°C from the time at the strain of 166 MPa (curve 1) and 642 MPa (curve 2)

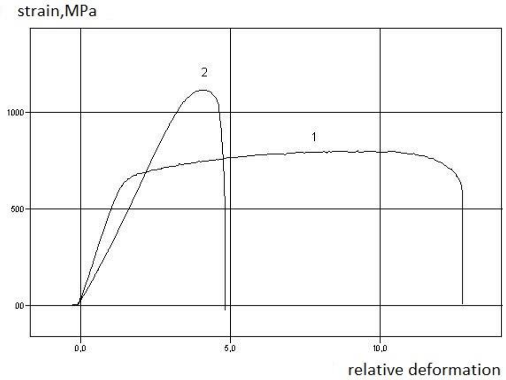

Figure 2. Stress-strain diagram at 20°C non-irradiated (curve 1) and irradiated Cr13Mo2NbVB steel (curve 2)

In the initial state the investigated steel has high ductility and a wide plasticity region necessary for the operation of the material in the fields of alternating power reactors (curve 1). Contrast with the non-irradiated steel, the material of the fuel assembly shell after long-time operation in the BN-350 radiation-hardened (curve 2). With the temperature increase of the tests to 350°C, the region of ductility of non-irradiated steel decreases 1.7 times and the elongation curve of irradiated steel does not change virtually, which is apparently due to similar values of the test temperature and radiation.

Using scanning electron microscopy it was revealed the presence in the structure of fracture non-irradiated Cr13Mo2NbVB steel after the SMT at 20°C the presence of knife-shaped cut with cup-shaped elements and practically complete absence of fractographic zone because of high ductility of the tested material. With the increase of the test temperature, the form of the cut was virtually unchanged. Irradiated Cr13Mo2NbVB steel is also characterized by knife-shape cut, but with elements of cleavage. Moreover, the cleavage area increases with the increase of test temperature. Therefore, the destruction of the irradiated Cr13Mo2NbVB steel occurs by the ductile-brittle mechanism.

Based on the analysis of mechanical characteristics of the Cr13Mo2NbVB steel indicated in table 1, it is shown that with the increase of the test temperature from 20 to 350°C, conventional yield strength $\sigma_{0.2}$ of non-irradiated and irradiated Cr13Mo2NbVB steel decreases to 4 and 22% respectively.

Table 1. Mechanical characteristics of Cr13Mo2NbVB steel

Material	Temperature of test, °C	$\sigma_{0.2}$, MPa	$\sigma_в$, MPa	Δl, mm	H_μ, MPa
Non-irradiated	20	729.0	803.6	0.18	320
	350	690.1	731.2	0.08	380
Irradiated	20	875.6	1116.8	0.05	320
	350	677.6	1005.2	0.05	350

Consequently, the main contribution to reducing $\sigma_{0.2}$ of irradiated steel is mainly due to radiation-induced processes that occur during long-time operation of the material of the fuel assembly shell. Further, we note that the contribution to the reduction of modulus of rupture of the irradiated Cr13Mo2NbVB steel is attributed to the thermal exposure during mechanical tests, whereas a zero change of elongation Δl at the increase of the test temperature, as noted above, is due to close test temperatures and radiation.

According to the microhardness measurements, irradiated Cr13Mo2NbVB steel is softer by ~ 3% (340 MPa) as compared to the initial state (350 MPa). As a result of SMT at 20°C both steels are softened; at higher temperature, on the contrary, hardened, where non-irradiated steel hardens more (table 1).

Figure 3 shows the diffraction patterns of non-irradiated Cr13Mo2NbVB steel taken with the pace of 0.07 and storage time of 7 s. before and after short-time tests at 350°C. From the comparison of diffraction patterns it can be seen that the phase composition of non-irradiated steel changes as a result of tests at 350°C (curve 2). Compared with the initial state (curve 1), three X-ray reflection lines are observed, which are, according to data files, identified as a combination close to FeMo with - body-centered cubic lattice (a = 0.3119 nm).

The amount of FeMo in the non-irradiated steel after the tests at 350°C, determined on the base of relations I_{FeMo} / I_{Fe}, is $\geq$ 50%. At that, the main line of FeMo is a superposition of 2 or more lines belonging to combinations of similar composition, indicating a prolonged shoulder from the side of higher angles 2θ. Further, it should be noted that in the initial state, this combination is also present, though in a small amount, according to the slightly outstanding line above the background at 2θ=40,87°.

According to the research data, by electron scan microscopy (figure 4) in the microstructure of non-irradiated Cr13Mo2NbVB steel after SMT at 350°C, one can clearly observe light (1) and darker microprecipitations (2) and microcracks of 5 to 25 nm, mainly located at the boundaries of these microprecipitations data. From the comparison of the elemental composition (table 2) it follows that the difference is determined mainly by the ratio of Fe and Mo in them. Thus, for light microprecipitations, approximately equal amount of both elements is typical, whereas for darker microprecipitations strong enrichment of Mo is typical.

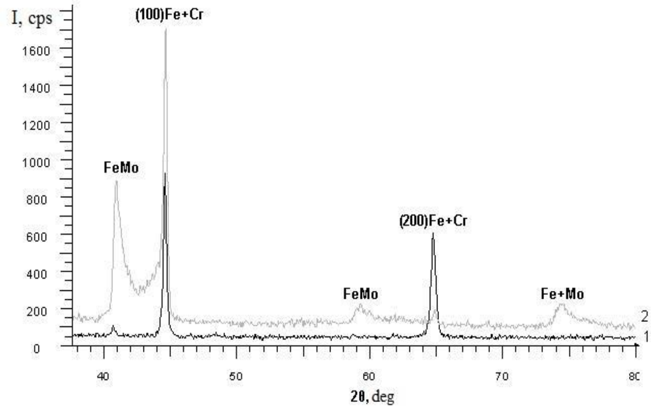

Figure 3. Sections of diffraction patterns of non-irradiated Cr13Mo2NbVB steel before (curve 1) and after SMT at 350°C (curve 2)

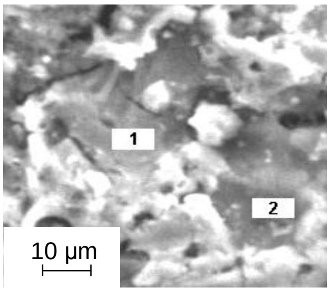

Figure 4. SEM image of the structure of Cr13Mo2NbVB steel after SMT at 350°C: 1 – ${}_{36}Fe_{40}Mo$; *2* – ${}_{17}Fe_{74}Mo$

However, such release is not detected after the test at 20°C. Only a slight reduction of carbides was found in comparison with microstructure obtained before the tests. Consequently, during the SMT at 350°C the formation occurs as a result of the cleavage of microprecipitations, enriched or depleted with Mo respectively [7].

Table 2. The elemental composition of non-irradiated Cr13Mo2NBVB steel after SMT at 350°C

№ microprecipitations	Elemental composition (in weight %)					
	Fe	Mo	Cr	O	Al	Si
1	36.55	40.30	6.89	3.78	0.38	0.34
2	16.82	73.89	2.72	6.58	-	-

Conducted studies of the structure by the method of optical metallography of irradiated and non-irradiated Cr13Mo2NbVB steel also indicate the formation of microprecipitations after SMT at 350°C, but their amount is little and the size is a bit more in the irradiated steel than for the initial state. It is also shown that there are virtually no microprecipitations after tests at 20°C, whereas an inconsiderable proportion of small microprecipitations presents in the microstructure of the non-irradiated steel before the test, which is consistent with the results of X-ray diffraction analysis. Further, is should be noted that in the material after long-time operation in the BN-350 reactor, primary carbides and microprecipitations are virtually unobserved, and this is the reason for the softening of the material.

4. Conclusion

As a result of the experimental research, were found the peculiarities of changes in the structure, phase composition and mechanical characteristics of Cr13Mo2NbVB steel – the material of the fuel assembly shell in reactor at fast BN-350 neutrons after various mechanical tests at 350°C. On the basis of long-time creep tests it is shown that the material of the spent fuel assembly shell has sufficient resources for long-time storage in the temperature and force conditions simulating long-time storage of spent nuclear fuel.

It is established that the decrease in conditional limit of ductility $\sigma_{0.2}$ and modulus of rupture σ_b is mainly due to radiation-induced processes that occur during long-time operation of the material of the fuel assembly shell, and thermal effect in the short-time mechanical tests at 350°C respectively.

The formation of FeMo microprecipitations is observed, enriched or depleted with molybdenum during mechanical tests at 350°C, which is the reason for additional thermal hardening of irradiated Cr13Mo2NbVB steel and its destruction by the ductile-brittle mechanism.

Acknowledgements

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