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Mechanical properties and microstructure of three Russian ferritic/martensitic steels irradiated in BN-350 reactor to 50 dpa at 490 °C

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Abstract

Ferritic/martensitic (F/M) steels are being considered for application in fusion reactors, intense neutron sources, and accelerator-driven systems. While EP-450 is traditionally used with sodium coolants in Russia, EP-823 and EI-852 steels with higher silicon levels have been developed for reactor facilities using lead–bismuth coolant. To determine the influence of silicon additions on short-term mechanical properties and microstructure, ring specimens cut from cladding tubes of these three steels were irradiated in sodium at 490 °C in the BN-350 reactor to 50 dpa. Post-irradiation tensile testing and microstructural examination show that EI-852 steel (1.9 wt% Si) undergoes severe irradiation embrittlement. Microstructural investigation showed that the formation of near-continuous χ-phase precipitates on grain boundaries is the main cause of the embrittlement.

1. Introduction

Ferritic/martensitic (F/M) steels are used in various fast reactor facilities and are considered as candidate materials for application in fusion reactors, intense neutron sources and accelerator-driven systems. Their advantages over other structural materials are higher resistance to void swelling, lower irradiation creep rate, and relatively low induced radioactivity compared to most austenitic alloys. It is known, however, that one of the shortcomings of F/M steels is their tendency to develop low-temperature irradiation embrittlement.

EP-450 is a typical 12% Cr steel commonly used for nuclear applications in Russia and other former Soviet countries with properties similar to Western 12% Cr steels [1]. Two Russian F/M steels designated EP-823 and EI-852 were developed specially for reactor facilities with lead–bismuth coolant [2]. The corrosion resistance of these steels in the

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coolant was obtained primarily by a higher content of silicon. To elucidate the influence of silicon additions on short-term mechanical properties and microstructure, three F/M steels EP-450, EP-823 and EI-852 with different contents of silicon were irradiated in the form of fuel cladding tubes in the BN-350 fast reactor in Kazakhstan.

2. Experimental details

The chemical composition of F/M steels EP-450, EP-823 and EI-852, and the sizes (diameter and thickness) of cladding tubes from which ring specimens for mechanical tests were cut are shown in Table 1.

The heat treatment for the EP-450 tubes consisted of quenching from 1050 °C followed by tempering at 780 °C for 1 h. The EP-823 tubes were normalized at 1050 °C and aged at 740 °C for 1.5 h. The EI-852 steel was aged at 850 °C for 0.5 h but no normalization or tempering was used.

Ring specimens of 2 mm length were cut from these tubes for measurements of mechanical properties and additional rings of 4 mm length were cut for electron microscopy. These rings were irradiated in flowing sodium in a special fuel assembly in the BN-350 fast reactor. This assembly used short fuel pins in the bottom of the assembly, which allowed heating of the specimens in the upper part of the assembly to 490 ± 10 °C. The neutron fluence was 1.12×10^{22} n/cm^2 ($E > 0.1$ MeV) corresponding to 50 dpa (NRT) at 1.47×10^{-7} dpa/s.

Mechanical properties of both non-irradiated and irradiated 2 mm specimens were measured using a standardized Russian ring-pull test (see Refs. [3,4] for details) in a shielded tensile testing machine at test temperatures ranging from 20 °C to 700 °C. The pre-test aging time for the specimens at any test temperature was 20 min. Two or three separate specimens were tested at any given test temperature. Fractography of fracture surfaces was not conducted so no determination of fracture mode was made.

Disks of 3 mm diameter were punched from the 4 mm rings and prepared using the two-jet 'TENUPOLE' polishing technique with an electrolyte of 5% $HClO_4$ + 95% acetic acid. The microstructure was observed using a JEM-100CX electron microscope operating at 100 kV and equipped with a lateral goniometer.

3. Results

Measured mechanical properties of unirradiated and irradiated F/M steels EP-450, EP-823 and EI-852 at various test temperatures are shown in Figs. 1 and 2. The mechanical properties of the three unirradiated steels are similar, namely, up to the test temperature of 500 °C the ultimate strength decreases rather smoothly, but at higher temperatures, it falls sharply, accompanied by a rise in total elongation. Of the three steels investigated, EI-852 in the unirradiated condition had the lowest strength and the highest ductility.

Neutron irradiation resulted in a significant change of mechanical properties, expressed primarily in hardening. At room temperature the ultimate strength increment equals 649 MPa for EI-852, 398 MPa for EP-450 and 225 MPa for EP-823. With increase of test temperature, the difference in

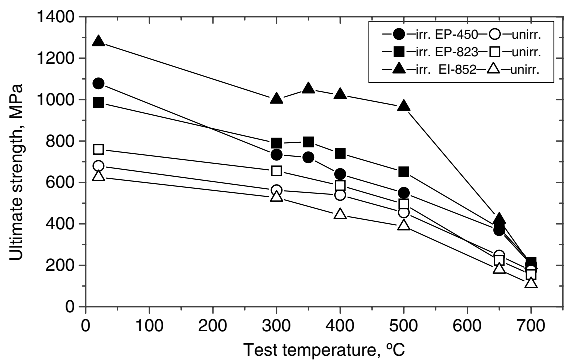

Fig. 1. Ultimate strength of both unirradiated and irradiated steels EP-450, EP-823 and EI-852 versus test temperature.

Table 1
Composition of steels EP-450, EP-823 and EI-852 (wt%)

Steel	Tube size (mm)	Content (wt%)														
		C	Mn	Si	P	S	Cr	Ni	Mo	Nb	V	W	Ti	Al	B	N
EP-450	6.0 × 0.3	0.13	0.32	0.18	0.023	0.006	13.28	0.22	1.5	0.45	0.28	–	–	–	0.004	0.040
EP-823	7.0 × 0.3	0.18	0.6	1.05	0.012	0.008	11.40	0.70	0.67	0.20	0.40	0.65	0.03	0.030	0.004	0.040
EI-852	7.0 × 0.3	0.13	0.52	1.9	0.014	0.012	12.85	0.22	1.66	–	–	–	–	0.014	–	0.034

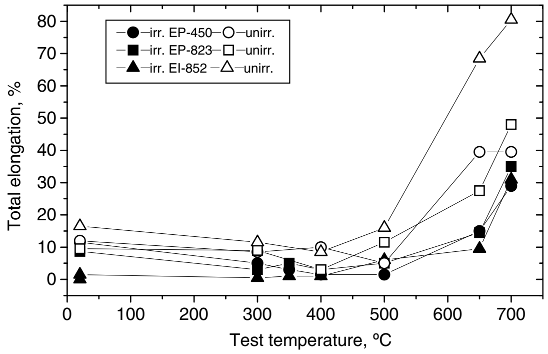

Fig. 2. Total elongation of both unirradiated and irradiated steels EP-450, EP-823 and EI-852 versus test temperature.

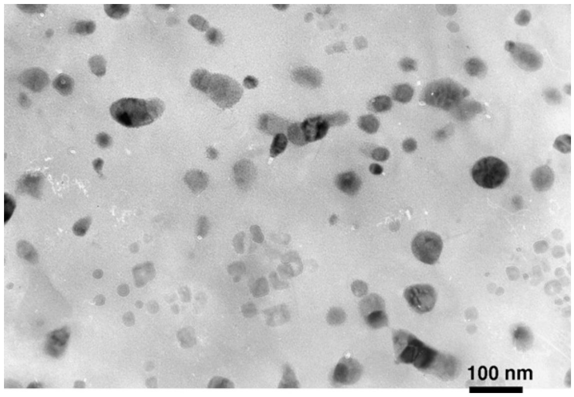

Fig. 3. Bright-field image of microstructure of ferrite grains of irradiated EP-450 at 490 °C and 50 dpa.

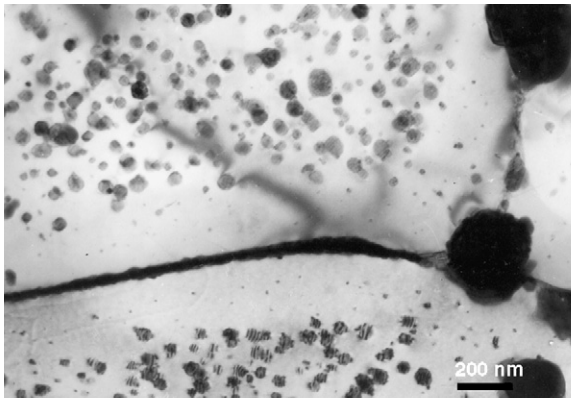

Fig. 4. Bright-field image of zone denuded of χ-phase precipitates in irradiated EP-450 at 490 °C and 50 dpa.

strength between unirradiated and irradiated steels becomes smaller and at a test temperature of 700 °C the difference almost disappears.

The ductility of the irradiated steels vs. test temperature is more complicated. In the range 300–500 °C the total elongation ε_t of EP-450 and EP-823 decreases with increasing test temperature, reaching 1.5% for EP-450 and 3.0% for EP-823, and then sharply rises. In contrast, EI-852 at room temperature has low ε_t values (0–1.5%) and, in the range 300–400 °C, the ductility is consistently lower compared with those of EP-450 and EP-823 steels.

The initial microstructures of EP-450 and EP-823 steels were similar and consisted of ferrite grains and tempered martensite grains. Carbide precipitates of type $M_{23}C_6$ were observed on grain and sub-grain boundaries of the tempered martensite grains. In the ferrite grains, fine needle precipitates of M_2X type were observed. The microstructure of unirradiated steel EP-852 was observed to consist of ferrite grains only, with $M_{23}C_6$ precipitates uniformly distributed in the grains.

Irradiation at 490 °C to 50 dpa has resulted in the formation of voids in all three steels at relatively low concentration ($<10^{18}$ m^{-3}) with diameters up to 20 nm. The distribution of voids in the steels was found to be non-uniform. Voids in ferrite grains, as a rule, were adjacent to precipitates. In grains of tempered martensite, voids were located primarily on sub-grain boundaries. After irradiation, the phase structure of the steels has changed, with both sizes and concentration of $M_{23}C_6$ increased, needle precipitates in ferrite grains of EP-450 and EP-823 dissolved, and new radiation-induced phases formed, as shown in Figs. 3 and 4.

Micro-diffraction analysis of precipitates formed during irradiation has shown that in EP-450 new precipitates are the α′- and χ-phases. In EP-823 and EI-852 only χ-phase has formed. The mean size, concentration, and volume fraction of radiation-induced phases are shown in Table 2.

As shown in Table 2, χ-phase (an intermetallic phase known to be rich in Si) is the major irradiation-induced phase increasing progressively with

Table 2
Mean size, concentration and volume fraction of phases formed under irradiation

Steel	Phase	Mean size (nm)	Concentration (10^{21} m^{-3})	Volume fraction (%)
EP-450	χ	40	0.8	4
	α′	20	1.0	1
EP-823	χ	50	0.8	5
EI-852	χ	36	3.0	12

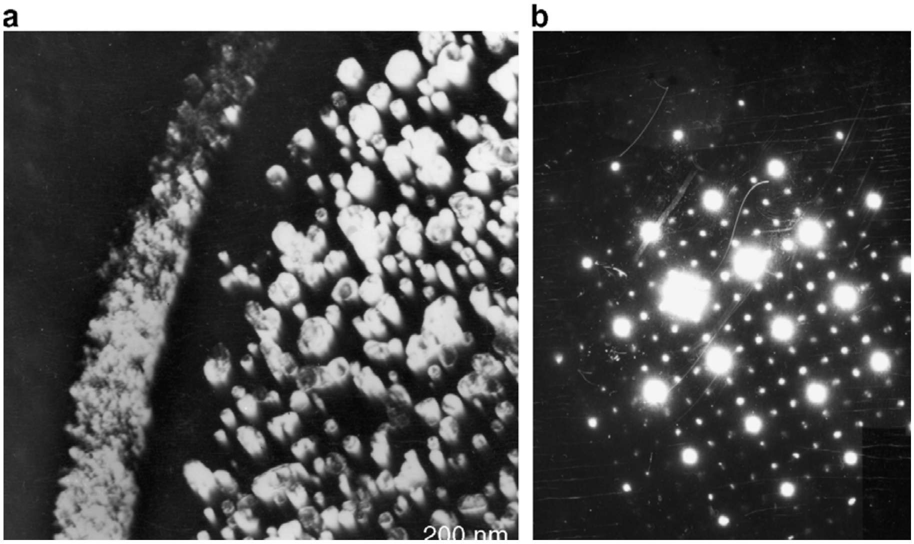

Fig. 5. Dark-field image of χ-phase reflection (a) and diffraction pattern of χ-precipitates (b) of EI-852 at 490 °C and 50 dpa.

silicon content. Note that the volume fraction of χ-precipitates in EI-852 reaches 12%.

There is a significant difference in the spatial distribution of χ -precipitates in the various alloys. In EP-450 and EP-823, these precipitates are rather uniformly distributed within grains, although at grain boundaries a precipitate-denuded zone of ∼300 nm width (Fig. 4) is observed. In contrast, the χ-precipitates in EI-852 have formed both in the grain interior as well as on grain boundaries, with these precipitates forming continuous chains on and near the boundaries (see Fig. 5).

4. Discussion

As mentioned above, F/M steels EP-823 and EI-852 were developed as materials capable of withstanding corrosion in Pb–Bi coolant [2]. The resistance to corrosion was achieved mainly by increased silicon content compared with that of EP-450 steel. Such compositional modifications might be used for fusion applications and therefore it is of interest to compare the effect of neutron irradiation on these three steels.

In all three steels, the void concentration was very low after irradiation to 50 dpa, irrespective of chemical composition and heat treatment. This observation confirms the generality of data showing a high resistance of F/M steels to swelling determined, most probably, by the crystallographic properties of the Fe–Cr bcc matrix.

It should be noted that, in spite of their distinctive chemical compositions, the mechanical properties of EP-450 and EP-823, both before and after irradiation, are similar. Both steels demonstrated rather high strength and ductility at all test temperatures. In contrast, EI-852 experiences much greater hardening and embrittlement following irradiation, and in some cases its ductility is essentially zero.

These data confirm previously published results on the irradiation hardening and embrittlement of the two Si-modified F/M steels when irradiated in the BR-10 and BOR-60 fast reactors at temperatures of 350 to 680 °C [5,6]. One of these studies showed that formation of χ-precipitates at higher irradiation temperatures contributes to ductility reduction [6].

Analyzing the relationship between microstructure and radiation-induced changes of mechanical properties, one can conclude that severe embrittlement of EI-852 steel is caused not only by the greater degree of precipitation of χ-phase in the matrix and therefore higher matrix strengthening, but also arises due to the near-continuous precipitation of χ-phase on grain boundaries. This conclusion is in agreement with results of Refs. [7,8] that show that the formation of χ-phase in ferritic and martensitic steels during either aging or irradiation, results in ductility reduction as revealed in both tensile and impact mechanical tests. In the other two F/M alloys, the matrix is also hardened by precipitation, albeit to a lower level, but the precipitate-denuded zones near grain boundaries allow enough grain boundary sliding during deformation to preserve some ductility. Therefore it appears that

~1 wt% Si is probably an upper limit for alloys developed for corrosion resistance in Pb–Bi coolant.

5. Conclusions

The changes in mechanical properties and microstructure of F/M steels EP-450, EP-823, and EI-852 following irradiation in the BN-350 reactor at 490 °C to 50 dpa can be summarized as follows:

1. All three steels are very resistant to swelling at these irradiation conditions, with voids occasionally observed at very low concentrations.
2. Irradiated EP-450 and EP-823 containing 0.18 and 1.05 wt% of silicon, respectively, demonstrate good mechanical properties both before and after irradiation. Steel EI-852 with 1.9 wt% silicon, however, hardens to a higher level and develops severe irradiation embrittlement.
3. The embrittlement of EI-852 steel by radiation is caused not only by a higher level (volume fraction of 12%) of Si-rich χ-phase precipitates inside grains, but especially by the near-continuous formation of χ-phase on grain boundaries. This latter microstructural feature probably precludes grain boundary sliding that can occur in the precipitate-denuded zones maintained in the other two alloys.
4. Increasing silicon additions appear to offer no large influence on the void swelling of the Russian F/M steels at these irradiation conditions, but the silicon content must be limited to ~1% in order to preserve sufficient post-irradiation ductility.

Acknowledgements

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