Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of Nuclear Materials 329–333 (2004) 314–318
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Microstructure and mechanical properties of ferritic/martensitic steel EP-823 after neutron irradiation to high doses in BOR-60

S.I. Porollo [a], A.M. Dvoriashin [a], Yu.V. Konobeev [a], F.A. Garner [b,*]

[a] *State Scientific Center of Russian Federation, Institute of Physics and Power Engineering, Obninsk, Russia*
[b] *Structural Materials Research, Pacific Northwest National Laboratory, P.O. Box 999, Richland, WA 99352, USA*

Abstract

Mechanical properties and microstructure of ferritic/martensitic (F/M) steel EP-823 were investigated following irradiation in BOR-60 to 63 dpa at 365–680 °C. Up to ~460 °C irradiated EP-823 was found to fracture in a brittle mode at low strength levels, but no significant change in mechanical properties occurred at temperatures >460 °C. When compared to EP-450 often used in Russia it is observed that EP-823 is rather more brittle, with the primary microstructural difference being the formation of M_2X in EP-823 instead of α' precipitates in EP-450. On the other hand, EP-823 appears to be somewhat more resistant to void swelling. Both of these attributes are thought to arise primarily from the higher silicon content of EP-823.

---

1. Introduction

Ferritic/martensitic (F/M) steels are proposed for use in nuclear applications, including fusion devices and accelerator-driven systems, due to their lower radioactivation, higher swelling resistance and lower irradiation creep rate compared to austenitic steels.

The well-characterized F/M steel EP-450 with nominal composition of 0.1wt%C–13%Cr–2%Mo–Nb–V–B has the largest experimental database, having been extensively irradiated in Russian and Kazakh fast reactors. However, for application in facilities using Pb-based coolants the Russian F/M steel EP-823 (0.16wt%C–12%Cr–Mo–W–Si–Nb–V–B) has been developed, possessing an improved corrosion resistance [1]. However, the radiation resistance of EP-823, especially at high dose levels, has not been investigated sufficiently.

2. Material and experimental technique

EP-823 was used as fuel pin cladding in an experimental assembly irradiated in the BOR-60 fast reactor. The cladding tubes were 6.9 mm in external diameter and 0.4 mm thick. The composition was 0.18C–11.4Cr–0.5Ni–0.7Mn–1.1Si–0.6Mo–0.8W–0.4V–0.4Nb, in wt%. The tubes were normalized at 1020–1070 °C with subsequent tempering at 760 °C for 1 h. The pins were irradiated in the 300–680 °C range to a maximum dose of 63 dpa.

After irradiation the pins were cut to sections of 2 or 4 mm width, which were then de-fueled and cleaned in HNO_3. The 2 mm rings were used to measure mechanical properties and the 4 mm rings for electron microscopy. The mechanical properties were measured at test temperatures equal to irradiation temperatures. Testing proceeded on both unirradiated and irradiated specimens using a standardized ring pull test employed in Russia [2].

Microstructural investigation of neutron irradiated specimens was conducted at three locations along the length of one pin, focusing on the middle wall region.

---

[*] Corresponding author. Tel.: +1-509 376 4136; fax: +1-509 376 0418.
E-mail address: frank.garner@pnl.gov (F.A. Garner).

0022-3115/$ - see front matter © 2004 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2004.04.310

Table 1
Irradiation conditions for TEM specimens

Specimen #	Irradiation temperature range (°C)	Mean irradiation temperature (°C)	Dose (dpa)
1	365–385	375	50.5
2	465–535	500	63
3	580–680	630	33

Irradiation conditions of the investigated samples are shown in Table 1. The ranges of irradiation temperature reflect a gradual decline in temperature with increasing burnup.

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

3. Experimental results

The mechanical properties are shown in Fig. 1. Up to 500 °C the unirradiated steel had sufficiently high strength properties. At higher test temperatures the ultimate strength of EP-823 fell sharply (reaching only 270 MPa at 700 °C). Concurrently, the total elongation of the steel increased from 4.2–6.6% in the range of 300–500 °C to 35.4% at 700 °C. For temperatures <460 °C the irradiated specimens failed at low strength levels in brittle mode with zero total elongation. At higher temperatures the mechanical properties of the unirradiated and irradiated steel were comparable.

3.1. Starting structure

The initial structure of EP-823 was a mixture of polyhedral grains of ferrite (10–20%) and tempered martensite (90–80%). The martensite grains consisted of sub-grains having the shape of equally-oriented thin plates 0.5–1.0 μm in width. Equiaxed MC particles of 0.3–0.5 μm diameter at 5×10^{11} cm^{-3} were observed inside the grains. The grain boundaries of both ferrite and martensite, as well as sub-boundaries in the martensite, were partially filled with blocky $M_{23}C_6$ carbides. In addition, plate-like precipitates of M_2X with mean diameter of 70 nm at 5×10^{15} cm^{-3} were observed inside ferrite grains. The dislocation density was $\sim3\times10^{10}$ cm^{-2} in ferrite grains and $\sim1\times10^{12}$ cm^{-2} in martensite sub-grains.

3.2. Post-irradiation structure

Irradiated specimens of EP-823 steel also had a two-phase structure consisting of the ferrite and tempered martensite, with the volume ratio between the two phases unchanged at all irradiation conditions. Isolated small voids (10–20 nm diameter) were observed in some, but not all, ferritic grains for specimens irradiated at 365–385 °C. No voids were observed at higher temperatures.

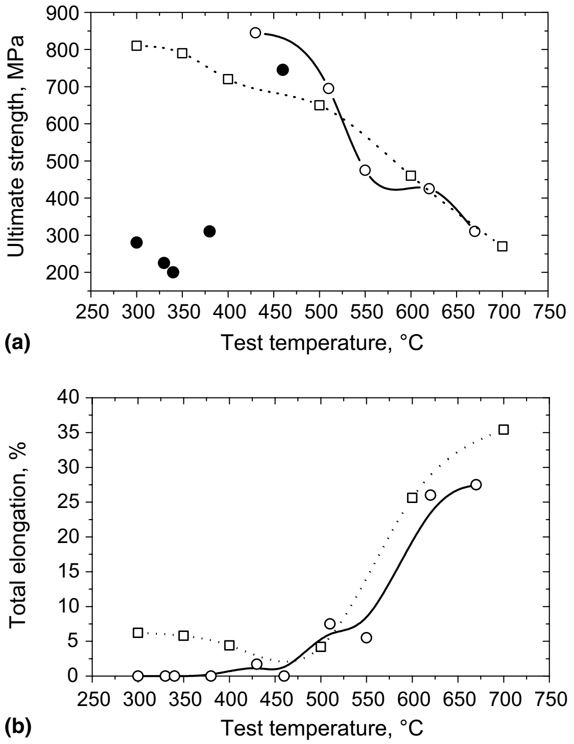

Fig. 1. Strength (a) and total elongation (b) of ring specimens made of EP-823 F/M steel. □ – unirradiated specimens; ○ – following irradiation in BOR-60 reactor to doses of 44–62 dpa with test and irradiation temperatures identical. Strengths corresponding to zero values of total elongation in (b) are shown by dark circles in (a).

At 365–385 °C perfect dislocation loops were uniformly distributed. In ferrite grains the loops had somewhat larger sizes and concentration compared to those in martensite grains. In the ferrite grains the mean loop diameter and loop concentration were equal to 14 nm and 2×10^{16} cm^{-3}, respectively, while in the martensite grains the values were 12 nm and 1.5×10^{16} cm^{-3}. The total dislocation density was 1.5×10^{10} cm^{-2} for ferrite and 2×10^{11} cm^{-2} for martensite grains. The radiation-induced change of dislocation structure in the EP-823 steel was similar for both 465–535 °C and 580–680 °C intervals, consisting of decreases from 2×10^{10} cm^{-2} in ferrite and $(4–5)\times10^{10}$ cm^{-2} in martensite.

Changes in precipitates were determined primarily by the irradiation temperature. At 365–385 °C finely-dispersed spherical M_6C particles with mean size of 5 nm

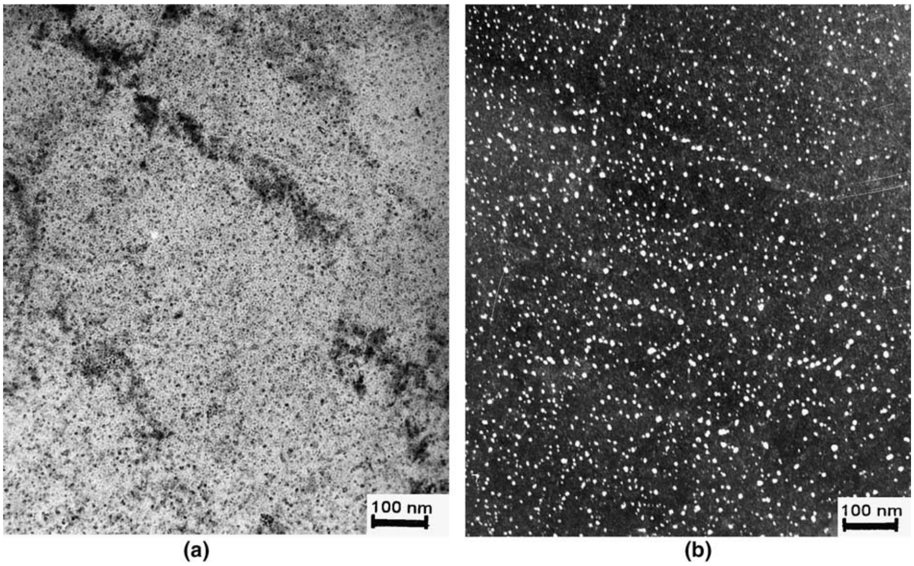

Fig. 2. Fine-dispersed M_6C precipitates in the EP-823 steel irradiated at temperatures in the range of 365–385 °C to dose of 50.5 dpa. (a) bright-field image, (b) dark-field image using precipitate reflection.

and concentration of 5×10^{16} cm^{-3} were observed, distributed randomly in both ferrite and martensite grains as shown in Fig. 2. Pre-existing precipitates did not appear to undergo any noticeable changes.

After irradiation at 465–535 °C, however, two types of precipitates with mean diameters of 13 and 48 nm formed in ferrite grains (Fig. 3), with the exception that a precipitate-free zone of ~ 200 nm in width was observed near grain boundaries. Precipitates of the larger size category were distributed uniformly within the grains, at $\sim10^{15}$ cm^{-3}. Analysis showed these larger precipitates to be intermetallic χ-phase.

Precipitates in the smaller size category were nonuniformly distributed in ferrite grains, forming small clusters (Fig. 4). Because of their small volume fraction it was not possible to unequivocally determine the identity of these precipitates, but they appeared to be M_6C carbides, similar to those at 365–385 °C. The M_6C

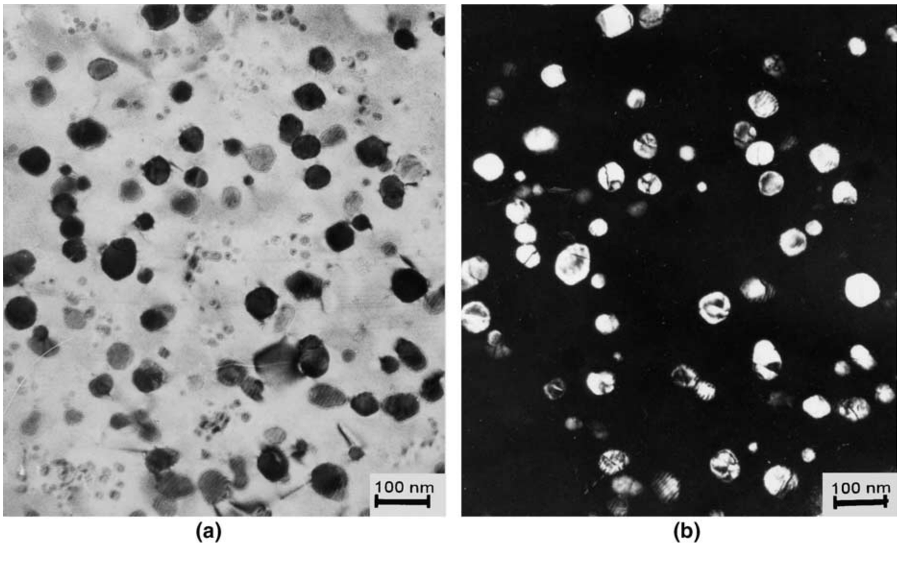

Fig. 3. Precipitates of χ-phase and M_6C precipitates in steel EP-823 irradiated at temperatures 465–535 °C to dose of 63 dpa: (a) bright-field image, (b) the dark-field image using χ-reflection.

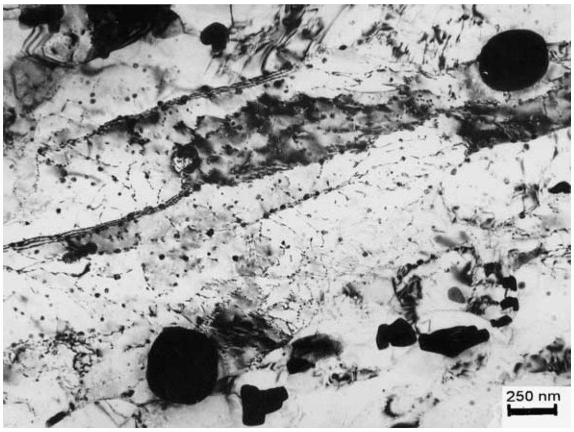

Fig. 4. Microstructure of the tempered martensite in steel EP-823 irradiated at temperatures 465–535 °C to dose of 63 dpa.

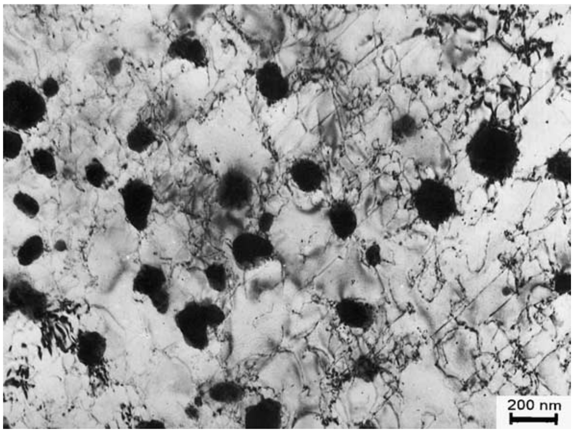

Fig. 5. Precipitates of χ-phase in ferrite grains of steel EP-823 irradiated at temperatures 580–680 °C to 33 dpa.

carbides formed in martensite grains were small in size and concentration and were usually situated on dislocations (Fig. 5). Most large sub-grains of the martensite were observed to possess a low number of precipitates of χ-phase with sizes of 20–40 nm.

Irradiation at temperatures in the 465–535 °C range resulted in complete dissolution of pre-existing M_2X plate particles in ferrite grains. Some growth and increase of the number of $M_{23}C_6$ carbides also took place on grain boundaries, with the degree of carbide occupation of sub-grain boundaries increasing in the martensite.

In ferrite grains irradiation at 580–680 °C resulted in formation of equiaxed particles of χ-phase with mean diameter of 130 nm at 7×10^{13} cm^{-3} (Fig. 5). In martensite the formation of additional precipitates was not observed. Pre-existing M_2X particles in ferrite grains had dissolved completely. On grain boundaries the size of carbides $M_{23}C_6$ increased with formation of continuous chains of 150 nm thickness. The degree of carbide occupation of sub-boundaries increased considerably in martensite, with sizes also increased.

4. Discussion

EP-823 was developed to have high corrosion resistance in lead–bismuth coolants by increasing the silicon content (1.1 wt%) relative to ~0.2 wt% of EP-450 and other common 12% chromium steels. Therefore, it is of interest to compare the radiation behavior of EP-823 and EP-450 steel, the latter having a larger database.

It appears that EP-823 is somewhat more swelling-resistant compared with EP-450 steel, which swells as high as 0.2–0.3% at 380–420 °C and doses ranging from 45 to 56 dpa [3]. One possible reason of higher swelling resistance might be the higher silicon content, which is known to strongly reduce void swelling in austenitic alloys [4,5] and simple Fe–Cr ferritic alloys [6].

Mechanical property data of EP-823 during irradiation at <460 °C show a complete embrittlement and significant reduction of strength. Therefore, EP-450 seems to be more attractive due to its better mechanical properties after irradiation under approximately the same conditions [3]. Residual ductility of the majority of EP-450 ring specimens was ~0.5–1.0%, and only a few failed in brittle mode. It is important to note, that compared to EP-823, all EP-450 specimens retained a high level of strength.

The conclusion that F/M steels with high silicon content suffer stronger embrittlement can be confirmed from Ref. [7], where gas-pressurized creep tubes fabricated from EP-450 and EP-852 (another high-Si F/M steel with 1.91% Si) were compared after irradiation in BN-350 to 61 dpa at 310 °C. In that study only one EP-450 tube out of seven failed during puncturing to release their gas while four EP-852 tubes out of seven failed.

By comparing microstructural features as a function of temperature one can conclude that the embrittlement of EP-823 steel is probably a consequence of formation of high concentrations of both dislocation loops and finely-dispersed precipitates M_2X. It is not clear from these results alone which of these defects are the main contributors to embrittlement. It is possible only to note that the microstructural characteristics (sizes and concentrations of dislocation loops and precipitates) in irradiated EP-823 and EP-450 are similar, except that in EP-823 steel the type M_2X precipitates form, whereas in EP-450 steel α'-phase precipitates form. This is an indication that second phase precipitates probably play the most important role in embrittlement.

5. Conclusions

Irradiation of F/M steel EP-823 irradiated in BOR-60 over the range 365 to 680 °C to 63 dpa leads to rather severe embrittlement at irradiation temperatures <450 °C, but no significant change in mechanical properties at temperatures >450 °C. When compared to EP-450 it is observed that EP-823 is somewhat more brittle, with the primary microstructural difference being the formation of M_2X instead of α' precipitates. On the other hand EP-823 appears to be somewhat more resistant to void swelling.

Acknowledgements

The Russian data presented in this report were generated in irradiation programs conducted under sponsorship of the Ministry of Science and later evaluated under sponsorship of MINATOM. The US portion of this work was jointly supported by the Office of Nuclear Science and Technology (NERI and AFCI programs) and the Materials Science Branch (Basic Energy Sciences, Fusion Energy), US Department of Energy, under Contract DE-AC06-76RLO 1830. Pacific Northwest National Laboratory is operated for the Department of Energy by Batelle Memorial Institute. The assistance of Natalia A. Brikotnina of Interpreter and Translation Services to facilitate discussions between US and Russian participants, and to aid in the preparation of the English draft is greatly appreciated.

References

[1] A.E. Rusanov, V.M. Troyanov, Yu.S. Belomytzev, A.A. Smirnov, G.A. Yachmenev, R.K. Gibadullin, V.N. Sugonyaev, V.M. Pykhtin, in: Proceedings of Conference on Heavy Metal Liquid Coolants in Nuclear Technology, Obninsk, Kaluga Region, Russia, vol. 2, 1999, p. 678.

[2] V.I. Prokhorov, A.G. Finko, R.I. Mineev, Report of the Research Institute of Atomic Reactors, Dimitrovgrad, Russia, 1977 (in Russian).

[3] A.M. Dvoriashin, V.D. Dmitriev, V.S. Khabarov, in: R.E. Stoller, A.S. Kumar, D.S. Gelles (Eds.), 15th International Symposium on Effects of Radiation on Materials, ASTM STP 1125, ASTM, Philadelphia, 1992, p. 1180.

[4] F.A. Garner, W.G. Wolfer, J. Nucl. Mater. 102 (1981) 143.

[5] F.A. Garner, Materials Science and Technology: A Comprehensive Treatment, vol. 10A, VCH, 1994, p. 419 (Chapter 6).

[6] D.S. Gelles, in: N.H. Packan, R.E. Stoller, A.S. Kumar (Eds.), 14th International Symposium, ASTM STP 1046, vol. 1, American Society for Testing and Materials, Philadelphia, 1989, p. 73.

[7] Yu.V. Konobeev, A.M. Dvoriashin, S.I. Porollo, S.V. Shulepin, N.I. Budylkin, E.G. Mironova, F.A. Garner, M.B. Toloczko, Semiannual Progress Report for Period Ending 30 June 2003 DOE/ER-0313/34. 30 June 2003, p. 70.