PHASE TRANSFORMATIONS OBSERVED IN EP-450 FERRITIC/MARTENSITIC STEEL IRRADIATED AT ~ 300ºC TO 40.3 DPA IN THE BN-350 FAST REACTOR–O. P. Maksimkin, L. G. Turubarova., T. A. Doronina (Institute of Nuclear Physics, National Nuclear Centre, Alma Ata, Kazakhstan), and F. A. Garner (Pacific Northwest National Laboratory)

OBJECTIVE

The objective of this effort is to establish the mechanisms by which ferritic/martensitic steels change their microstructure and properties during irradiation.

SUMMARY

At ~ 300ºC, the ferritic/martensitic steel EP-450 was observed to start swelling (0.4%) after irradiation to 40.3 dpa in BN-350. Comparing to similar behavior from a comparable component irradiated to 33.5 dpa and 305ºC, the swelling rate appears to be accelerating strongly in the last 10 dpa.

This two-phase steel is unstable under irradiation with sorbite grains converting to ferrite grains, producing a densification that obscures the onset of void swelling. The relative proportions of the sorbite and ferrite grains in the unirradiated steel was 1.55:1, while in the irradiated steel this relationship changes in favor of the ferritic structure, becoming 1:2.5.

PROGRESS AND STATUS

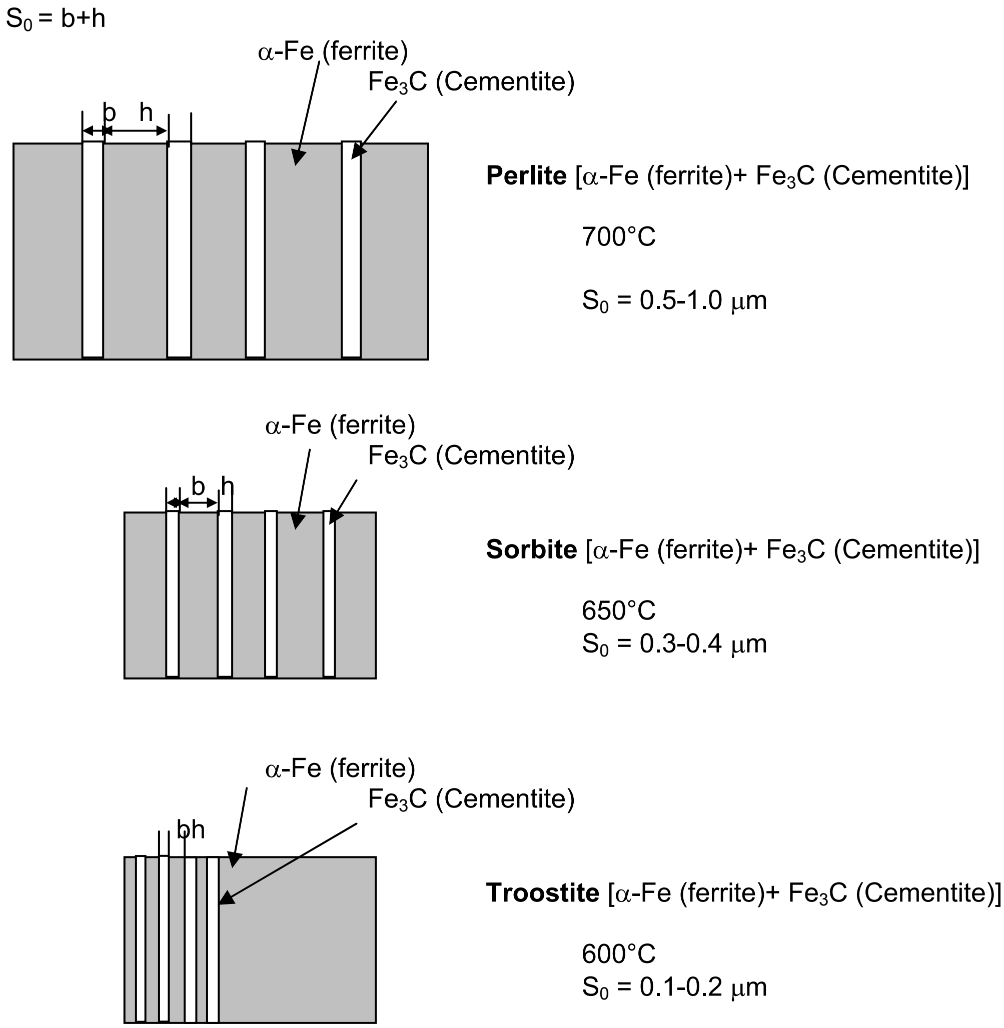
Introduction

Many irradiation experiments conducted on structural materials are rather well-controlled and do not involve large periodic changes in environment, application of varying stress fields, and other features that are characteristic of real reactor operation. It is important that results from such tests be compared with results involving reactor components that were subjected to more realistic histories. For ferritic/martensitic steels that are suggested as structural components in fusion or generation 4 reactors, it is also important to collect data at temperatures that were not attainable in U.S. fast reactors EBR-II and FFTF, since these reactors had inlet coolant temperatures on the order of 365–370ºC. Some Russian-built fast reactors such as BOR-60 and BN-350 have much lower inlet temperatures.

In the current study, we address the behavior of Russian steel EP-450 that served in the BN-350 fast reactor as a protective wrapper for a fuel assembly. The specimen chosen for examination in this first effort reached 40.3 dpa at ~ 300ºC and 0.96×10^{-6} dpa/s.

Experimental Procedure

A hexagonal wrapper (designated № 715.1700.31188) removed from a spent fuel assembly previously irradiated in the BN-350 fast reactor was chosen for study. The wrapper was made from the Russian stainless ferritic-martensitic steel designated as 12Cr13Mo2NbVB, more often known as EP-450. Its composition prior to irradiation was C-0.13; Cr-12.83, Ni-0.14; Mo-1.62; Nb-0.45; Si-0.4; Mn-0.34; V-0.16; B-0.004. The thermal treatment involved normalization at 1050ºC and subsequent annealing at 750ºC for 1 hr. In its initial state, the EP-450 steel is usually employed in a two-phase condition with sorbite dominating over ferrite. Sorbite is a term used by Russians to characterize the desired optimal result of the decomposition of γ-Fe to α-Fe + Fe_3C upon cooling, as shown in Fig. 1.



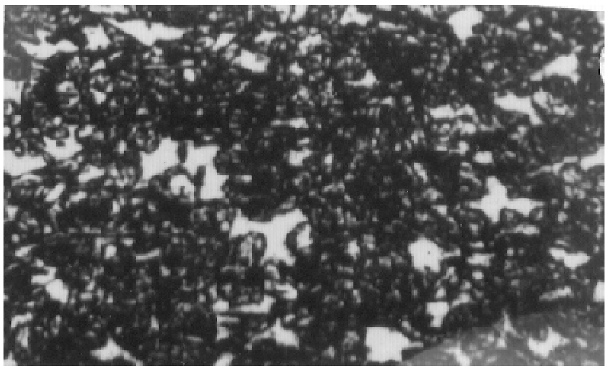
S_0 (perlite) > S_0 (Sorbite) > S_0 (Troostite)

Fig. 1. Transformation of γ-Fe ⇒ α-Fe + Fe_3C with cooling.

A specimen with dimensions 5 x 5 x 2 mm was cut in the hot cell from the wrapper at -375 mm with respect to the core center. The structure of the steel was studied using the MeF-2 optical microscope and the JEM 100CX electron microscope. The irradiated specimens were weighed in air and in water on the KERN 770-12 scales with an accuracy of ±0.05 mg. Vickers microhardness measurements were also performed. Similar measurements were made on an archive duct of the same heat.

Metallographic studies showed considerable differences in microstructure of the steel in unirradiated and irradiated states (Fig. 2). The relative proportions of the sorbite and ferrite grains in the unirradiated steel is 1.55:1, while in the irradiated steel this relationship changes in favor of the ferritic structure, becoming 1:2.5.

It should also be noted that in the unirradiated steel the sorbite grains are very distinct, i.e., the grain boundaries are very clearly defined and always continuous in nature. After irradiation, the boundaries are less distinct, becoming smaller, with their grain boundaries broken and unfinished. It was discovered that many of the "secondary" ferrite grains, i.e., grains that formed as a result of the simultaneous effect of radiation, temperature, and stress, were extended (in some cases in twin formation) along certain directions in a specimen. When viewed on the macroscale, these grains appear to be organized and one can discern certain recurring patterns (Fig. 3).



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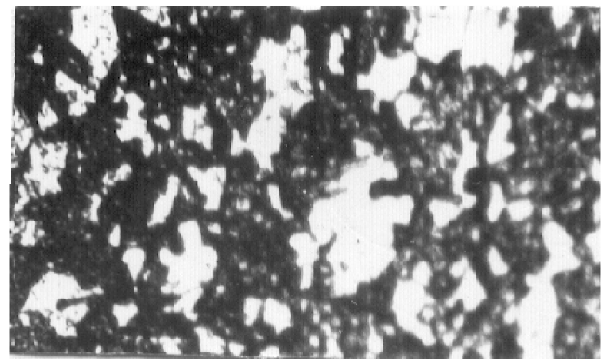

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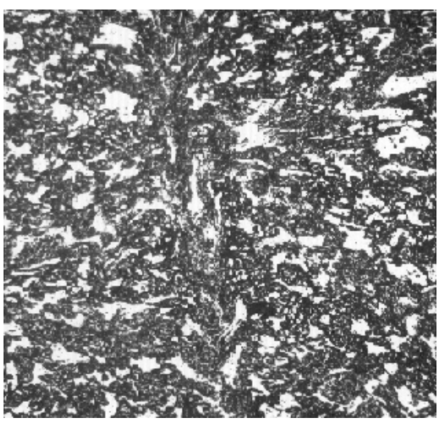
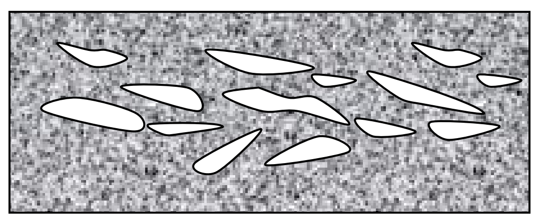
Fig. 2. Microstructure of unirradiated (a) and irradiated (b) EP-450 steel (x 200). Dark areas are sorbite grains; light areas are ferrite grains.





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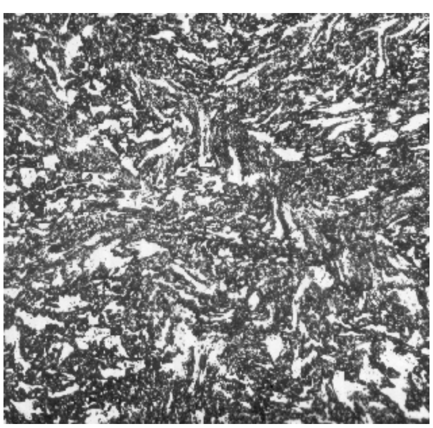

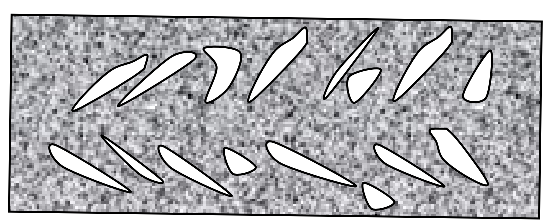

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Fig. 3. Microstructure and sketch of the organized grain structure of irradiated EP-450 steel (x 200).

Studying the microstructure of the irradiated steel with high magnification showed the presence of occasional cementite macro-chains which were located inside both ferrite and sorbite grains and sometimes along grain boundaries (Fig. 4).

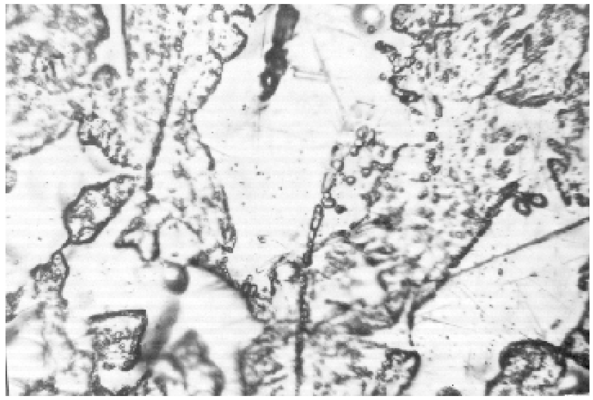

Fig. 4. Chains of cementite phase observed in the irradiated steel (x 1200).

Microhardness measurements Hμ of the steel specimens showed that before the irradiation the total microhardness Hμ was equal 280 kg/mm^2 and that the microhardness of the ferrite component alone did not exceed 225 kg/mm^2. After irradiation the total (ferrite and sorbite) microhardness was 355 kg/mm^2, while the hardness of the ferrite alone was 365 kg/mm^2.

The effect of radiation hardening of the two phases was estimated using the following relationships. Hardening of ferrite = 225 ΔH ferrite = 365-225 x100% ≈ 60%. Hardness of sorbite was determined via 2H total = H ferrite +H sorbite.

For unirradiated steel, 2 x 285 = 225 + H sorbite, hence H sorbite ≈ 340 kg/mm^2. At the same time for the irradiated steel it follows: 2 x 355 = 365 + H sorbite, or H sorbite ≈ 340 kg/mm^2. Thus, the hardness of the sorbite component of the steel did not change due to irradiation, but the ferrite grains got 60% harder.

Some specific features of the microhardness experiments draw special attention. Note in the irradiated specimens the prints of the diamond pyramid often have irregular shapes, with both increases and decreases of square diagonals observed compared to the "normal" length (Fig. 5). No similar behavior was observed in the unirradiated steel, with the prints in sorbite and ferrite components having a regular square shape (Fig. 6).

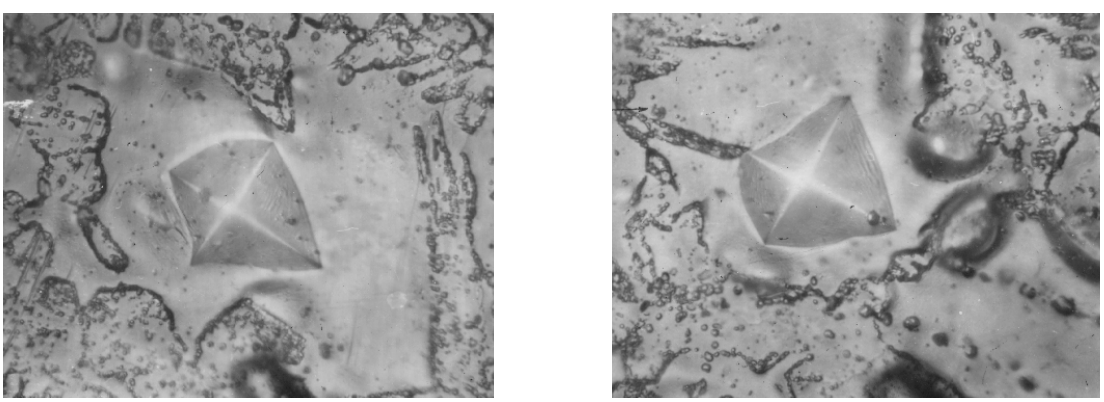

Fig. 5. Distorted imprints of the diamond pyramid in neutron irradiated EP-450 steel (x 2000).

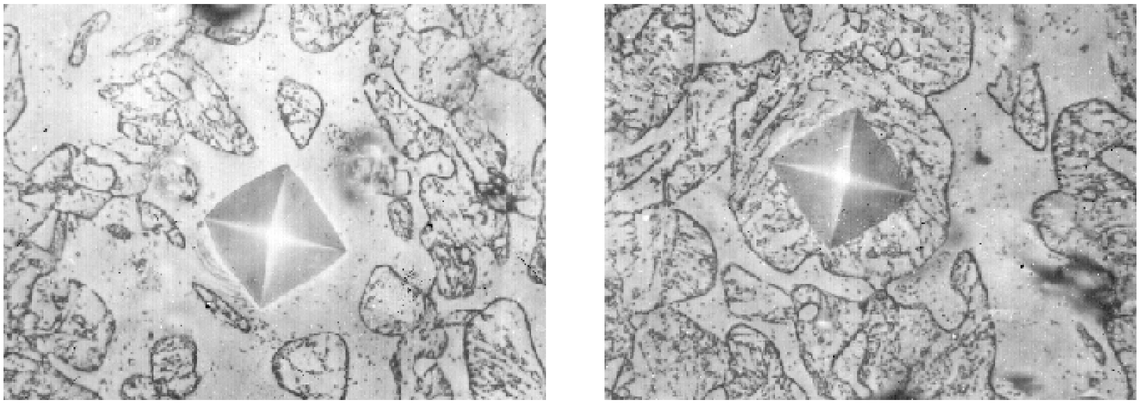

Fig. 6. Imprints of the diamond pyramid in ferrite (a) and sorbite (b) of unirradiated EP-450 steel (x 2000).

There is considerable difference observed between density change data and the microscopy data. Hydrostatic measurement showed a density decrease with respect to the unirradiated steel of only 0.04 %, and microscopy showed the presence of small voids, the concentration and size distribution of which imply ~ 0.4% swelling (Fig. 7). This implies that there is a densification of ~ 0.4% that obscures the onset of swelling as might be observed via density measurements.

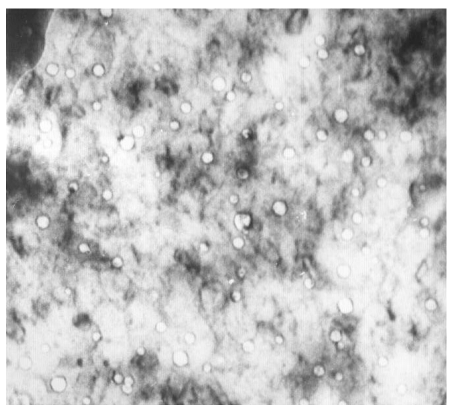

Fig. 7. Voids observed in irradiated EP-450 stainless steel after irradiation in BN-350 to 40.3 dpa at 300ºC.

Results

It is obvious from these results that EP-450 under neutron irradiation can swell at ~ 300ºC, but depending on one's point of view, the amount of swelling is either rather small or significantly larger than expected. Swelling of 0.4% at 40.3 dpa and a low temperature of only 300ºC is relatively larger than usually expected and may be a reflection of unanticipated swelling at low-temperature and low-flux predicted by Garner and coworkers for ferritic and ferritic/martensitic steels (1-4). This steel has recently been shown to swell at temperatures as low as 275ºC in BN-350 (3). At 305ºC and 33.5 dpa in another BN-350 wrapper swelling of 0.015% was observed, indicating an average swelling rate of only 4 x 10^{-4} %/dpa. (3).

As shown in Fig. 8, swelling data on EP-450 was compiled by Dvoriashin and coworkers as an average swelling rate (swelling divided by dpa) over the range of temperatures experienced in the BR-10, BN-350, and BN-600 fast reactors (3). Note that the average swelling (1 x 10^{-2} %/dpa) observed at 300ºC and 40.3 dpa in this study falls at the high end of the swelling rates observed by Dvoriashin, indicating that the swelling rate at 300ºC is increasing between 33.5 and 40.3 dpa.

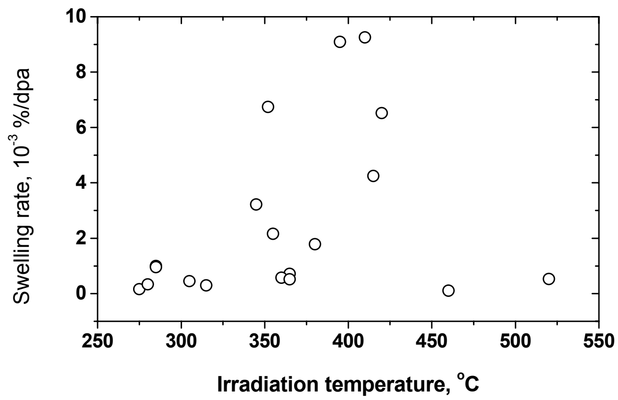

Fig. 8. Temperature range of void swelling observed in EP-450 irradiated in three fast reactors, with swelling expressed as an average swelling rate (3).

A more significant observation is that the sorbite component of this two-phase steel is not stable under irradiation, with the sorbite being progressively replaced by ferrite. The distortion of the hardness imprints observed in ferrite grains following irradiation is another indication of the continuing instability, probably arising from gradients in carbon content as the adjacent sorbite grains continue to dissolve. It is instructive to note that the aggregate hardening due to irradiation arises from increases in both the ferrite content and its hardness, and not from any change in the sorbite intrinsic hardness. Some changes in density observed as a consequence of radiation are not yet understood, but may reflect some consequence of the evolving phase changes.

The changes in phase stability observed in this study have been observed in other studies of EP-450. The proportion of ferrite and sorbite volume fractions at 1:1.6 obtained by the thermal treatment: quenching from 1100°C and tempering at 720°C for 1 h is similar to that obtained in this study at 1: 1.55 (5). Precipitation chains analogous to those in the current study (see Fig. 4) were also seen for EP-450 steel irradiated in the BN-600 fast reactor (6).

Therefore it can be assumed that the instability of EP-450 is a relatively reproducible process, and the mechanisms that drive this behavior are also occurring in Western steels when irradiated under similar conditions.

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