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Swelling and void-induced embrittlement of austenitic stainless steel irradiated to 73–82 dpa at 335–365°C

S.I. Porollo [a], A.N. Vorobjev [a], Yu.V. Konobeev [a], A.M. Dvoriashin [a], V.M. Krigan [a], N.I. Budylkin [b], E.G. Mironova [b], F.A. Garner [c,*]

[a] Institute of Physics and Power Engineering, Obninsk, Kaluga Region, Russian Federation

[b] Bochvar's Research Institute of Nonorganic Materials, Moscow, Russian Federation

[c] Pacific Northwest National Laboratory, Structural Materials Research, P.O.Box 999, Richland WA 99352, USA

Abstract

Conventional wisdom holds that void swelling of stainless steels falls strongly in swelling rate at temperatures below 400°C and should not reach large swelling levels even at high dpa levels. This perception may be incorrect as demonstrated by an irradiation of the Russian steel EI-847 which, in both the annealed and cold worked conditions, reached swelling levels in the BN-350 fast reactor on the order of $\geqslant 10\%$ at temperatures of 335–365°C and doses of 73–82 dpa. Consistent with expectations based on earlier studies of other steels at these swelling levels, the steel was found to be exceptionally brittle and to have failed with a rather unusual failure mode.

1. Introduction

It is generally accepted that void swelling of austenitic stainless steels ceases below some temperature in the range 340–360°C, and exhibits relatively low swelling rates up to ~420°C. It is also generally understood that the swelling rates remain low in the temperature range 340–420°C, and do not increase toward the 1%/dpa post-transient swelling behavior observed above 420°C [1]. These perceptions may not be correct at all irradiation conditions, however, since they were largely developed from data obtained in fast reactors such as EBR-II and FFTF whose inlet temperatures were in the range of 365–370°C. Thus the strong gradient in neutron flux at the bottom of these cores did not allow large dose levels to be reached at ~365°C, and no data are available below this temperature.

Garner and coworkers have recently called these perceptions into question, however, especially for lower flux environments found in light water power reactors, accelerator-driven spallation neutron devices, and some fusion applications [2–5]. They cite the well-known "temperature shift" phenomenon and the higher levels of helium and hydrogen generation in such environments, and then draw the conclusion that the swelling regime can move significantly downward in temperature under these conditions.

Garner and coworkers also noted that if $\geqslant 10\%$ void swelling is accumulated at temperatures of $\leqslant 400°C$, stainless steels were subject to a new form of severe embrittlement arising from the combined effects of stress concentration between voids, nickel segregation at void surfaces, and the resultant tendency toward martensite formation when the steel was deformed at room temperature. When deformed at the irradiation temperature the embrittlement was much less severe.

Voids have now been observed at lower-than-expected temperatures and relatively low dose levels under low dose rate conditions, supporting the possibility that a "temperature shift" does indeed operate [3,4]. Until recently, however, no data has been available to support the possibility of large levels of void swelling at high dpa levels for temperatures well below 400°C. This paper provides the first evidence to support the possibility of high swelling and resultant embrittlement. Some partial and preliminary results of this work were published earlier [4,5].

* Corresponding author. Tel.: +1 509 3764136; fax: +1 509 3760418; e-mail: frank.garner@pnl.gov.

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2. Experimental details

The steel used in this experiment was EI-847, a Russian niobium-stabilized 16Cr–15Ni stainless steel, and was irradiated in both the solution annealed and 20% cold-worked condition. Compositions and treatments of these two variants are shown in Table 1.

The specimens were in the form of relatively long, argon-pressurized, thin wall creep tubes, with hoop stress levels ranging from 0 to 196 MPa. These tubes have been used in other experiments, and the dimensions of these tubes are shown in a companion paper in these proceedings [6].

The tubes were irradiated in the BN-350 fast reactor located at Aktau, Kazakhstan in a special test subassembly. This subassembly is similar to fuel driver assemblies used in BN-350, but the central fuel pins were replaced by an extractable container 30 mm in diameter. Cylindrical baskets of 27 mm diameter containing seven creep tubes each were placed in the extractable container. All surfaces of the baskets had perforations to allow the reactor sodium coolant to flow over the tubes. The inlet coolant temperature for this reactor is ~285°C.

The test subassembly was irradiated for 14,400 h over the period 12/3/86 to 11/10/89 in the low-enrichment zone of BN-350. The irradiation was conducted in two segments with a transfer into a second subassembly on 4/3/88. The temperatures of the tubes were determined by calculations involving the known coolant flow and the heat production of surrounding fuel pins, and are thought to be accurate to ±10°C over the lifetime of the experiment.

Two groups of specimens were irradiated. The first group was the annealed tubes which had average temperatures from top to bottom of 324–350°C and dose levels of 65–78 dpa, respectively. The second group was 20% CW tubes ranging from 350–385°C and 78 to 95 dpa. At the center of these tubes the conditions were 73 dpa, 335°C and 82 dpa, 365°C for the annealed and cold-worked tubes respectively. These conditions will be the ones quoted in the following sections.

Table 1
Composition of EI-847 steels in wt%

EI-847	C	Si	Mn	S	P	Cr
S.T. [a]	0.050	0.29	0.78	0.009	0.011	15.74
C.W. [b]	0.060	0.20	0.73	0.004	0.012	15.61

	Ni	Mo	Nb	B	N
S.T.	15.32	2.95	0.54	0.001	0.035
C.W.	14.90	2.96	0.59	0.001	0.022

[a] S.T. Solution treated 1040°C/3 min, grain size 12 μm.
[b] C.W. Solution treated 1040°C/3 min, 18 ± 2% C.W., grain size 10–15 μm.

3. Results

Upon disassembly of the baskets all tubes, except those in the stress-free condition, were found to have lost pressure, with most having failed in a very brittle manner without any obvious physical insult having been experienced during disassembly. Some tubes were reduced to fragments. Many of the tubes were also bent toward one or both ends, having elongated enough to interact with the ends of the basket. Such elongation is known to arise only from swelling and not irradiation creep. Both failure and bending can be seen in Fig. 1.

Although it was difficult to measure diameter changes and density on such broken tubes, it became obvious, where measurements could be made, that diameter changes as large as 5.4% on the annealed tubes and 7.6% on the cold-worked tubes had occurred, indi-

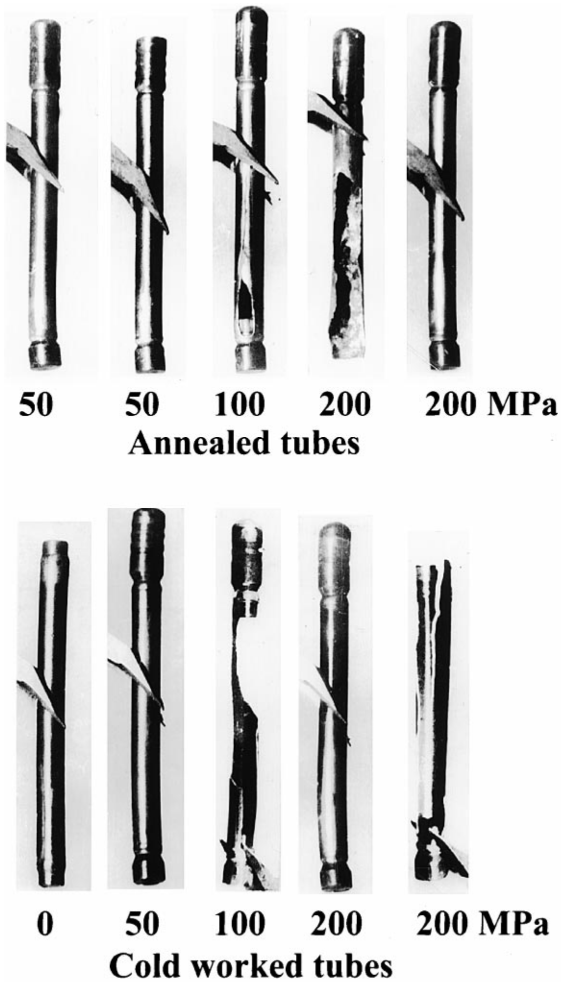

Fig. 1. Post-irradiation condition of EI-847 tubes at 335°C (top) and 365°C (bottom), showing swelling-induced fracture and bending.

cating that substantial combined creep and swelling strains had occurred. On some of the cold-worked tubes at 365°C and 82 dpa, three density change measurements were obtained ranging from 10.3% to 15.5%, supported by microscopy measurements of 11.7–14.5% swelling.

Examination of these specimens using transmission electron microscopy is still in progress, but it is very clear that significant void swelling has occurred in these specimens. Fig. 2 shows ~6.2% swelling in the stress-free annealed tube at 335°C and 73 dpa, while Fig. 3 shows ~11.7% swelling in the stress-free cold-worked tube at 365°C and 82 dpa. In earlier papers [4,5], void microstructures producing ~14% swelling were shown for the cold-worked tube at 100 MPa hoop stress. Fig. 4 shows that large voids form parallel to the grain boundary

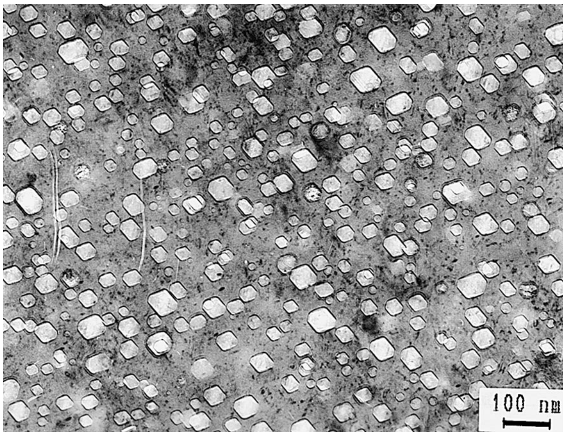

Fig. 2. Microstructure of annealed EI-847 after irradiation at 335°C, 73 dpa and zero stress. The voids have a mean diameter of 260 Å, number density of 4.0×10^{15} cm^{-3}, with 6.2% swelling.

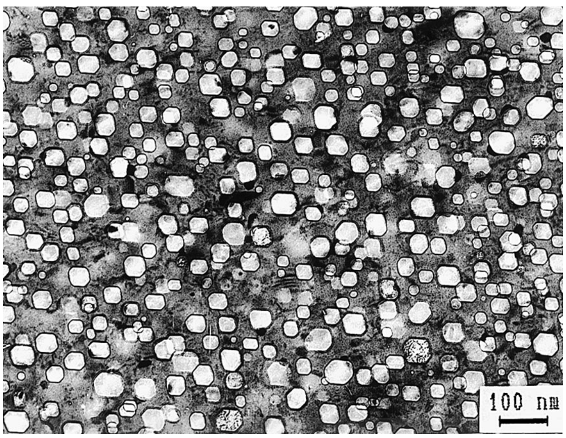

Fig. 3. Microstructure of cold-worked EI-847 after irradiation at 365°C, 82 dpa and zero stress. The voids have a mean diameter of 312 Å, number density of 4.8×10^{15} cm^{-3}, with 11.7% swelling.

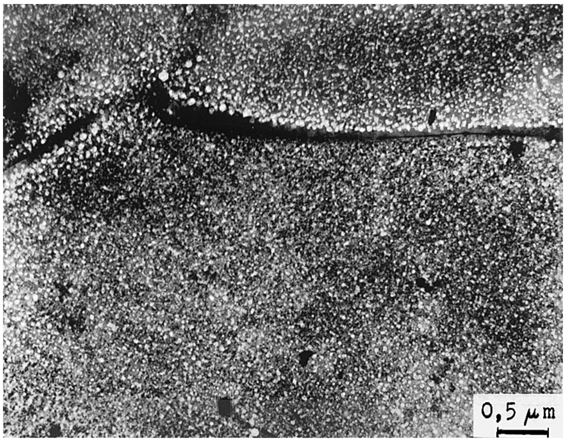

Fig. 4. Large voids associated with cracking along denuded zone adjacent to grain boundaries at 335°C and 73 dpa in an annealed stress-free tube.

denuded zone. Such zones frequently develop cracks parallel to the grain boundaries along and through these larger void layers during specimen preparation. Alpha ferrite is also observed on these fracture surfaces. When broken easily with tweezers, new fracture surfaces are highly irregular, and appear largely to be traveling along grain boundaries, as shown in Fig. 5. Upon closer examination it appears that the fracture surface jumps back and forth across the grain boundary, with fracture arising from ductile coalescence of the large voids in the vicinity of the grain boundary.

4. Discussion

Contrary to conventional wisdom, the large levels of void swelling produced by this irradiation imply that the void swelling rate must be increasing at 335–365°C at the higher dose levels reached in this experiment. It should be noted that these unexpectedly large levels of void swelling were developed at displacement rates typical of fast reactors and therefore are not a consequence of the "temperature shift" phenomenon arising from changes in displacement rate. At lower dpa rates, more swelling might therefore occur.

Since void-related embrittlement is a well-known phenomenon at temperatures below 420°C [1,7], the resultant embrittlement is not so unexpected, however. What is found to be unusual is that the fracture surface travels along, or most likely, parallel to grain boundaries, involving ductile tearing of the larger voids that almost always form the border of the denuded zone. Exactly the same behavior was observed by Anderson, Garner and Stubbins, however, in highly voided (18%) pure copper [8]. In the steel, however, the material on the fracture surface appears to have undergone a transition to martensite.

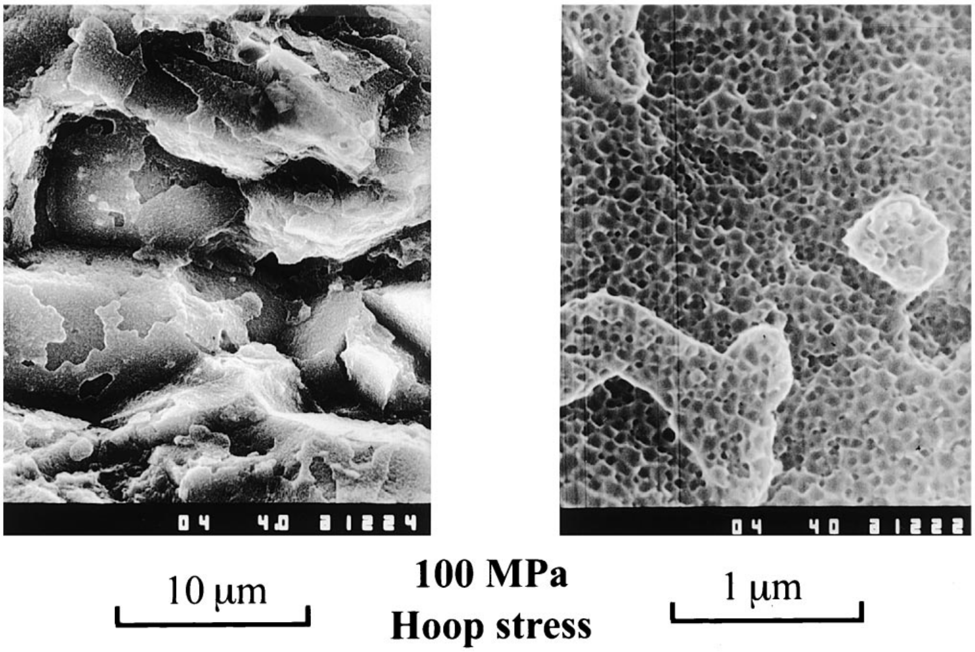

Fig. 5. Fracture surface of failed cold-worked creep tube at 365°C and 83 dpa, showing ductile coalescence of voids parallel to grain boundaries.

Since dpa levels of 73–83 dpa have not been previously reached at temperatures in the 335–365°C range, there is no way to know if the response of this particular steel is typical and therefore to be expected in all steels, or whether this steel is somehow more susceptible to high swelling levels. This steel has served well, however, in other reactor applications in Russia. In addition, previous experience with void swelling of stainless steels leads us to the conclusion that if one steel can swell in this manner under these conditions, all stainless steels are capable of reaching this state, with only the incubation time for swelling altered by compositional and/or thermomechanical variations.

One last consideration should be mentioned. The fact that the stress-free tubes did not break into fragments while the stressed tubes did, may imply that slow strain rates typical of irradiation creep may have contributed to the failure mechanism during the experiment. If correct, this would be very unusual, since irradiation creep in general is thought not to directly cause failure [1]. It is expected, however, that even the unbroken stress-free tubes will be found to be very fragile with respect to post-irradiation handling.

5. Conclusions

At temperatures of 335–365°C and doses in excess of 70–80 dpa it appears possible to reach rather high ($\geqslant 10\%$) swelling levels in at least one stainless steel when irradiated at displacement rates typical of fast reactors. Associated with such high swelling levels is a pronounced embrittlement.

Acknowledgements

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