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Journal of Nuclear Materials 251 (1997) 252–261

journal of nuclear materials

Irradiation creep and void swelling of austenitic stainless steels at low displacement rates in light water energy systems

F.A. Garner *, M.B. Toloczko

Pacific Northwest National Laboratory, Structural Materials Research, PO Box 999, Richland, WA 99352, USA

Abstract

The majority of the high fluence data on the void swelling and irradiation creep of austenitic steels was generated at relatively high displacement rates characteristic of fast reactors. The application of these data to other reactor systems which operate at lower displacement rates requires that some estimate be made of the dependence of swelling and irradiation creep on displacement rate. Austenitic components in typical light water reactors (LWRs) experience displacement rates that are an order of magnitude or more lower than that found in fast reactors. Since irradiation creep has a component that is proportional to swelling, it is anticipated that irradiation creep will exhibit a sensitivity to displacement rate that is a direct expression of the dependence of swelling on displacement rate. Until recently, however, the non-swelling-related component of creep was also thought to exhibit its own dependence on displacement rate, increasing at lower fluxes. This perception originally arose from the work of Lewthwaite and Mosedale on irradiation creep of cold-worked steels in the Dounreay Fast Reactor at temperatures in the 270–350°C range. It now appears that this interpretation is incorrect and, in fact, the steady-state value of the non-swelling component of irradiation creep is relatively insensitive to displacement rate. The earlier perceived flux dependence appears to have arisen from a failure to properly separate the transient and steady-state regimes of irradiation creep. As evidence begins to accumulate on void swelling of stainless steels at LWR-relevant displacement rates, voids are being observed at lower-than-expected temperatures and often at very low dpa levels, implying that the 'temperature shift' phenomenon may indeed be operating to increase swelling. It is demonstrated in this paper that if swelling exceeds ~ 10% at PWR-relevant temperatures, stainless steels become very brittle.

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1. Introduction

The majority of high fluence data on the void swelling and irradiation creep of austenitic stainless steels was generated at relatively high displacement rates found in fast reactors and some high flux test reactors. The application of these data to light water reactors (LWRs) requires that some estimate be made of the dependence of swelling and irradiation creep on displacement rate.

Whereas swelling has not previously been seriously addressed as a potential problem in light water reactors (LWRs), it now appears that it can occur in some LWR components under the right conditions. Contributing factors to produce these conditions are relatively high dpa levels, gamma heating of relatively thick components, the 'temperature shift' phenomenon, and increased levels of helium and hydrogen generation characteristic of LWR neutron spectra [1].

Garner and co-workers [1,2] recently predicted that components of the baffle assembly in some pressurized water reactors (PWRs) might meet these conditions. Several important conclusions of this work were: (1) based on fast reactor data alone, swelling in excess of 10% was predicted to occur late in life in some portions of the annealed 304L steel used in the baffle plates; (2) due to the differences in neutron spectrum and displacement rate between fast reactors and PWRs, a 'temperature shift' in the swelling regime would probably lead to more swelling per dpa in the baffle plate than would be predicted from high-flux fast reactor data; and (3) swelling levels > 10% would produce a very brittle steel that would be vulnerable

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* Corresponding author. Tel.: + 1-509 376 4136; fax: + 1-509 376 0418; e-mail: fa_garner@ccmail.pnl.gov.

0022-3115/97/$17.00
PII S0022-3115(97)00260-2

not only to seismic and thermal shocks, but also to slow rates of strain characteristic of irradiation creep.

Irradiation creep and void swelling are known to be intimately connected phenomena [3] and therefore the dependence of irradiation creep on displacement rate must also be examined to determine whether irradiation creep is also subject to a 'temperature shift' of some sort. This paper reexamines the validity of the previously accepted flux sensitivity of irradiation creep.

Table 1
Compositions of the alloys studied by Lewthwaite and Mosedale

Alloy	Fe	Cr	Ni	Mo	Mn	C	Si	Ti	Nb
316 1	Bal	16.7	13.7	2.3	1.9	0.04	0.6	< 0.02	< 0.05
316 2	Bal	17.4	13.8	2.4	1.8	0.04	0.6	–	–
316 3	Bal	16.8	13.3	2.3	1.7	0.04	0.4	–	–
FV548	Bal	16.5	11.8	1.4	1.1	0.11	0.7	–	0.9
EN58J	Bal	18	9	< 0.5	< 1	0.06	–	0.5	–
EN58B	Bal	17.6	11	< 0.5	< 1	0.03	–	–	–
EN58E	Bal	17	9	2.8	< 1	0.08	–	–	–

2. The dependence of irradiation creep on displacement rate

As reviewed by Garner [3], irradiation creep exhibits a short and often ignored transient regime, followed by a steady-state creep rate B, per MPa and dpa, that is composed of two contributions which can be written as

$$B = \frac{\bar{\dot{\varepsilon}}}{\bar{\sigma}} = B_o + D\dot{S}, \tag{1}$$

where $\bar{\dot{\varepsilon}}/\bar{\sigma}$ is the effective strain rate per MPa and dpa, B_0 is the creep compliance, a swelling-independent contribution. The parameter D is the creep-swelling coupling coefficient, and $\dot{S}$ is the instantaneous volumetric swelling rate.

Garner [3] earlier reviewed the available data and reached the conclusion that there appeared to be a neutron flux-dependence of both contributions. The data showed that the flux-dependence of the second term arose from the inherent flux-dependence of $\dot{S}$ via the 'temperature-shift' at lower irradiation temperatures. The temperature regime of swelling is dependent on the displacement rate and 'shifts' in temperature as the displacement rate changes. The shift at lower irradiation temperatures is thought to arise from the dependence of Frenkel pair recombination on displacement rate, and causes an increase in swelling at PWR-relevant temperatures as the dose rate decreases. The coefficient D in the $D\dot{S}$ contribution was shown not to be constant as originally thought, but to depend either on the total swelling or the instantaneous swelling rate.

With respect to the flux-dependence of B_0, the data were not so conclusive, however, with some data supporting a flux-dependence and some not. Garner concluded

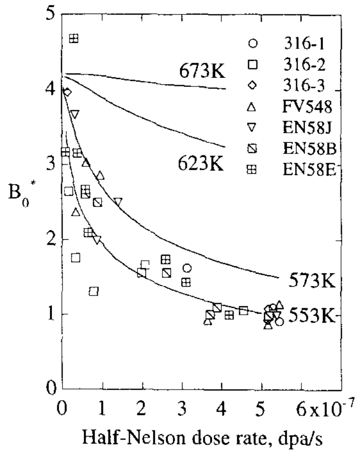

Fig. 1. Apparent flux-dependence of the B_0^* component of the irradiation creep rate, as presented by Lewthwaite and Mosedale [4], for a variety of cold-worked austenitic stainless steels irradiated in and below the DFR core at 270–305°C (543–578 K). B_0^* is the creep rate normalized by the average creep rate of specimens of the same alloy type irradiated at dose rates greater than 5×10^{-7} dpa/s. 100 Half-Nelson dpa ≅ 85 NRT dpa. The trend lines indicate the expected behavior as a function of irradiation temperature if Frenkel pair recombination dominates at lower irradiation temperatures.

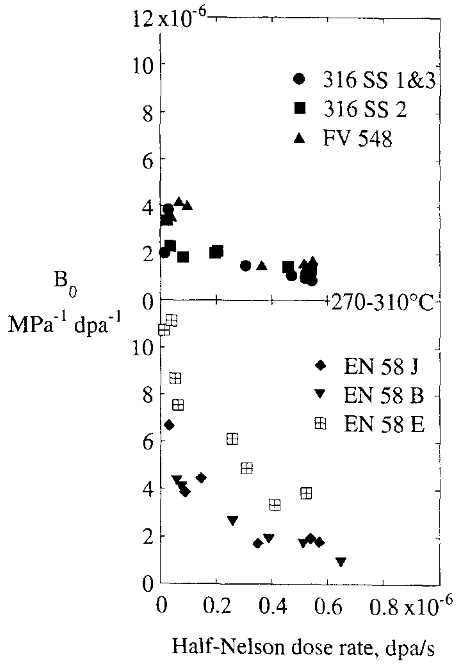

Fig. 2. Creep rate, B_0, calculated from the data used to construct Fig. 1. Dose is defined by the Half-Nelson ($N/2$) model used by Lewthwaite and Mosedale [4], assuming that no swelling is occurring.

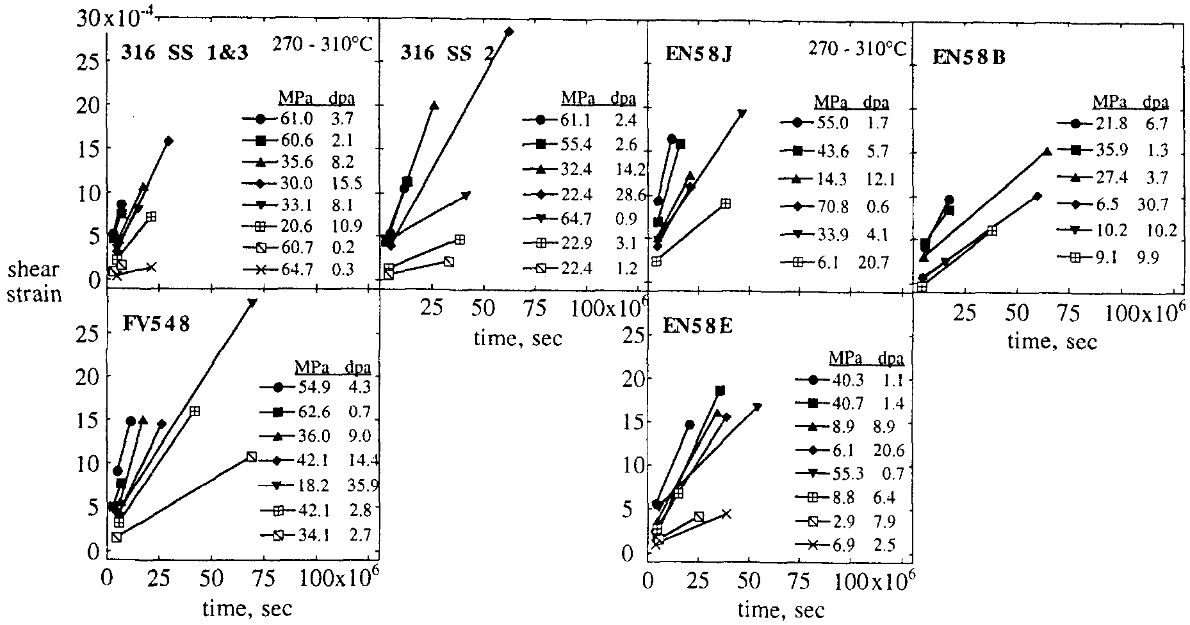

Fig. 3. Original strain data from Lewthwaite and Mosedale [4] used to derive the creep coefficients plotted in Fig. 2. Intermediate points on each line which were shown in Lewthwaite and Mosedale's original plots, have been omitted here for clarity. Dose is defined by the Half-Nelson ($N/2$) dpa model.

that the apparently best-defined data supported an inverse square root dependence of B_0 at temperatures below 350°C, with the B_0 creep contribution increasing as the displacement rate decreases. Based on this perceived dependence, creep data derived from fast reactors would significantly underpredict the creep strain at the neutron flux levels characteristic of LWRs.

Based on a later analysis of data from other as-yet unpublished sources that did not support this conclusion, the flux dependence of B_0 has been revisited with more detailed attention paid to the data sets which appeared to support a flux dependence of B_0. As shown below, it now appears that these data were misinterpreted by the original authors, and B_0 is essentially independent of displacement rate.

Fig. 1 shows that Lewthwaite and Mosedale [4] saw an apparent inverse square root flux-dependence of irradiation creep at low temperatures in a variety of cold-worked

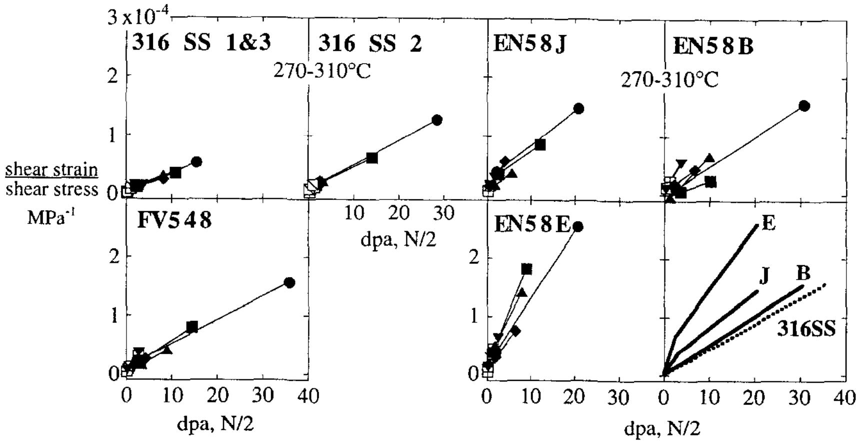

Fig. 4. Stress-normalized data from Fig. 3, plotted versus dpa, showing that higher creep rates occurred only in the low-dose transient regime of creep. Again, intermediate points on the lines have been omitted for clarity.

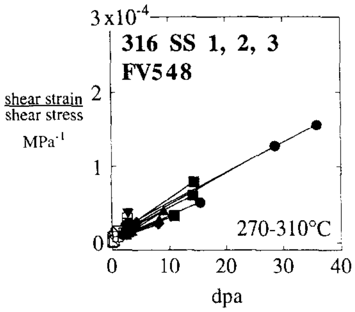

Fig. 5. Superposition of creep data from FV548 and two 316 alloys.

stainless steels (Table 1) irradiated as springs in and below the core of the Dounreay Fast Reactor (DFR). These data were presented in reduced and normalized form, and unfortunately produced a very misleading impression, implying that B_0 is flux-sensitive. If the unnormalized creep coefficients are broken into subsets such as presented in Fig. 2, it is obvious that FV548 and 316-type steels exhibit much less of an apparent flux-dependence than do the EN58 variants. The shear strain data cover a wide range of (stress, dpa, dpa rate) combinations, and when presented by Lewthwaite and Mosedale versus time, as shown in Fig. 3, do not allow an easy visualization of a flux dependence. When plotted as stress-normalized strain versus dpa, however, as shown in Fig. 4, it becomes clear that the larger creep rates occurred only at the very lowest dpa levels. Therefore these higher creep rates represent only the low-dpa transient regime of creep and are not a direct consequence of the lower-flux level at which these low-dpa levels were attained. As shown in Fig. 5, the FV548 and 316 variants can almost be considered to behave as a single alloy.

As shown on the lower right-hand-side of Fig. 4, the

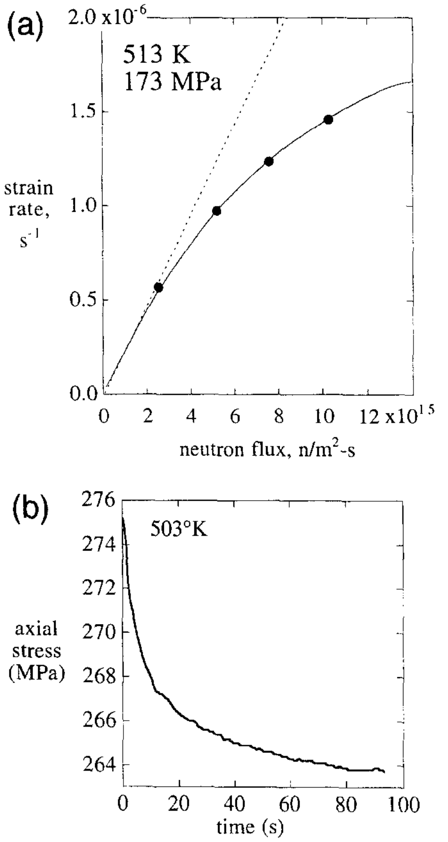

Fig. 7. (a) Irradiation creep observed by Barmore and coworkers in annealed polycrystalline nickel minitensile specimens irradiated in a stress relaxation test at 240°C in RTNS-II with 14 MeV neutrons [6], suggesting an inverse square root dependence on neutron flux. (b) Typical stress relaxation curve observed by Barmore and coworkers for annealed polycrystalline nickel under steady 14 MeV neutron irradiation at 9×10^{15} n/m^{2}s^6, showing that most relaxation during this test is a consequence of the transient regime of irradiation creep.

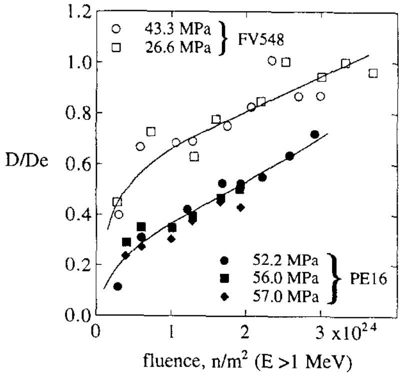

Fig. 6. Irradiation creep of cold-worked FV548 and Nimonic PE16 springs in the DMTR reactor at 100°C, as observed by Lewthwaite and Proctor [5]. Note that the post-transient creep rates are essentially the same.

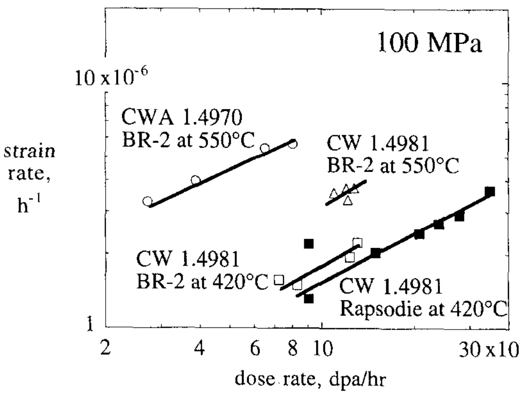

Fig. 8. Dependence of irradiation creep on displacement rate of 1.4970 and 1.4981 steels as presented by Grossbeck et al. [7].

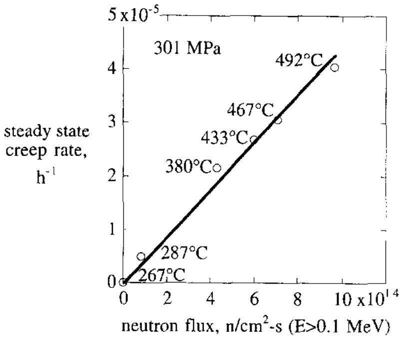

Fig. 9. Secondary creep rates observed in annealed 09Kh16N15M3B irradiated in the BR-10 fast reactor by Kruglov and co-workers [8].

largest transient regimes are associated with two of the three EN58 variants. There is significant scatter in the low dpa EN58B creep behavior, but its behavior at high dpa is very similar to that of 316 and FV548. The EN58E alloy is a high molybdenum alloy compared to the other two EN58 variants. Its larger post-transient creep rate suggests that this alloy may already be swelling, yielding a $D\dot{S}$ contribution, while the low molybdenum variants may not yet be swelling. Based on the lower nickel content of this alloy and its lack of silicon, it would be expected to swell sooner than would the 316 alloys [3].

Interestingly, Lewthwaite and Proctor recognized the strong role of the transient regime in earlier work conducted in the DMTR reactor at 100°C, as shown in Fig. 6. Note the relative difference in transient magnitude of the two steels, as well as the nearly identical post-transient creep rates.

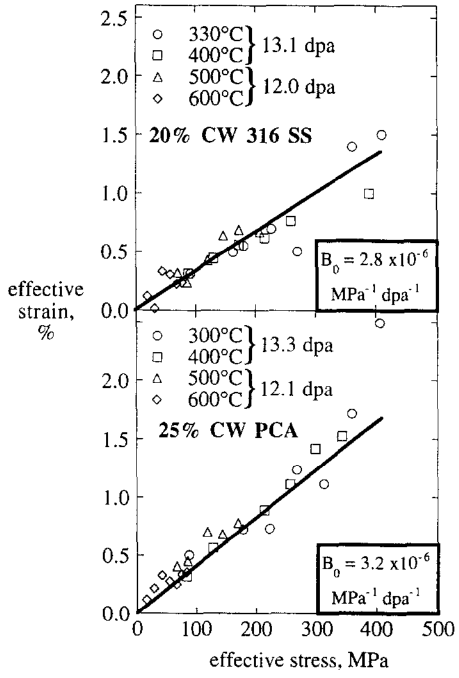

Fig. 10. Temperature-independent creep strain observed by Grossbeck and Horak in 20% cold-worked 316 and 25% cold-worked PCA during irradiation in ORR [9].

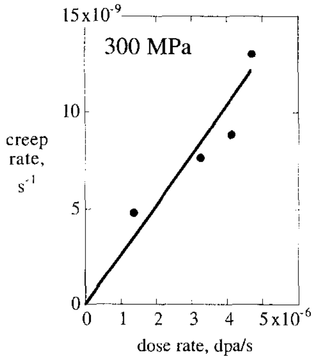

Fig. 11. Effect of displacement rate on irradiation creep of 20% cold-worked 316 SS at 300°C as observed by Schwaiger and co-workers using deutreron irradiation [10].

Having realized that the data of Lewthwaite and Mosedale had been misinterpreted, the other data cited in Ref. [3] showing a flux sensitivity were also revisited. Fig. 7a shows the reduced data of Barmore and co-workers [6] for irradiation creep of annealed polycrystalline nickel derived from a stress relaxation experiment using 14 MeV neutrons in the RTNS-II facility. Fig. 7b shows a typical high-flux relaxation curve, demonstrating that most of relaxation arises from transient creep. All tests at lower fluxes reached correspondingly lower neutron fluences. Therefore, the creep rates at lower fluxes were even more

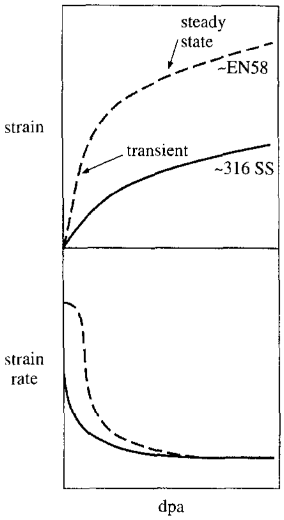

Fig. 12. Schematic illustration of the impact of the transient regime on the interpretation of irradiation creep data. Creep rates at lower dpa levels are strongly dominated by the transient regime of creep.

Table 2
Voids observed in annealed 16Cr-15Ni-3Mo-Nb austenitic stainless steel, EI-847, irradiated in a defense PWR

Fluence (10^{21} n/cm^2) ($E > 0.5$ MeV)	T_{irr} (°C)	Dose rate (NRT) (10^{-8} dpa/s)	Dose (NRT) (dpa)	Void data		
				mean void diameter (Å)	void number density (10^{14} cm^{-3})	swelling from TEM (%)
2.2(1)	330[a]	8.9	2.1	65	23	0.033
4.6(2)	325	10.5	4.4	50	20	0.013
6.3(3)	316	7.3	6.1	92	5	0.019
4.9(3)	314	5.6	4.7	112	5	0.035
2.2(3)	310	2.5	2.1	91	3	0.073
2.4(3)	295	2.8	2.3	n.o.[b]	n.o.	n.o.

(1–3) Denotes different fuel pins.

[a] Temperature measured using chromel–alumel thermocouple on inside of fuel clad.

[b] n.o.: not observed.

Table 3
Voids observed in annealed 16Cr–15Ni–3Mo–Nb–B[a] austenitic stainless steel, EP-172, irradiated below core in the BOR-60 fast reactor

Fluence (10^{21} n/cm^2) ($E > 0.1$ MeV)	T_{irr} (°C)	Dose rate (NRT) (10^{-8} dpa/s)	Dose (NRT) (dpa)	Void data		
				mean void diameter (Å)	void number density (10^{14} cm^{-3})	swelling from TEM (%)
2	310	5.7	2.01	n.o.[b]	n.o.	n.o.
4	312	7.8	2.68	n.o.	n.o.	n.o.
6	314	12.6	4.29	25	0.15	$\leq 10^{-4}$
11	315	14.9	5.09	27	1.1	0.001
18	320	22.8	7.77	45	3.3	0.015
40	340	62.4	21.3	89	1.2	0.044

[a] Fe–15.83Cr–15.15Ni–2.90Mo–0.84Nb–0.39Mn–0.12Si–0.08C–0.01B–0.012S–0.009P–0.02N_2.

[b] n.o.: not observed.

Table 4
Voids observed in annealed 18Cr–9Ni–Ti austenitic stainless steel irradiated in the reflector region of the BN-350 fast reactor

Fluence (10^{21} n/cm^2) ($E > 0.1$ MeV)	T_{irr} (°C)	Dose rate (NRT) (10^{-8} dpa/s)	Dose (NRT) (dpa)	Void data		
				mean void diameter (Å)	void number density (10^{14} cm^{-3})	swelling from TEM (%)
43	332	4.0	14.5	173	6.4	0.28
48	321	4.6	16.8	124	12	0.17
130	310	15.6	56	n.o.[a]	n.o.	n.o.
17	280	2.1	7.7	n.o.	n.o.	n.o.

[a] n.o.: not observed.

Table 5
Composition of EI-847 steels in wt%

EI-847	C	Si	Mn	S	P	Cr	Ni	Mo	Nb	B	N_2
S.T.[a]	0.050	0.29	0.78	0.09	0.011	15.74	15.32	2.95	0.54	0.001	0.035
C.W.[b]	0.060	0.20	0.73	0.004	0.012	15.61	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.

strongly dominated by the transient regime, leading to an illusory dependence of the creep rate on displacement rate.

In another published inverse square-root study by Grossbeck et al. [7], a possible inverse square-root dependence of irradiation creep on displacement rate was also presented, as shown in Fig. 8. However, the untimely

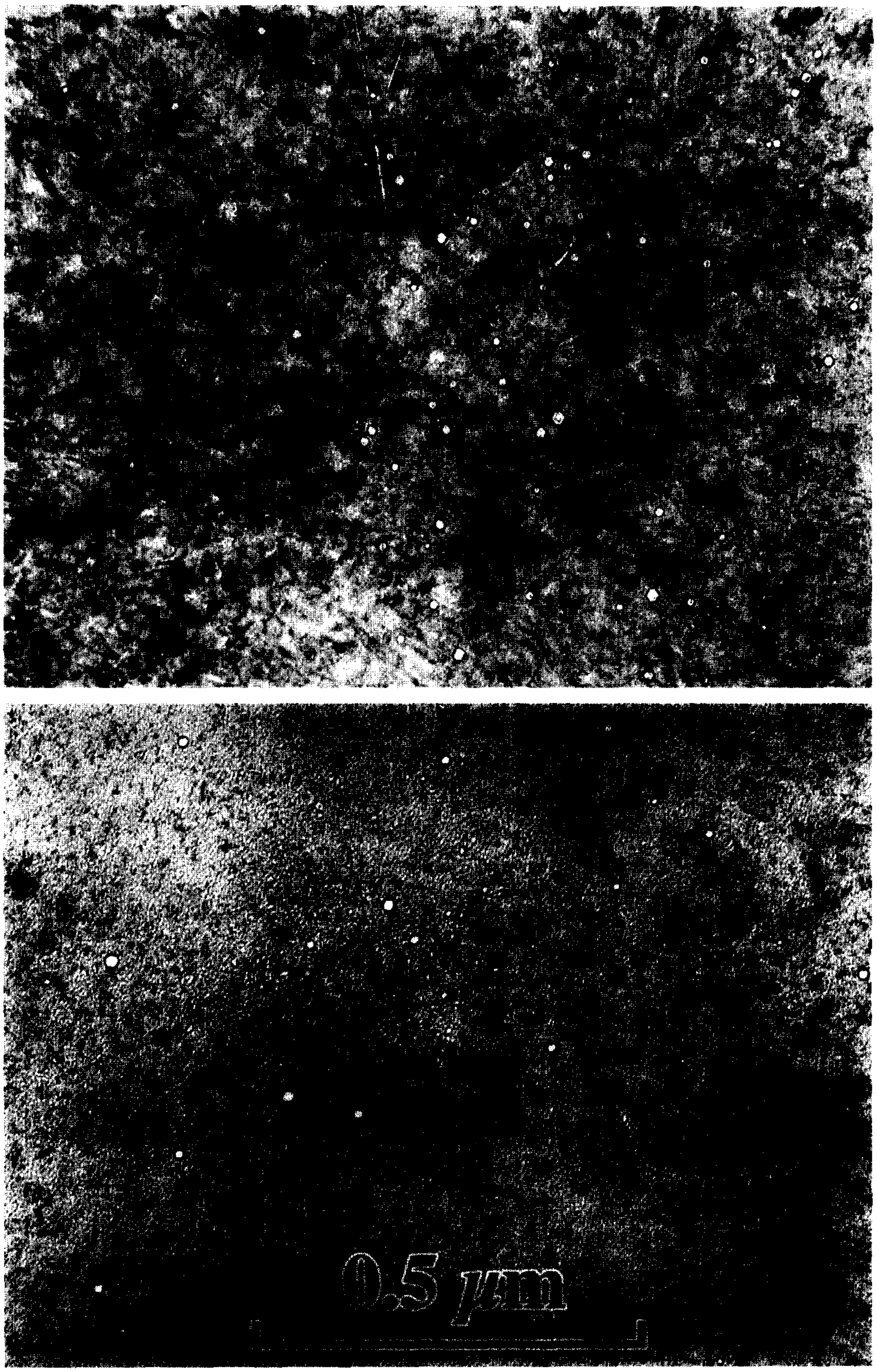

Fig. 13. Voids observed in annealed Fe–16.3Cr–15.3Ni–2.7Mo–0.4Nb–0.8Mn–0.15Si–0.02C stainless steel after irradiation at 316°C in a defense-related PWR at 4.7 and 6.1 dpa (top and bottom, respectively) [12].

death of the major German researcher, Christo Wassilew, does not allow us to re-examine this work in terms of transient influence.

The interpretation of irradiation creep data addressing the possible influence of displacement rate is often complicated by concurrent variations in neutron flux and irradiation temperature, as shown in the data of Kruglov and co-workers [8] in Fig. 9, which implies that there is no dose rate sensitivity. The temperature dependence of irradiation creep before the onset of void swelling appears to be relatively low, however, as demonstrated by Grossbeck and Horak [9] in Fig. 10. Garner reached a similar conclusion concerning the relatively small influence of temperature on the B_0 component of irradiation creep [3].

The flux-independence of irradiation creep has also been confirmed with deuteron irradiation of stainless steels [10,11]. These experiments do not suffer from dpa rate and temperature coupling effects. An example is shown in Fig. 11. Therefore, it appears that the perception that the B_0 component of irradiation creep is dependent on displacement rate is largely an artifact of incorrect data analysis. As shown schematically in Fig. 12, strain rate data plotted versus dpa (or dpa rate for a series of irradiations conducted at different flux rates over the same time interval, as in Refs. [4,6]) can give the illusion of a flux dependency, but one which arises only from the unrecognized influence of the transient regime of swelling.

3. Creep contributions arising from void swelling

If swelling is sensitive to displacement rate, the irradiation creep must also be sensitive, but when it was originally proposed that void swelling would occur at lower-than-expected temperatures in PWRs due to the dependence of swelling on both displacement rate and temperature, there were no supporting data available. A variety of both open and proprietary data are now being produced that provide confirmation of this proposal.

Based on fast reactor data, swelling is thought not to occur below 350°C [3]. The current authors, working with scientists at the Institute of Physics and Power Engineering (IPPE) in Obninsk, Russia, have recently published data showing low levels of void formation in austenitic steels at temperatures approaching 300°C and at very low doses following irradiation in low-flux conditions in both fast and thermal reactors [12]. These data are reproduced in Tables 2–4 and some examples are shown in Figs. 13 and 14. Other as-yet unpublished studies demonstrate that the irradiation creep moduli also increase under these conditions, as would be expected from a $B_0 + D\dot{S}$ creep model.

Even more importantly, the IPPE work showed that it was possible to reach swelling levels of > 10% in at least one austenitic steel designated EI-847 (see Table 5 for composition) at doses exceeding 70 dpa in the BN-350 fast reactor at temperatures in the range 335–365°C [12,13], and that the steel was very brittle under these conditions as had been earlier predicted [1]. Typical micrographs of the void microstructure and resultant fracture surface are shown in Figs. 15 and 16 for a cold-worked EI-847 pressurized tube irradiated at 100 MPa hoop stress to 82 dpa at 365°C, producing ~ 14% swelling. Similar behavior was observed in an annealed pressurized tube irradiated to 73 dpa at 335°C. In these irradiations, all tubes irradiated under stress failed, often shattering [13]. It appears that the slow

Fig. 14. Voids observed in annealed Fe–18.7Cr–10.8Ni–1.4Mn–0.5Si–0.3Ti stainless steel at 16.8 dpa and 321°C after irradiation in a low-flux reflector region of the BN-350 fast reactor [12].

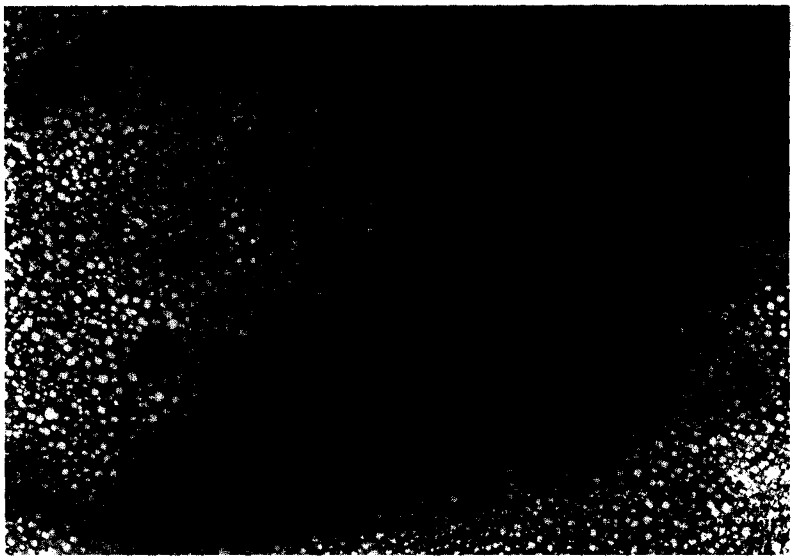

Fig. 15. Void microstructure observed in 20% cold-worked Fe–16Cr–15Ni–3Mo–Nb stainless steel after irradiation in BN-350 to 82 dpa at 365°C, and a hoop stress of 100 MPa, showing swelling of 14% [12].

strain rates inherent in irradiation creep tests contributed to the failure.

It is interesting to note that Fisher and co-workers also found that voids formed in Nimonic PE16 when irradiated at low dose rates and to very low dose levels at the ~ 320°C coolant inlet temperature in the Advanced Gas-Cooled Reactor (AGR) [14]. This high nickel alloy is expected to form very large amounts of helium and hydrogen when irradiated in the AGR neutron spectrum, and these gases may influence the swelling behavior.

The observation of such high swelling levels at 335°C implies that the swelling rate continues to accelerate at higher fluence, in contradiction to the conventional wisdom that swelling rates always remain low at such temperatures. It is not clear, however, why this Russian steel experienced such a high swelling level, and it cannot be

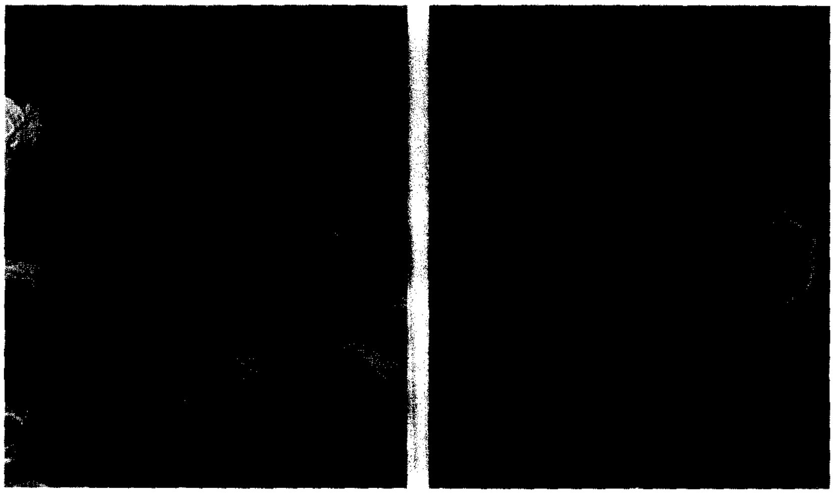

Fig. 16. Fracture surface of a failed creep tube with microstructure similar to that shown in Fig. 15 [12]. Fracture appears to have arisen from ductile coalescence of voids in the vicinity of the grain boundaries. Why the fracture surface hugs the grain boundary region is under investigation. Although the fracture surface is ductile, the high void density requires very little deformation to effect coalescence, thus leading to the observed macroscopically brittle behavior. This fracture surface was produced by manually applying pressure with two sets of tweezers.

stated at this time that typical PWR steels will exhibit comparable behavior.

Another as-yet-unresolved issue is whether the enhanced helium and hydrogen formation relative to that of fast reactor conditions during PWR irradiation will accelerate the void swelling process at relevant, i.e., lower, temperatures. If so, the lower temperatures of void formation under low-flux conditions may reflect the action of the spectral dependence of gas formation as well as that of the rate-dependent 'temperature shift'. Two upcoming papers will address this issue [15,16]. In these papers, the possibility is examined, both experimentally and theoretically, that an artificially high value of B_0 may be calculated if the researcher does not recognize that transmutation-induced gas bubbles may also contribute a $D\dot{S}$ contribution.

4. Conclusions

At temperatures and displacement rates typical of PWR components, it appears possible to develop voids in stainless steels at doses and temperatures lower than anticipated from higher-flux fast reactor data. This implies that the 'temperature-shift' concept applies to stainless steels at PWR low-flux conditions.

While the lower displacement rate of PWRs may accelerate the onset of void swelling and thereby the $D\dot{S}$ component of irradiation creep, it does not appear to affect the B_0 component of creep. This latter conclusion is a reversal of a long-standing previous perception.

Acknowledgements

This research is sponsored at PNNL by the Electric Power Research Institute, and the US DOE under the auspices of the Basic Energy Science Program. The research at the IPPE is supported by both the Civilian Research and Development Foundation and the Pacific Northwest Laboratory of Battelle.

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