Journal of Nuclear Materials 386–388 (2009) 249–253

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Anisotropic swelling observed during stress-free reirradiation of AISI 304 tubes previously irradiated under stress

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ABSTRACT

A 'history effects' experiment was conducted in EBR-II that involved the reirradiation of AISI 304 cladding and capsule tubes. It is shown that when irradiated tubes had not previously experienced stress, subsequent irradiation led to additional swelling strains that were isotropically distributed. However, when tubes previously irradiated under a 2:1 biaxial stress were reirradiated without stress the additional swelling strains were not isotropically distributed. The tubes obviously retained a memory of the previous stress state that appears to be attempting to distribute strains in the directions dictated by the previous stress state. It is clear, however, that the memory of that stress state is fading as the anisotropic dislocation microstructure developed during irradiation under stress is replaced by an isotropic dislocation microstructure during subsequent exposure in the absence of stress. It is also shown that once the transient regime of swelling nears completion, further changes in stress state or irradiation temperature have no influence on the swelling rate thereafter.

1. Introduction

Structural steels anticipated for fission and fusion applications will experience time-dependent changes in the radiation environment, i.e., stress level, stress state, irradiation temperature and dpa rate. All of these variables are known to affect void swelling when maintained at constant values [1]. There are insufficient data available, however, to allow confident prediction of the effects of changes in these variables on subsequent behavior of swelling during continued irradiation. Data on the effect of changes in stress state or irradiation temperature are especially lacking.

In this paper are presented the results of a reirradiation experiment conducted in EBR-II that addresses the effect of an abrupt loss of the stress and/or an increase in the irradiation temperature once swelling is already in progress. This 'history effect' experiment was designed to determine whether there is some memory of previous conditions that persists after a change has occurred. The experiment was conducted in the mid-1970s but the collected data has only recently been examined for relevance to current design needs. It involved the reirradiation of concentric tube segments where the inner tube was originally irradiated under stress but the outer tube was stress-free.

2. Experimental details

2.1. Materials

Annealed Type 304L stainless steel was obtained from a surveillance program conducted to qualify EBR-II Mark-II metal driver fuel pins. With the exception of a few unirradiated archive specimens, the specimens used in this study were taken from an fuel pin experiment consisting of encapsulated driver fuel in which the fuel elements, clad with Type 304L stainless steel, were sodium-bonded and sealed in Type 304L stainless steel capsules. The cladding and the capsule tubes were manufactured from different heats of Type 304L stainless steel. Sections of the unirradiated, as-fabricated cladding and capsule tubing materials were analyzed by wet chemistry techniques, and the results are given in Table 1, showing some differences in elemental composition that are not thought to impact the interpretation of the derived data.

2.2. Specimen preparation and measurements

Cladding-capsule specimen pairs were obtained by sectioning through cladding and capsule tubing at the same elevation. The cladding-capsule pairs experienced essentially the same flux-spectral exposures, but the capsules were stress-free and operating at ~50 °C lower temperatures. In each of four fuel pins specimens

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doi:10.1016/j.jnucmat.2008.12.105

Table 1
Composition, wt%.

	Mn	P	S	Si	Ni	Cr	Ti	Cu	Mo	Co	C
Cladding	1.66	0.016	0.014	0.59	10.6	18.3	<0.01	–	0.02	–	0.03
Capsule	1.37	0.010	0.007	0.62	9.26	18.3	0.02	0.074	0.02	0.05	0.03

were extracted above the fuel column while second specimens were extracted from the fueled regions at somewhat higher fluences. Under gas loading the stress levels are essentially identical at both positions in a given pin.

Fuel was removed from the cladding sections by chemical dissolution of the fuel. Cladding and capsule sections had nominal outer diameters of 4.42 and 7.37 mm, respectively, and were cut to be approximately 25.4 mm long. Each tube had its ends ground and polished to ensure suitable surfaces for length measurements before and after reirradiation.

Specimen length, diameters, and densities were measured before and after reirradiation. Length and diameter measurements were obtained from an optical-reading dial gauge, having a precision of ±0.005 mm. Densities were determined by immersion techniques having an accuracy of approximately ±0.0005 of the measured values.

The specimens selected for this study are described in Table 2 which presents the initial irradiation temperature, neutron exposure ($E > 0.1$ MeV), and peak hoop stress level. The initial irradiation temperatures are calculated, time-averaged, mid-wall values, and the stresses are based on the end-of-life plenum pressures measured on the fuel elements prior to sectioning into specimens for reirradiation.

There was originally some uncertainty in the design phase of the experiment concerning the nature and history of the stress state in the cladding. Initially the stress level induced in cladding by fission gases is low but builds continuously with irradiation. It is unknown whether the fuel contacted the cladding in these pins and if so, when contact was made. The general nature of the stress state can be deduced, however, by examining the post-irradiation relationship between length strains and diametral strains.

During the initial irradiation cladding stresses can increase progressively for two reasons. First, fission-product-gas pressure increases linearly with fuel burn-up above 2.0 at.% [2–4]. Second, the fuel swells and is known to contact the cladding near burn-up levels of ~2.0 at.% [2–4]. However, once swelling in the cladding becomes large, the cladding tends to move away from the fuel. In the absence of fuel–clad mechanical interaction, the ratio of hoop stress to axial stress is expected to be 2/1 arising only from gas loading. The possible deviation from the 2/1 value depends on the time-averaged contribution from mechanical interaction. If fuel swelling is the only source of stress and the shear strength of the fuel is large, the stress state approaches 1/1. The actual stress state history lies somewhere between these two states and possibly changes with time.

The nature of the stress state can be determined from a comparison of the integrated length changes with the integrated diametral strains. If the total length change measured for the fuel element corresponds to the integrated average of the total diameter profile (creep plus swelling), the strain state and therefore the stress would be 1/1. If, however, the measured total length change corresponds to the integrated average of the swelling profile, the stress state would be 2/1, since creep, which is volume conservative, is not expected to contribute to length change for this stress state [1].

Results of the dimensional analysis for four fuel elements are shown in Table 3. It was concluded that the stress state that best describes the time-averaged behavior of the fuel elements is nearer to 2/1 biaxial than 1/1 biaxial, since the measured length changes of the element correspond closely to the average swelling strain. Thus, if fuel cladding contact occurred it had very little effect on the integrated strains, probably because the cladding was observed to swell very strongly and thereby minimize fuel–clad interaction.

The observation of a primarily 2/1 biaxial stress state is important for analysis of the subsequent deformation state, because we can assume that the stress on the cladding is a function only of

Table 3
Results of analysis of length change of four fuel elements to determine the biaxial stress ratio, showing that the stress state was determined primarily by gas loading and not by fuel–clad mechanical interaction.

Pin number	1/3 of integrated average of swelling (mm)	Integrated average of total diameter change (mm)	Total length change (mm)
265	21.3	35.8	19.7
267	15.6	26.3	14.5
284	16.0	30.1	15.8
200	17.3	28.1	17.3

Table 2
Initial conditions of previously irradiated cladding and capsule tubes.

Specimen number	Fluence, 10^{26} nm^{-2} ($E > 0.1$ MeV)	Time averaged temperature, (°C)	Total diametral change (%)	Swelling (%)	Peak hoop stress (MPa)
27	5.92	528	9.42	18.33	116
28	4.10	557	2.97	6.10	116
31	6.02	532	8.56	15.81	118
32	4.18	552	4.09	7.92	118
35	5.62	548	9.29	15.87	100
36	4.19	577	2.77	5.19	100
41	0	–	0	0	0
23	4.34	498	2.28	4.74	40
24	2.85	556	0.88	0.90	40
29	5.92	480	2.03	6.66	0
30	4.10	513	0.39	1.53	0
33	6.02	477	2.21	7.03	0
34	4.18	509	0.50	1.93	0
37	6.02	493	2.11	7.38	0
38	4.19	527	0.37	1.77	0
43	0	–	0	0	0

the plenum gas pressure, which is known as a function of time. The hoop stresses ascribed to the cladding specimens in Table 2 were calculated using the gas-pressurized thin-wall tube approximation.

2.3. Reirradiation conditions

The reirradiation vehicle was a NaK-filled sub-capsule centered inside a Mark B-7 capsule. The specimens were placed in four tiers inside the sub-capsule. All specimens were axially located within 66 mm of the reactor core center-plane. Within this region, the neutron flux profile was relatively flat, and therefore all specimens attained nearly the same additional fluence of 2.0×10^{26} n/m^2 ($E > 0.1$ MeV) or ~10 dpa.

The thickness of the helium insulation gap and the gamma heating generated in the sub-capsule and its contents determined the temperature which was measured using passive thermal-expansion-difference (TED) monitors known to have an accuracy of ±5 °C. The monitors indicated 540 °C in the top-tier and 546 °C in the bottom tier after corrections were made for irradiation-induced swelling of the stainless steel shell of the TEDs.

3. Results

Cladding and capsule strains were determined from the length, diameter, and immersion density measurements taken before and after each irradiation period. Length and diameter strains were taken as the ratio of the change in length and diameter to the initial length and diameter of each specimen.

The total swelling strain for the cladding and capsule specimens before and after the reirradiation experiment is shown in Figs. 1 and 2 as a function of neutron exposure. If differences in prior temperature are ignored, the cladding, even after the fuel has been removed, continues to swell at a rate that is significantly greater than that for the capsule tubing. Contrary to expectations prevalent when this experiment was conducted in the late 1970s, the swelling rate did not fall upon removal of the applied stress.

Note that once swelling in excess of several percent is attained the average swelling rate of the cladding at 540 °C in the second irradiation segment is ~1%/dpa as is known to be typical of AISI 304 and other 300 series steels [1,5,6]. While the average incubation intercept is on the order of 2–3 $\times 10^{22}$ n/cm^2 ($E > 0.1$ MeV) in the cladding, it is somewhat larger in the capsule material which was irradiated initially at lower temperatures, and the average

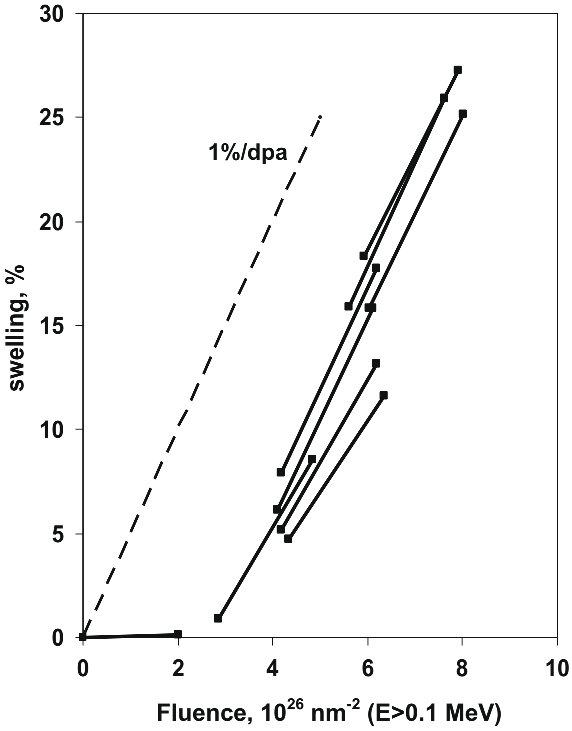

Fig. 1. Swelling observed in reirradiation of previously stressed cladding specimens.

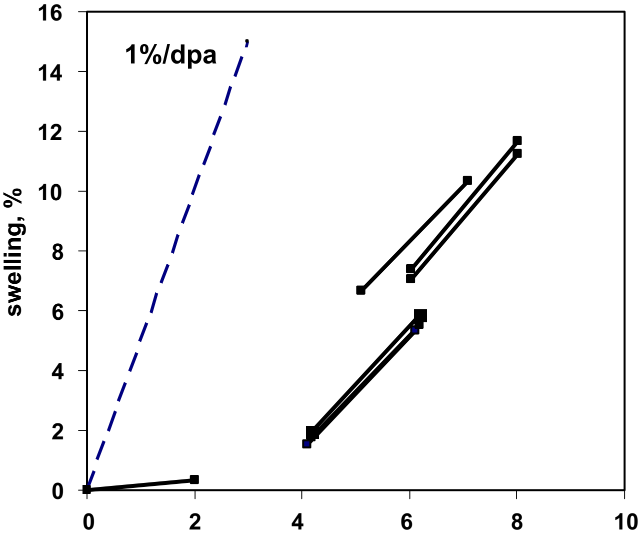

Fig. 2. Swelling observed in previously irradiated but unstressed capsule specimens.

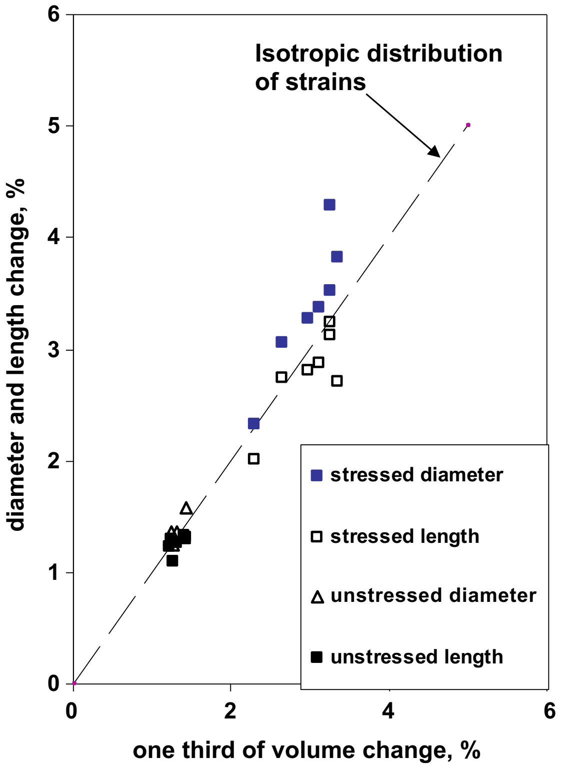

Fig. 3. Isotropic swelling was observed in the second irradiation segment in unstressed capsule specimens, but highly anisotropic swelling was observed in previously stressed cladding specimens. Note that in the stressed specimens the diameter changes are consistently larger than the corresponding length changes.

swelling rates are lower, indicating that transient swelling regime is probably still in progress in the second segment.

The spatial distribution of dimensional changes in the cladding and capsule specimens after reirradiation show the most interesting effects, as can be seen in Fig. 3. In this figure, the diameter and length changes that occurred during reirradiation are plotted against one-third of the volume swelling strain. This figure shows that while the capsule specimens essentially swelled isotropically during the reirradiation, the cladding appears to have swelled anisotropically. In all cases the diameter strain for the cladding is significantly larger than the length strain, as would be expected if the cladding retained some memory of the original 2:1 biaxial stress state.

The anisotropic components of the diameter strain in the cladding after reirradiation are shown in Fig. 4. The slope of the solid line drawn through the initial anisotropic strain values agrees well with the anisotropic strain rate expected for these claddings when driven by the calculated time-dependent gas loading. For each reirradiated cladding specimen, the dashed lines connect the initial and final strain values.

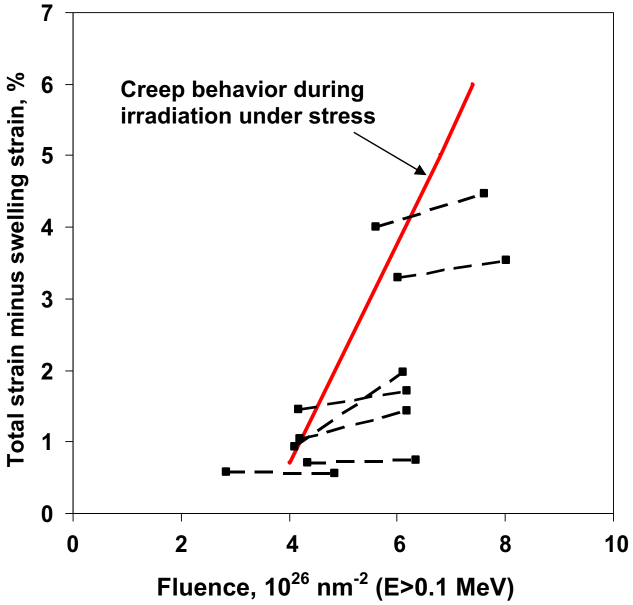

Fig. 4. Anisotropic strain components observed in previously stressed cladding specimens. The solid line represents both the trend line of the first irradiation sequence but also the expected behavior if the 2:1 biaxial stress state had been maintained in the second irradiation segment.

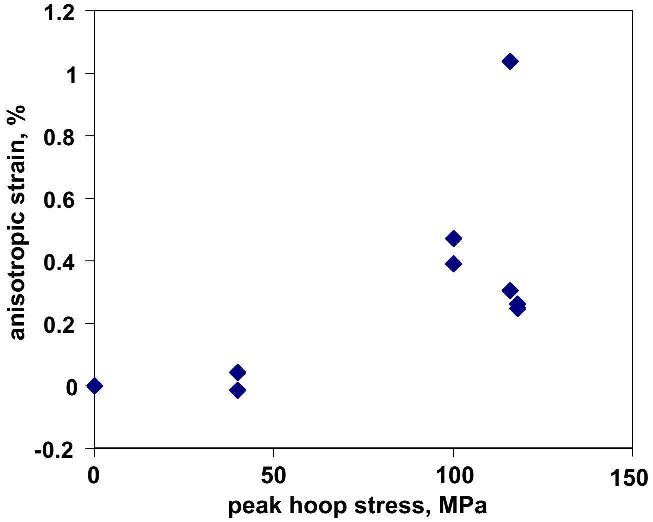

Fig. 5. Anisotropic growth strain observed as a function of final hoop stress level, showing the tendency of growth strain to increase with the stress level.

Fig. 4 shows that during the second irradiation segment, the average rate of anisotropic growth is substantially less than during the initial irradiation, but is nevertheless significant. It therefore appears that memory of the previous stress state was fading over the second irradiation segment. One would expect that the degree of anisotropy would increase with the hoop stress and roughly this is the behavior observed in Fig. 5, indicating qualitatively that the retained anisotropy is most likely related to the stress level.

4. Discussion

The results clearly show that there is a retained but probably diminishing memory in the microstructure of the previous stress state and that the memory is strongest for higher stress levels. This memory is probably expressed in the stress-induced anisotropic distribution of Burgers vectors of Frank loops and line dislocations that is known to develop during irradiation under stress [7–9]. Continued irradiation in the absence of stress is expected to erase this anisotropy eventually, but averaged over the lifetime of the dislocation array there is a net anisotropic growth attained. We can not from these data alone determine whether complete relaxation of dislocation anisotropy has occurred at or before 10 dpa, or whether relaxation might still be in progress, since we are seeing only the integrated effect of the anisotropy. Another radiation segment would be required to determine if the level of anisotropy is still growing or not.

There was also some speculation when the experiment was designed, however, that the steady-state swelling rate might depend on the stress level and possibly the stress state. It is now well-known, but it was not so well-known at the time this experiment was conducted, that stress only accelerates the end of the transient regime of swelling and not the steady-state swelling rate [1]. The expectation that the swelling rate would increase until it reached the terminal swelling rate of ~1%/dpa, and not fall for any reason thereafter, was also unknown when this experiment was conducted.

This experiment also involved substantial increases in the irradiation temperature for the unstressed capsule specimens while most of the cladding specimens were not subjected to large changes. From this study alone, we cannot with confidence determine the effect that temperature changes may have had on swelling. In an earlier study by Yang and Garner [10] on AISI 316 stainless steel it was shown that if the transient regime was nearly complete when temperatures were changed, neither increases or decreases in temperature had any effect on the subsequent swelling rate, especially if it was already approaching 1%/dpa.

Bloom and Wolfer performed a reverse version of our experiment [11]. They made flat tensile specimens from the unstressed walls of an irradiated EBR-II safety rod thimble and then subjected them to applied stresses arising from differential swelling that was driven by a higher-swelling steel. While their objective was to study in-reactor stress rupture, we can use their data to observe the influence of late-term increases in stress state on subsequent swelling.

In the Bloom–Wolfer experiment the specimens in the first segment were irradiated over a wide range of fluences and at temperatures of 400, 450 and 550 °C, while the second irradiation segment proceeded only at 450 °C. Thus the experiment not only explored increases in stress state but also isothermal and non-isothermal (both increases and decreases) temperature histories.

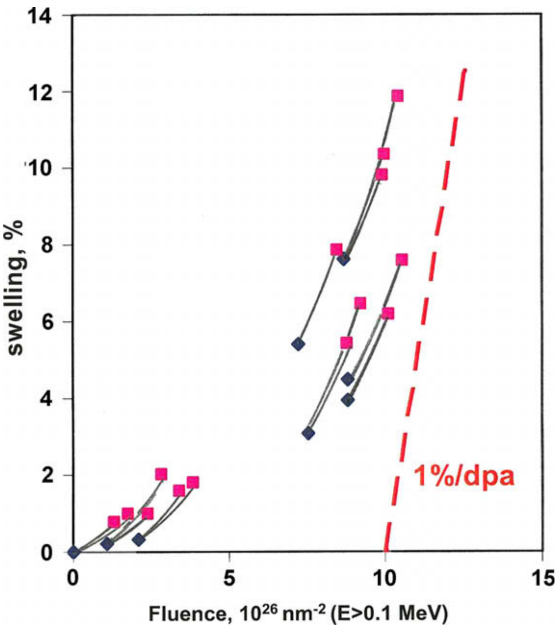

Fig. 6. Swelling observed by Bloom and Wolfer in another reirradiation experiment conducted on AISI 304 stainless steel where previously non-stressed specimens were subjected to stress in the second irradiation segment. Temperature changes were also involved in this experiment.

Fig. 6 shows that all swelling curves developed as now expected, increasing toward 1%/dpa with increasing fluence, although it appears that the low fluence specimens may have experienced a somewhat shorter transient regime as a result of stress application when compared to the higher fluence specimens that were beyond the transient regime before the second irradiation segment started. There was no obvious effect of temperature changes observed in this experiment, especially once swelling has exceeded ~3%.

Finally, it follows from these results that dislocation anisotropy arising from any source should lead to anisotropic growth during irradiation. Indeed, Bates and coworkers [12] showed that a preferred orientation of Burgers vectors is induced in AISI 316 stainless steel tubes during production by drawing. This leads to preferred growth in the length direction of gas-pressurized tubes during irradiation. As would be expected, the effect was most pronounced in the cold-drawn condition, but was still somewhat observable in identical tubes that were annealed prior to irradiation.

Interestingly, Bates showed that the effect of increasing gas pressure and thereby higher hoop stress was to progressively reduce the anisotropy. This can now be understood to arise from the fact that the gas-induced stress tends to generate growth in the diameter rather than in the length, and the competition of the two reduces the net effect of the drawing-induced anisotropy. In the current experiment any drawing-induced dislocation anisotropy should have been erased and replaced during the long first irradiation sequence. Therefore drawing-induced anisotropy played no role in our experiment.

5. Conclusions

If AISI 304 stainless steel was already swelling at a significant rate while under neutron irradiation and stress, removal of the stress will not change the subsequent swelling rate. However, a memory of the previous stress state is retained and produces an anisotropic distribution of strains consistent with the strain distribution behavior operating when the stress was present. It is clear however, that this is a transient behavior with the memory fading during continued irradiation. When there was no stress previously operating on the specimen, the strains during subsequent irradiation are isotropically distributed as would be expected in stress-free swelling [1].

Once the transient regime of swelling is nearing completion, changes in stress state or temperature appear to have no effect on the subsequent swelling rate.

Acknowledgements

The experimental portion of this work was conducted in the late 1970s under the auspices of the US Liquid Metal Reactor Program of USDOE. The recent analytical participation of F.A. Garner was sponsored by the Office of Fusion Energy, USDOE under Contract DE-AC06-76RLO 1830 at Pacific Northwest National Laboratory.

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