ELSEVIER

Journal of Nuclear Materials 283–287 (2000) 380–385

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Swelling, irradiation creep and growth of pure rhenium irradiated with fast neutrons at 1030–1330°C

F.A. Garner [a,*], M.B. Toloczko [a], L.R. Greenwood [a], C.R. Eiholzer [a], M.M. Paxton [b], R.J. Puigh [b]

[a] *Materials Resources Department, Pacific Northwest National Laboratory, 902 Battelle Boulevard, P8-15, Richland, WA 99352, USA*

[b] *Fluor Daniel Northwest, Richland WA, USA*

Abstract

This paper discusses the results of two series of experiments conducted on pure hcp rhenium in the EBR-II and FFTF fast reactors. In FFTF, density change data were derived from open tubes and solid rods irradiated at temperatures and fluences in the range of 1020–1250°C and 4.4–8.3 $\times$ 10^{22} n cm^{-2}, respectively ($E > 0.1$ MeV). Both density change and diametral change data were obtained from pressurized tubes irradiated in EBR-II to $\sim$0.65 and $\sim$ 5.1 $\times$ 10^{22} n cm^{-2} at temperatures between 1030°C and 1330°C. Analysis of the data shows that four concurrent processes contribute to the radiation-induced strains observed in these experiments. These are void swelling, transmutation-induced densification via production of osmium, irradiation creep and irradiation growth.

1. Introduction

Various bcc and hcp refractory metals and alloys have been proposed for high temperature applications in fusion reactors. However, with the exception of zirconium and to a lesser extent titanium, there is very little information on radiation damage of hcp metals. Rhenium in particular has been considered for fusion application both as a metal and as an alloy constituent, but very little published irradiation data are available, especially at sufficiently high dpa levels, to qualify rhenium as a structural component. Some limited data at low dose were published much earlier, and these data established that dislocation loops and dislocation line segments, as well as voids, form in irradiated rhenium [1,2].

This paper presents the results of two separate experiments at high irradiation temperature and high dpa levels, one experiment conducted in the Fast Flux Test Facility (FFTF) and another in the Experimental Breeder Reactor-II (EBR-II).

2. Experimental details

In the first experiment, thin-walled pressurized tubes of 22.4 mm length, inner and outer diameter of 4.57 and 4.06 mm, respectively, were prepared from pure (99.98%) rhenium wrought sheet. The sheet was rolled into an approximate cylindrical shape, electron beam welded, vacuum annealed at 1430°C for 1 h, cold swaged to round and annealed in hydrogen at 1650°C for 20 min. The welded tubing was then reduced 6% by cold drawing and annealed in hydrogen at 1650°C for 20 min. A second 6% cold drawing was followed by a vacuum anneal at 1450°C for 1 h. The tubes were then cold drawn 4.3% to final size. Rhenium end-caps were electron beam welded to the tube ends, heat treated in vacuum at 1150°C for 1 h, and pressurized with pure helium prior to sealing of the fill hole by laser beam welding.

Ten of these tubes were irradiated in EBR-II in two separate lithium-filled B-7A capsules. The tubes were distributed axially below the core mid-plane, receiving

* Corresponding author. Tel.: +1-509 376 4136; fax: +1-509 376 0418.

E-mail address: frank.garner@pnl.gov (F.A. Garner).

0022-3115/00/$ - see front matter
PII: S 0 0 2 2 - 3 1 1 5 (0 0) 0 0 2 2 5 - 7

doses ranging from 0.60 to 0.70×10^{22} n cm^{-2} ($E > 0.1$ MeV) in one capsule and 4.7 to 5.5×10^{22} n cm^{-2} in the other capsule. The exact location of each tube cannot be reconstructed from the available records, introducing some small uncertainty in the relative neutron exposure for each tube.

The subcapsule temperatures were 1030°C and 1330°C with an uncertainty of ±50°C. The capsules were filled with isotropically enriched 99.98% ^{7}Li which served to reduce pressurization by helium generation via the ^{6}Li (n, α) reaction. Another 10 tubes were irradiated at 1330°C, but only at the higher dose range.

The temperature of each subcapsule, which was not actively controlled, arose from the competition between gamma heating and gas gap resistance. The hoop stress levels during irradiation ranged from 25.1 to 71.5 MPa at 1030°C, and 16.1 to 44.9 MPa at 1130°C. At 1330°C the stresses ranged from 5.1 to 13.1 MPa.

Diameter changes were measured before and after irradiation by a non-contacting laser system described by Gilbert [3]. Selected tubes were cut to remove the end-caps, and immersion density measurements accurate to ±0.005% were used to determine the density before and after irradiation.

In the second experiment, five open tubes, 6.35 mm long and 6.27 mm diameter, of chemical vapor deposition (CVD) rhenium were irradiated in the Materials Open Test Assembly (MOTA) in FFTF with active temperature control to ±30°C at temperatures of 1017°C, 1077°C (two tubes), 1127°C and 1252°C. Also included at each of 1017°C and 1077°C were two small rods, 3.16 mm in diameter and 3.67 mm in length. At each temperature, one rod was in the as-received cold-worked condition, while the other was recrystallized prior to irradiation. The purity of both tubes and rods was 99.97% rhenium. Once again, 99.98% ^{7}Li was used to cover the specimens. Immersion measurements were performed on these two sets of specimens after irradiation to determine the *net* swelling. The fluence levels ranged from 4.4 to 8.3×10^{22} n cm^{-2} ($E > 0.1$ MeV), which were attained by placing the specimens at different axial positions in MOTA levels 4 and 5.

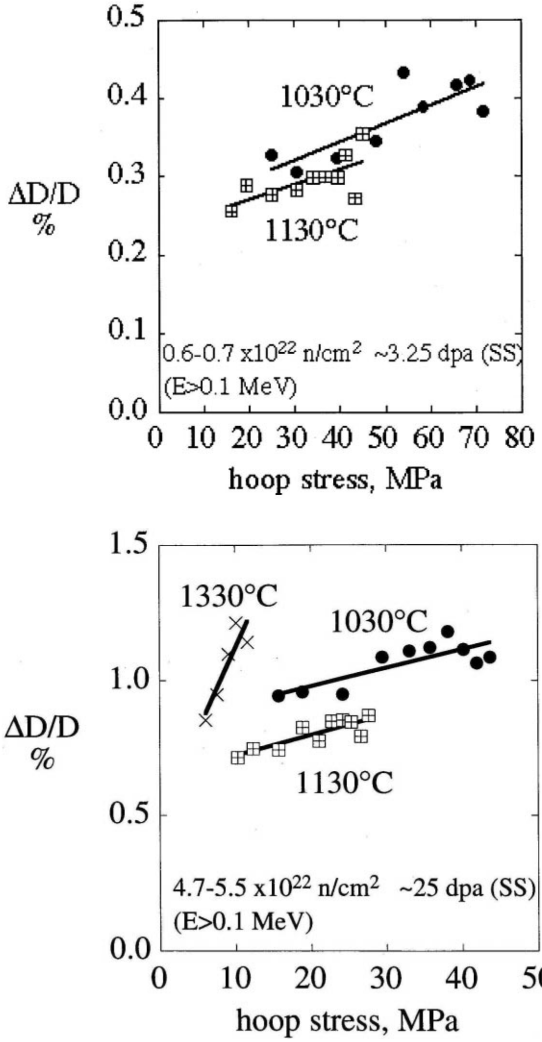

Fig. 1. Total diametral strains observed in pressurized tubes constructed from pure rhenium and irradiated in EBR-II.

3. Results

As shown in Fig. 1, the total diametral strains of the pressurized tubes irradiated in EBR-II were linear with stress at both low and high dose levels and at both irradiation temperatures, with the strains at 1030°C a little larger than those at 1130°C. The strains at 1330°C at the high dose are significantly larger than at lower temperatures. Note that all three high dose sets of data extrapolate to non-zero strains at zero stress, indicating volume strains of ~2.4%, 2.0%, and 1.5% at 1030°C, 1130°C and 1330°C, respectively, assuming the distribution of strain was isotropic. Based on the observation of voids in other studies of rhenium [1,2], it is assumed that these strains are largely due to void swelling, but as we will see later there was another contribution to the volume change.

Note that the dpa levels in Fig. 1 are for stainless steel rather than rhenium. This choice of units reflects two considerations. First, the dosimetry results for EBR-II and FFTF are routinely supplied in terms of fluence above 0.1 MeV for stainless steel, which can be used to assess the relative impact of the differing spectra in the two reactors. Second, there is no published displacement threshold for rhenium. This point will be addressed in more detail in Section 4.

Fig. 2 and Table 1 present the swelling of these tubes measured by density change. There is no apparent

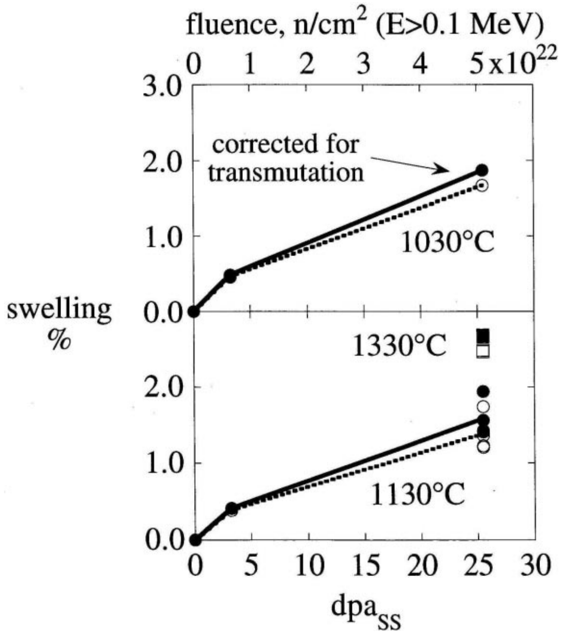

Fig. 2. Swelling of pressurized rhenium creep tubes, as measured by immersion density. It is assumed that void growth accounts for this increase in volume. Note that a small correction must be made to account for the transmutation-induced formation of osmium.

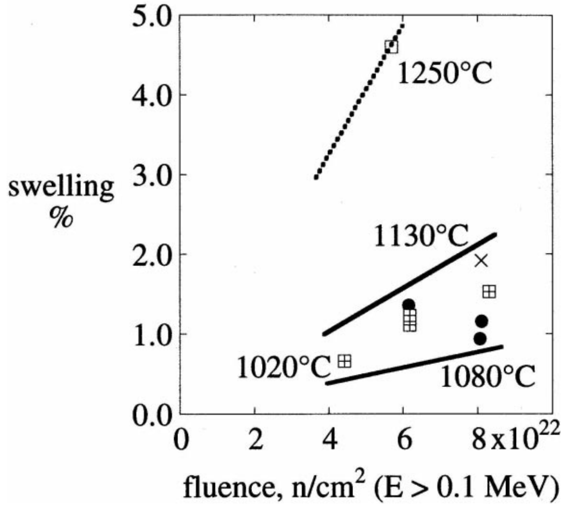

Fig. 3. Stress-free 'net swelling' of pure rhenium in FFTF, as measured by immersion density. These values are strong underestimates of the void swelling due to very large densification contributions occurring from osmium formation, as shown in Table 2.

correlation of swelling with stress level. Note that a relatively small correction has been made in Fig. 2 to account for volume decreases arising from transmutation of Re to Os. This correction must be calculated and will be described in Section 4 of this paper.

Fig. 3 and Table 2 show the density change and corrected swelling observed in FFTF, with the highest values, 4.6% and 5.51%, respectively, observed at 1250°C. Note that in the range 1017–1127°C there appears to be no significant effect of either the recrystallization or the CVD treatment on the swelling. Based only on the one datum at 1250°C there appears to be some influence of temperature on swelling rate, increasing as the temperature increases. Note in Table 2, however, that in contrast to the EBR-II data, rather large changes must be made in the FFTF data to account for osmium formation.

4. Discussion

It is important to recognize that rhenium is easily transmuted to osmium even in fast reactors, and that the amount of transmutation is very sensitive to the differ-

Table 1
Swelling of pressurized tubes after irradiation in EBR-II

Irradiation temperature (°C)	Neutron fluence, (n cm^{-2}) ($E > 0.1$ MeV)	Dose in stainless steel (dpa)	Hoop stress (MPa)	Swelling (%) $\Delta\rho/\rho_0$, without and (with) correction for osmium formation
1030	0.60–0.70	3.0–3.5	54.0	2.27[a]
1030	0.60–0.70	3.0–3.5	71.5	0.464 (0.494)
1130	0.60–0.70	3.0–3.5	30.4	0.455 (0.485)
1130	0.60–0.70	3.0–3.5	43.2	0.385 (0.415)
1030	4.7–5.5	23.5–27.5	43.7	1.673 (1.903)
1130	4.7–5.5	23.5–27.5	10.3	1.739 (1.969)
1130	4.7–5.5	23.5–27.5	15.7	1.225 (1.455)
1130	4.7–5.5	23.5–27.5	18.9	1.360 (1.590)
1130	4.7–5.5	23.5–27.5	27.7	1.204 (1.434)
1330	4.7–5.5	23.5–27.5	5.1	2.484 (2.714)
1330	4.7–5.5	23.5–27.5	13.1	2.465 (2.695)

[a] This value is thought to represent an experimental error.

Table 2
Swelling of specimens after irradiation in FFTF–MOTA as determined by density change measurements

Specimen type	MOTA level	Irradiation temperature (°C)	Neutron fluence (10^{22} n cm^{-2}) ($E > 0.1$ eV)	Dose in stainless steel (dpa)	Swelling (%) $\Delta\rho/\rho_0$, without and (with) correction for osmium formation[a]
Open tube, CVD	5	1017	6.15	25.1	1.36 (2.28)
Open tube, CVD	4	1077	8.31	35.1	1.53 (2.76)
Open tube, CVD	5	1077	4.42	17.2	0.66 (1.38)
Open tube, CVD	4	1127	8.09	34.2	1.92 (3.10)
Open tube, CVD	5	1252	5.69	23.0	4.60 (5.51)
Rod, recrystallized	4	1016	8.10	34.2	1.16 (2.34)
Rod, cold worked	4	1016	8.06	34.1	0.94 (2.12)
Rod, recrystallized	5	1077	6.18	25.3	1.23 (2.21)
Rod, cold worked	5	1077	6.18	25.3	1.11 (2.09)

[a] $\Delta\rho/\rho_0$ is the change in density.

ences in neutron spectra between that in EBR-II and the somewhat softer spectra in FFTF. This sensitivity arises from some strong neutron resonance reactions, and therefore the osmium formation rate is also very sensitive to the relatively minor spectral differences between levels 4 and 5 in FFTF–MOTA [4,5]. In EBR-II at the highest exposure of 5.5×10^{22} n cm^{-2}, the calculated amount of Os formed was ~3.2%, and in the low exposure experiment it was only 0.22%. In FFTF, however, 11.0% Os was formed at mid-plane in level 5 and 17.4% at mid-plane in level 4. Our estimates of the uncertainties in these calculations are dominated by self-absorption corrections, but the calculated values of osmium are felt to be accurate to ±20%.

Although Re and Os are fully soluble in each other at all concentrations, Os formation leads to a reduction in lattice parameter. Using a correlation by King [6], this produces a maximum densification of only 0.2% in the EBR-II experiment, but 0.78% and 1.23% densification is calculated for FFTF–MOTA at the mid-plane of levels 5 and 4, respectively. Thus, the measured density changes represent underestimates of the void swelling by these amounts. In the absence of any contrary information it is reasonable and necessary to assume that the densification is isotropically distributed and there is no significant enhancement of swelling by stress. The net swelling values shown in Table 2 are in rough agreement with the swelling values derived from the zero-stress intercepts of the creep data at 1030°C (2.0% vs 2.4%) and 1130°C (1.6% vs 2.0%), but somewhat larger at 1330°C (2.5% vs 1.5%). It therefore appears that these are relatively safe assumptions.

When corrections are made for both displacive and transmutation aspects of the neutron spectra, the data from both reactors can be plotted together as shown in Fig. 4. Considering the differences in transmutation rate, displacement rate, stress level and starting condition, all data in the range 1016–1330°C, except one at 1250°C, exhibit remarkably similar behavior, with a mean swelling rate of 0.08 (±0.02)% dpa^{-1}, based on stainless

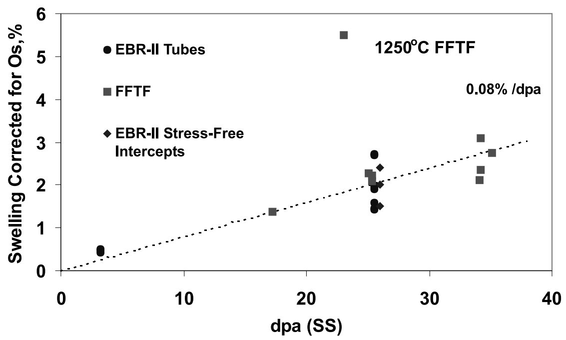

Fig. 4. Compilation of transmutation-corrected swelling data from both experiments vs. dpa for stainless steel, showing that all data except one at 1250°C exhibit very similar behavior.

steel dpa. This implies that osmium formation probably does not strongly influence the swelling rate. It is also interesting that there does not appear to be a large incubation regime of swelling, although the data in Fig. 2 might imply a higher initial swelling that saturates to a lower level at higher exposure. Based on one data point, however, this is too speculative to entertain.

Unfortunately, there is no published and accepted value of the atomic displacement threshold E_d for rhenium, leading to some uncertainty in the calculation of the dpa levels for these experiments. If we assume that the accepted threshold of 90 eV for neighboring tungsten can be used [7], then the dpa calculated for these spectra is ~44% of that calculated assuming stainless steel with $E_d = 40$ eV.

Note that the highest swelling value obtained in this experiment was ~5.5% at ~ 5.7×10^{-22} n cm^{-2} and 1250°C. Assuming the E_d value currently used for W leads to a swelling rate of ~0.55% dpa^{-1}, indicating that hcp rhenium may have a maximum swelling rate comparable to fcc metals such as Cu at ~0.5% dpa^{-1} [8] and austenitic steels at ~1.0% dpa^{-1} [9]. However, this is only a single data point. If we base our estimates of the swelling rate on all other data except that at 1250°C, we come to the conclusion that swelling in rhenium proceeds at a relatively temperature-independent rate of ~0.2% dpa^{-1}, similar to that of pure iron and Fe–Cr binary alloys [10].

Turning to the non-hydrostatic strains, it is known that irradiation creep is an inherently anisotropic process, and that hcp metals and alloys often exhibit irradiation growth, a separate process that is also anisotropic and highly dependent on the starting dislocation and grain texture. If we calculate the stress-normalized creep strains for the EBR-II experiment after deducting the net swelling and transmutation-induced strains, we can see in Fig. 5 that the creep strains are similar at 1030°C and 1130°C. The creep behavior mirrors that of the swelling in that it is also non-linear with dose. These two conclusions are in agreement with expectations based on other studies [11].

If we calculate the creep strain rates in EBR-II, Fig. 6 shows that the creep modulus B is the same at both 1030°C and 1130°C as expected from previous experience in iron-based fcc alloys [11]. The measured value of the creep-swelling coupling coefficient, D, for rhenium is $\sim 1.3 \times 10^{-2}$ MPa^{-1}, comparable to that observed in various fcc and bcc alloys, but the value of the creep compliance in the absence of swelling, B_0, is negative at about -3×10^{-6} MPa^{-1} dpa^{-1}, based on dpa for stainless steel. This negative value implies that the irradiation growth is strongly contributing to shrinkage of the tube diameter by making a strain contribution that is larger than that of irradiation creep, but smaller than the combined effects of void swelling and transmutation.

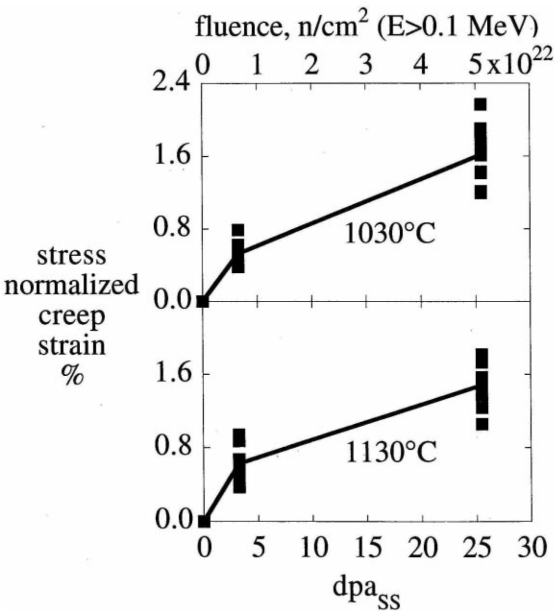

Fig. 5. Stress-normalized diametral creep strains in the EBR-II experiment.

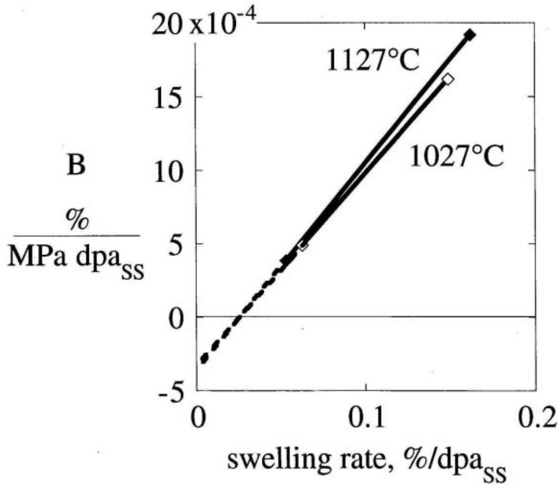

Fig. 6. Derived creep coefficients from the EBR-II experiment.

The creep coefficients are also comparable to those observed in bcc and fcc iron-based alloys [10,11], but the growth strains are comparable in magnitude and perhaps somewhat larger than the creep strains.

5. Conclusions

The radiation-induced changes in dimensional stability of hcp rhenium are much more complicated than those of typical fcc and bcc metals, especially those metals which do not undergo significant transmutation. Whereas both void swelling and irradiation creep contribute to increases in tube diameter, there are potentially very strong negative strain contributions to the tube diameter from irradiation growth and transmutation. The latter process is dependent on sometimes rather small differences in neutron spectra.

When all of these factors are taken into consideration, it appears that the creep and swelling rates of hcp rhenium in the range 1030–1330°C are comparable to those of fcc and bcc alloys. The influence of stress, starting condition and dpa rate do not appear to exert a strong influence on the swelling behavior in the limited temperature range studied.

Acknowledgements

This work was supported by the Office of Fusion Energy, US Department of Energy under Contract DE-AC06-76RLO 1830.

References

[1] J.L. Brimhall, G.L. Kulcinski, H.E. Kissinger, B. Mastel, Radiat. Eff. 9 (1971) 273.

[2] J.L. Brimhall, H.E. Kissinger, G.L. Kulcinski, in: J.W. Corbett, L.C. Ianniello (Eds.), in: Proceedings of the International Conference on Radiation-induced Voids in Metals, AEC Symposium Series 26, 1972, p. 338.

[3] E.R. Gilbert, B.A. Chin, In-reactor measurement technology, in: Proceedings of the 10th Conference on Effects of Radiation on Materials, ASTM STP, vol. 725, 1980, p. 665.

[4] L.R. Greenwood, F.A. Garner, J. Nucl. Mater. 212–215 (1994) 635.

[5] F.A. Garner, L.R. Greenwood, Radiat. Eff. Def. Solids 144 (1998) 251.

[6] H.W. King, J. Mater. Sci. 1 (1966) 79.

[7] Recommendations of IAEA Working Group on Reactor Radiation Measurements, Nucl. Eng. Des. 33 (1975).

[8] F.A. Garner, M.L. Hamilton, T. Shikama, D.J. Edwards, J.W. Newkirk, J. Nucl. Mater. 191–194 (1992) 386.

[9] F.A. Garner, J. Nucl. Mater. 122&123 (1984) 459.

[10] F.A. Garner, M.B. Toloczko, B.H. Sencer, J. Nucl. Mater. 276 (2000) 123.

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