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Received by OSTI

FEB 0 8 1991

PNL-SA--19061

DE91 007202

SWELLING AND MICROSTRUCTURE OF HIGH PURITY NICKEL IRRADIATED WITH FAST NEUTRONS IN EBR-II

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January 1991

To be included in the Fusion
Reactor Materials Semiannual
Progress Report DOE/ER-0313/9

Work Supported by
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

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Swelling and Microstructure of High Purity Nickel Irradiated with Fast Neutrons in EBR-II--
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OBJECTIVE

The objective of this effort is to provide an understanding of the radiation-induced evolution of microstructure in pure nickel, a metal often used in fission-fusion correlations.

PROGRESS AND STATUS

Introduction

In an earlier report,⁽¹⁾ a review was presented of the parametric sensitivity of void swelling in nickel, a metal often selected in fission-fusion correlation studies as a model material for studying neutron spectral effects. It was shown that nickel's response to radiation is often dominated by its tendency to saturate in swelling. At moderate dpa levels, this tendency often overshadows the influence of other variables and calls into question the validity of uncritically using nickel for comparative irradiations conducted in widely disparate irradiation environments.

Figure 1 shows the swelling response of pure nickel to thermomechanical starting condition and irradiation temperature as observed in a recent experiment. These data were presented in an earlier report and were derived from an experiment conducted in EBR-II to a nominal level of 14 dpa.⁽¹⁾ Reevaluation of the exposure levels has shown that seven of the nine specimens were irradiated at 14 dpa, but two (annealed and cold-worked conditions at 600°C) were irradiated at a somewhat lower elevation in EBR-II. These two specimens reached only 12 dpa.

Note that the cold-worked and the cold-worked and aged conditions yield swelling values that are relatively insensitive to irradiation temperature, declining very slowly at higher temperatures. The annealed condition, however, yields swelling that is essentially the same as that of the other two conditions at 425°C but is strongly sensitive to irradiation temperature above 425°C. Given the consistency of the data at 12 and 14 dpa for the cold worked and the cold worked and aged conditions, it appears that saturation of swelling for these conditions must occur well below 12 dpa and is only slightly sensitive to irradiation temperature. It is not certain whether the annealed condition has reached a saturation state at either 500 or 600°C. A second discharge of this experiment at higher fluence is now being examined and should answer this question. Saturation definitely appears to have been reached at 425°C, however.

Since most fission-fusion correlation experiments are based on microstructural comparisons involving voids, dislocations and loops, it was decided to examine the microstructures of these specimens in order to provide an understanding of the saturation process.

Experimental

High purity (99.999%) nickel in three thermomechanical treatment conditions was irradiated in EBR-II in the AA-14 fusion materials experiment at irradiation temperatures of 425, 500, and 600°C to doses of 12-14 dpa. The thermomechanical treatment conditions were solution annealed, cold worked, and cold worked and aged. The specific treatment conditions are given in Table 1.

Swelling values for the 12-14 dpa discharge were determined using immersion density measurements accurate to $\pm 0.16\%$ swelling. The microstructures of both the initial and irradiated material conditions were determined using a JEOL JEM 1200EX transmission electron microscope operated at 120 kV. Specimen thicknesses were established by means of stereo pairs. The data for specimens irradiated to 12-14 dpa at 425 and 600°C have been analyzed and are presented here. Analysis of the 500°C data at 12-14 dpa is still in progress. Density measurements for the second discharge will be completed shortly.

(a) Pacific Northwest Laboratory is operated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830.

Figure 1. Swelling of pure nickel observed after 12-14 dpa in EBR-II as a function of irradiation temperature and starting condition. All data taken at 14 dpa with the exception of two data at 12 dpa, each marked with an asterisk.

Table 1 Thermalmechanical Treatment Conditions for Nickel		
Alloy Code	Condition	Treatment History
BN	Solution Annealed (SA)	975°C for 10 min/water quench
BP	Cold Worked (CW)	30% cold rolled
BR	Cold Worked and Aged (CWA)	30% cold rolled/650°C for 10 h/ water quench

Results

Micrographs showing the initial dislocation microstructures for the three thermomechanical treatment conditions are presented in Figure 2, and the dislocation densities are tabulated in Table 2. The presence of a non-trivial dislocation structure in the solution-annealed state was surprising and is probably a consequence of the use of rapid quenching after annealing. A number of dislocation loops are evident in the structure, an observation that is consistent with microstructures typically developed by rapid quenching. In general, there were no dislocation cells or highly pinned networks of dislocations in the annealed specimen in contrast with that of the other two specimen conditions. The aged condition contained small dislocation cells with internal dislocation-free areas, whereas the cold worked condition contained moderately high densities of dislocations inside the cells.

After irradiation, the dislocation densities have fallen in all specimens, especially in the solution annealed condition at 600°C. At each temperature the dislocation densities in the cold worked and the cold worked and aged conditions have converged at each irradiation temperature. These convergence values decrease slightly with temperature.

Figure 2. Micrographs of starting dislocation structures of (a) annealed and quenched, (b) cold worked, and (c) cold worked and aged pure nickel.

A comparison of the irradiation-produced microstructures at 425°C is shown in Figure 3, and at 600°C in Figure 4 (note the difference in magnification in these two sets of micrographs). The low void density in the solution annealed material at 600°C is clearly evident, as are the smaller average void sizes in the solution-annealed material at both temperatures.

Microstructural parameters for both the unirradiated and irradiated material conditions are presented in Table 2. A detailed analysis of the distribution in the void population following irradiation is shown in Figure 5 where the void number densities are plotted as a function of size. The void population in the cold worked and in the cold worked and aged materials involves a large range of sizes at both temperatures that was not observed in the annealed material.

Table 2
Microstructural Parameters

Condition	Immersion Density Swelling (%)	IIM Measured Swelling (%)	Void Density 10^{19} (m^{-3})	Mean Void Diameter (nm)	Dislocation Density ($10^{13} m^{-2}$)
Solution Annealed					
Unirradiated	--	--	--	--	4.6
425°C to 14 dpa	5.8	4.1	25.4	62	3.0
600°C to 12 dpa	2.0	1.8	1.3	124	<0.2
Cold Worked					
Unirradiated	--	--	--	--	>20
425°C to 14 dpa	6.1	7.6	7.6	111	1.8
600°C to 12 dpa	5.1	7.0	0.48	268	~1
Cold Worked and Aged					
Unirradiated	--	--	--	--	14
425°C to 14 dpa	6.3	5.2	20.0	72	2
600°C to 14 dpa	5.3	7.6	0.78	241	-0.8

The void data presented in Table 2 and Figure 5 also show that the cold worked and aged material at both temperatures develops a void structure that is intermediate to that of the other two conditions. Cold working leads to the lowest void number densities at both irradiation temperatures, but the largest void sizes. The solution annealed material has the highest void densities and the smallest void sizes at both irradiation temperatures. The intermediate void sizes and number densities of the cold worked and aged material, however, are associated with the highest swelling levels. It is considered particularly significant that at 425°C, the postirradiation microstructure and the swelling of the annealed and the aged

Figure 3. Irradiation-produced microstructures at 425°C for (a) annealed, (b) cold worked, and (c) cold worked and aged pure nickel.

Figure 4. Irradiation-produced microstructures at 600°C for (a) annealed, (b) cold worked, and (c) cold worked and aged pure nickel. Note the difference in magnification compared to that of Figure 3.

Figure 5. Influence of temperature and starting condition on void distributions in irradiated nickel.

Figure 6. Heterogeneity observed in the void population in the (a) cold worked condition after irradiation at 600°C and (b) the cold worked and aged condition after irradiation at 500°C.

specimens are rather comparable, but at 600°C this is not true. This probably reflects the strong dependence of Frank loop density on irradiation temperature. At the lower temperature the loop microstructure is known to be the most dense and therefore probably the most persistent, resisting the tendency toward collapse in density that accompanies unfauling and interaction of loops.

While the void population in the annealed condition was very homogeneous, those of the cold worked and the cold worked and aged conditions were quite heterogeneous and contained a wide variability in size and number distribution in a given volume. This is emphasized in Figure 6 where two material conditions are shown, each with large local variations in void size and density in a relatively small region. This also leads to a larger uncertainty in void size and density determinations for those conditions. The heterogeneity probably arises from the heterogeneity of the starting dislocation and cell network, as illustrated in Figure 2.

The interaction of voids with the dislocation population is also evident in Figure 6. It was common at all irradiation conditions to find dislocations connecting voids, with almost no independent dislocations that were not associated with voids.

It appears that those material conditions that had either preexisting or easily developed and persistent loop microstructures all evolved to essentially the same saturation microstructure at a given temperature. For those cases where dislocation networks were harder to form, especially at higher temperatures, saturation microstructures either failed to develop or were still in the process of developing at 12-14 dpa. Examination of the second discharge of this experiment at higher fluence is underway and may illuminate which of these two possibilities was occurring. It does appear, however, that by using the microstructural data we can identify the major reasons for the behavior observed in this experiment.

First, nickel has a relatively high stacking fault energy. One consequence of this is that dislocations are not stabilized against glide by separation into partials and stacking faults. Therefore, during cold working, nickel does not develop dislocation densities that approach those of stainless steel ($\geq 3 \times 10^{15} \text{ m}^{-2}$)⁽²⁾ since the higher level of glide leads to greater levels of dislocation annihilation. When the mobility of dislocations is enhanced by radiation-induced point defects, the density falls faster and even further. The dislocations formed either by quenching or by Frank loop unfauling in annealed material glide and annihilate at rates at 600°C that do not allow the attainment of a high saturation density. It has been shown that the saturation density at a given irradiation temperature and displacement rate is determined by the competition of various loss and production mechanisms of dislocation line length.⁽²⁾ The loss rate associated with glide and annihilation events is probably affected by the high stacking fault energy much more than is the production rate.

Second, it appears that voids, once formed, thereafter become the most effective agents in maintaining the dislocation density, trapping the ends of dislocations at void surfaces. It is probably the case that only in starting conditions where voids can nucleate prior to the complete collapse of the dislocation network can a saturation density be reached and maintained. Even then the density of dislocations will be temperature dependent, decreasing somewhat as the void density decreases with irradiation temperature. On the other hand, the voids probably also serve as internal surfaces that attract, annihilate and thereby reduce any "excess" length of individual dislocations between voids. Thus the onset of swelling probably contributes to the collapse in dislocation density.

Discussion

If the saturation level of swelling is related to the time averaged dislocation density in the early phase of the irradiation, then it would be expected that the most swelling would occur in conditions where heavily pinned networks existed prior to irradiation or at lower irradiation temperatures where loop nucleation would proceed at higher and more stable levels. It is quite possible that the steepness of a swelling curve for an annealed and slow cooled condition with irradiation temperature might be somewhat different from that shown in Figure 1 for the annealed and quenched condition. Aging of heavily pinned cold worked structures provides small open areas within the dislocation cells that may enhance void nucleation a small amount in the early stage of the irradiation and thus lead to slightly more swelling than attained in cold-worked material.

It is obvious, however, that the postirradiation dislocation densities observed here for all conditions and temperatures are at least one order of magnitude lower than that observed in typical stainless steels at this temperature ($3-8 \times 10^{14} \text{ m}^{-2}$).⁽²⁾ This radiation induced collapse in dislocation density probably accounts for the saturation in swelling. It has been shown that if the void sink strength is much larger than the dislocation sink strength, then the swelling rate will strongly decrease.⁽³⁾ Stainless steels do not exhibit such a collapse in density and have not exhibited saturation behavior at any relevant swelling level (<80%). It has also been suggested that swelling saturation may in part result as a consequence of dislocation segments that are relatively short and that terminate on both ends on void surfaces.⁽³⁾ The microstructural observations of this study are consistent with both of these proposals.

If it is assumed that voids with larger diameters have been growing longer and that voids nucleate easiest at higher dislocation densities, then the void and dislocation data derived in these studies are consistent

with these assumptions at both irradiation temperatures. This suggests that voids nucleate relatively early in the irradiation but only grow while the dislocation density is relatively high. The dislocation density probably falls with irradiation at a rate dependent on both the initial dislocation density and the irradiation temperature, and when it reaches the low saturation value dictated by the current void density, the growth of voids stops.

Conclusions

Void nucleation in pure nickel during irradiation probably proceeds while the dislocation density is relatively high. This condition can be reached most easily in either of two sets of conditions. First, in annealed nickel, it may occur at relatively low irradiation temperatures where loop nucleation rates and therefore loop stability are the highest and most persistent. Second, it also occurs when the starting dislocation density is relatively high and strongly pinned in dislocation cell walls produced during cold working. Aging leads to reduction of the dislocation density within the cells but retains the cell walls themselves. This leads to a slightly higher level of swelling.

The population of dislocations will eventually collapse regardless of the starting state since the high stacking fault energy of nickel precludes the formation of partial dislocations with their stronger level of pinning. This causes easy glide and annihilation of dislocation line length. The collapse in turn leads to an imbalance of sink strengths and a strong reduction of the void growth rate. In highly pinned starting structures, the dependence of the saturation levels of both dislocations and void swelling are only slightly sensitive to temperature. In unpinned structures, however, the temperature dependence is very strong, probably reflecting the temperature dependence of loop size and density and the resultant influence on dislocation evolution.

Future Work

Analysis of the 500°C microstructural data will proceed. Measurement of the density changes produced by irradiation at the higher displacement level will also be completed.

References

1. Garner, F. A., "Influence of Material and Environmental Variables on the Neutron-Induced Swelling of Nickel." Fusion Reactor Materials Semiannual Progress Report, DOE/ER-03138, March 31, 1990, pp. 125-135.
2. Garner, F. A., and W. G. Wolfer, "A Model for the Evolution of Network Dislocation Density in Irradiated Metals." ASTM STP 782 (1982) pp. 1073-1087.
3. Wolfer, W. G., J. Nucl. Mater., 122-123 (1984) 367-378.

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02 / 20 / 91