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MICROSTRUCTURAL EVOLUTION OF NEUTRON-
IRRADIATED Fe-Cr-Ni TERNARY ALLOYS
AT HIGH HELIUM GENERATION RATES

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MICROSTRUCTURAL EVOLUTION OF NEUTRON-IRRADIATED Fe-Cr-Ni TERNARY ALLOYS AT HIGH HELIUM GENERATION RATES.

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Abstract

A series of Fe-Cr-Ni ternary alloys were irradiated to 12-14 dpa in the Oak Ridge Research Reactor (ORR). In this experiment, most of helium atoms were produced by the $^{58}\text{Ni}(n,\gamma)^{59}\text{Ni}(n,\alpha)^{56}\text{Fe}$ sequential reaction, and, therefore, the helium generation rate was almost proportional to the nickel content, ranging from 27 to 58 appm He/dpa in 20 to 45 wt.% Ni alloys. Irradiation temperatures were 330, 400, 500 and 600°C, and the specimen variables were the nickel level, the chromium level and cold working. The high levels of helium, combined with details of the temperature history, were found to have dominated to microstructural development, sometimes overwhelming and even reversing the action of a given material variable. Microscopy examination showed that there was an unprecedented refinement of cavity microstructure at all temperatures, which accounts for the substantial alterations in both swelling and mechanical properties observed in earlier studies.

Key Words: neutron irradiation, Fe-Cr-Ni alloys, helium generation rate, temperature history, cold-work.

Introduction

The influence of helium generation rate on microstructural evolution in austenitic stainless steels during neutron irradiation has been the focus of many experimental studies over the past two decades. Recently, the focus of some studies has shifted toward experiments in which variations in the helium generation rate were put in competition with variations in other important material or environmental variables. This allows for the relative importance of helium and other variables to be assessed. Three major approaches have been used. These are isotopic tailoring (1,2), spectral tailoring (3), and comparative irradiations in two reactors with significantly different neutron spectra, and, therefore, different helium generation rates (4,5).

The latter approach has been used in a recent series of studies conducted in both EBR-II and ORR on simple ternary austenitic alloys. The principle difference in the two reactor environments was originally anticipated to be the magnitude of the helium/dpa ratio, although it was recognized that the ORR irradiation proceeded at a displacement rate roughly an order of magnitude lower than that in EBR-II. It was later discovered, however, that the ORR experiment was subjected to frequent and numerous (~600) intermittent reductions in temperature, while the EBR-II irradiation proceeded in a much more isothermal manner (5,6).

These reductions in temperature caused periodic and profuse production of small dislocation loops, known to be efficient nucleation sites for small helium bubbles. Once formed, these bubbles are sessile and remain after the loop disappears when the target temperature is re-established. This leads to an extensive and unprecedented refinement of

the cavity microstructure that is very atypical of isothermal irradiation. This, in turn, leads to much larger increases in yield strength compared to that developed during isothermal irradiation (4).

The comparative irradiation experiments in ORR and EBR-II were designed to examine the relative influence of helium/dpa ratio, alloy composition, irradiation temperature and cold-work level. Some portions of the influence of nickel content on microstructural development in competition with helium generation rate were reported earlier for annealed specimens (5). This paper presents the influence of chromium content and cold-working at 500 and 600°C, and then compares these results with those arising from variations in nickel content and helium/dpa ratio.

Experimental Details

The specimens used in the ORR experiment were in the form of standard 3 mm diameter microscopy disks with nominal composition Fe-15Cr-xNi (x=20, 25, 30, 35, 45 wt%) and Fe-yCr-35Ni (y=7.5, 15, 22 wt%). Table I presents the actual compositions. While most specimens were prepared in the annealed condition, Fe-15Ni-25Cr and Fe-15Cr-35Ni were also prepared in the 30% cold-worked condition.

The various alloys were irradiated side-by-side in sealed helium-filled subcapsules in ORR at 330, 400, 500 and 600°C. The large thermal component of the neutron spectrum in ORR caused a high generation rate of helium to occur via the two step $^{58}\text{Ni}(n,\gamma)^{59}\text{Ni}(\alpha,n)^{62}\text{Ni}$ sequential reaction. Spectral tailoring was employed during the latter portion of the experiment to produce a nearly constant generation rate of helium.(3,4)

Thus, the generation rate was roughly proportional to the nickel content of the alloys, with measured values varying from 27 to 58 appm/dpa (5).

As shown in Table 2, the dpa levels also varied as a function of nickel content and local neutron spectrum (12.2 to 14.3 dpa). This difference in dose level arises from the large recoil energy of ^{56}Fe and, therefore, varies with nickel level and neutron spectrum (7). Thus, the experiment does not allow for a complete separation of the relative influence of nickel level and helium/dpa ratio.

Density measurements were conducted on the disk specimens using a immersion technique established to be accurate to $\pm 0.16\%$ swelling for typical disks. The specimens were then electropolished using standard techniques and examined in an electron microscope operating at 200 KeV. Examination focused primarily on cavity microstructure.

Results

Figure 1 and Table 3 show the bulk swelling of Fe-15Cr-xNi alloys for both the ORR and EBR-II experiments. Significant swelling occurred in ORR only at 500°C and was dependent on nickel content.

For all nickel levels, the cavity densities of these alloys at 330°C were so high ($\geq 10^{24}\text{m}^{-3}$) that they could not be measured accurately, although it was possible to measure the average sizes. As shown in Figure 2a, the cavity density is a strong function of irradiation temperature, a relatively weak function of nickel content and is essentially insensitive to cold-working. Figure 2b shows that the average cavity size was not a strong function of nickel content except at 500°C, where it declined strongly with nickel content.

Cold-working had no significant effect at this temperature. At 600°C, the effect of cold-work on the cavity distribution was small, but somewhat variable, depending on the nickel content.

The size distribution of cavity sizes at 500°C extended down to the smallest resolvable size, while at 600°C there are no cavities at the smallest resolvable size, as shown in Figures 3a and 3b, respectively. This implies that cavities are continuously nucleating and growing at 500°C, but nucleation at the 12-14 dpa level has ceased at 600°C. The cavity size distributions are somewhat reduced in width in the cold-worked alloys at 500°C (Figure 4), suggesting that some small delay in cavity nucleation may have occurred, probably in the early stages of the irradiation. The effect of cold work is not as large as usually observed in irradiations conducted at lower helium generation rates, however.

The cavity swelling measured by microscopy (Fe-15Cr-xNi alloys) is shown in Figure 5 and agrees well with that determined by immersion density. While cold-working reduced swelling at 500°C, the effect at 600°C varies with nickel content, but was not large at any nickel level.

Swelling of Fe-35Ni-Cr alloys measured by immersion density is shown in Figure 6, increasing a small amount with increasing chromium content except at 600°C, where swelling is very low. These trends are confirmed by microscopy, as shown in Figures 7 and 8. The effect of chromium on swelling is smaller than that observed at lower helium generations rates (9).

Discussions and Conclusions

It is well known that swelling in fast reactors with low helium generation rates decreases with increasing nickel content in the range 20-45% Ni and increases with increasing chromium level (8,9). Similar but less pronounced behavior appears to occur in this study at significantly higher helium generation rates, as shown in Figure 9. The cavity distribution as a function of nickel content behaves very differently in ORR, however, compared to that in EBR-II. Not only are the cavity densities much higher in ORR, but their dependence on nickel content in general exhibits the opposite sign, reflecting the coupling between nickel content and helium generation rate.

The significantly higher density of cavities in ORR is not only a result of the higher helium generation rates (27 to 58 appm/dpa compared to 0.66 to 1.2 appm/dpa in EBR-II), but is thought to arise as a consequence of the frequent set-backs in temperature experienced in ORR. These set-backs induce dense populations of small dislocation loops that are instrumental in nucleating helium bubbles (6). Independent confirmation of such history-dependent loop nucleation was provided by Sekimura and Ishino in another similar experiment conducted in FFIF (11).

Another consequence of the higher cavity density is that the mean cavity size at lower irradiation temperatures (330, 400°C) is pushed below the critical cavity size for bubble-to-void conversion, thus suppressing the onset of void growth as shown in Figure 1. The cavity size distributions at 330 and 400°C suggest that, at all nickel levels, most cavities are most likely helium bubbles. At 500°C the cavity size distribution shifts from void-like to bubble-like character as the nickel level increases. At 45% Ni and 500°C, the cavity distribution is clearly typical of bubbles. It is important to note that the critical cavity size is also

predicted to be a function of nickel content, decreasing at higher nickel levels due to nickel's influence on vacancy diffusivity (12). Thus, the normal tendency toward suppression of swelling and void nucleation with increasing nickel content is amplified by nickel's indirect effect on helium levels and thereby on bubble nucleation.

It appears, therefore, that the combined influence of helium and temperature history dominates the microstructural evolution of Fe-Cr-Ni alloys irradiated in ORR and thereby reduces or even reverses the trends observed with cold-work, nickel content, and chromium content at lower helium generation rates.

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Figure Captions

1. Comparison of swelling determined by an immersion density technique for Fe-15Cr-xNi alloys irradiated in the ORR and EBR-II reactors.
2. Influence of irradiation temperature, nickel content and cold-work on cavity characteristics; a) cavity density and b) average cavity diameter.
3. Cavity size distributions observed in Fe-15Cr-xNi alloys irradiated at a) 500°C and b) 600°C in ORR.
4. Comparison of cavity distributions in annealed and cold-worked alloys irradiated at 500 and 600°C in ORR.
5. Swelling determined by microscopy for Fe-15Cr-xNi alloys irradiated in ORR at 500 and 600°C.
6. Comparison of swelling determined by an immersion density technique for Fe-35Ni-Cr alloys irradiated in the ORR and EBR-II reactors.
7. Average cavity characteristics and swelling in Fe-35Ni-Cr alloys irradiated at 500 and 600°C in ORR.
8. Cavity size distributions observed in Fe-35Ni-Cr alloys irradiated at 500 and 600°C in ORR.
9. Comparison of cavity densities observed in irradiations conducted in ORR and EBR-II. Data at 510°C taken from ref. 10.

Table 1 Alloys used in the EBR-II and ORR experiments^{a)}

Composition (wt%)	EBR-II	ORR	
	Tensile	TEM Disk	Tensile
Fe-19.7Ni-14.7Cr	--	SA	--
Fe-24.4Ni-14.9Cr	SA	SA,CW	SA
Fe-30.1Ni-15.1Cr	--	SA	--
Fe-34.5Ni-15.1Cr	SA	SA,CW	SA
Fe-45.3Ni-15.0Cr	SA	SA	SA
Fe-35.5Ni-7.5Cr	SA	SA	--
Fe-34.1Ni-21.7Cr	SA	SA	SA

^{a)} SA = solution annealed (954 °C/15 m/AC),
CW = 30% cold worked.

Table 2 Displacement and helium levels in the MFE-4 experiment^{a)}

Composition (wt%)	330 and 400 °C		500 and 600 °C	
	dpa	appm He	dpa	appm He
Fe-19.7Ni-14.7Cr	13.4	371	12.2	332
Fe-24.4Ni-14.9Cr	13.6	463	12.4	414
Fe-30.1Ni-15.1Cr	13.8	555	12.6	495
Fe-34.5Ni-15.1Cr	14.0	647	12.7	573
Fe-45.3Ni-15.0Cr	14.3	832	13.1	740

^{a)} These values were calculated using dosimetry calculations and measurements provided in ref. [13] for individual elements irradiated in the ORR equipment. Selected measurements on four of the ORR microscopy specimens later confirmed that these calculations were correct within 4-8% (14).

Table 3 **Density data on the effect of cold-work**

Temperature (°C)	Swelling* (%)			
	<u>Fe-25Ni-15Cr</u>		<u>Fe-35Ni-15Cr</u>	
	SA	CW	SA	CW
330	0.21	0.15	-0.05	-0.10
400	0.04	0.04	0.14	-0.06
500	1.04	0.29	0.55	-0.11
600	-0.21	0.16	-0.14	-0.07

* Negative density changes, particularly for Fe-15Cr-35Ni, can result from radiation-induced spinodal-like decomposition (15).

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