Conf-911111--21

PNL-SA--193...

DE92 006420

THE EFFECT OF HELIUM ON MICROSTRUCTURAL EVOLUTION AND MECHANICAL PROPERTIES OF Fe-Cr-Ni ALLOYS AS DETERMINED IN A SPECTRAL TAILORING EXPERIMENT

F. A. Garner
N. Sekimura
R. D. Griffin

November 1991

Presented at the
5th International Conference on
Fusion Reactor Materials
November 17-22, 1991
Clearwater, Florida

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

Pacific Northwest Laboratory
Richland, Washington 99352

THE EFFECT OF HELIUM ON MICROSTRUCTURAL EVOLUTION AND MECHANICAL PROPERTIES OF AUSTENITIC STEELS AS DETERMINED BY SPECTRAL TAILORING EXPERIMENTS

N. Sekimura
University of Tokyo
Ibaraki, Japan

F. A. Garner
Pacific Northwest Laboratory
Richland, WA

R. D. Griffin
University of Wisconsin-Madison
Madison, WI

ABSTRACT

Fe-15Cr-XNi alloys irradiated at both low (0.66 to 1.2) and very high (27 to 58) helium/dpa levels exhibit significantly different levels of strengthening due to an unprecedented refinement of cavity microstructure at the very high helium levels. When compounded with the nickel dependence of helium generation, the cavity distribution for some irradiation conditions and alloy compositions can be driven below the critical radius for bubble-to-void conversion, leading to a delay in swelling. The critical radius also appears to be dependent on the nickel level. The refinement may not have resulted from the high helium levels alone, however but also may have been influenced by differences in displacement rate and temperature history in the two experiments.

1. Introduction

A variety of experiments are underway to assess the interactive influence of helium generation rate and other important variables on the radiation-induced evolution of both microstructure and mechanical properties of simple metals and Fe-Cr-Ni austenitic alloys. One of these involves a comparison of the response of a series of Fe-15Cr-XNi and Fe-YCr-35Ni alloys in two reactors, EBR-II and ORR. The details of these experiments were published earlier by Hamilton, Okada and Garner.(1) They showed that significant differences in both swelling and mechanical properties developed between the EBR-II and ORR specimens (1). While the swelling values at higher irradiation temperatures were comparable, and exhibited the expected dependence on composition and temperature, swelling was suppressed in ORR at lower temperatures and lower nickel levels, as shown in Figure 1. Even though the swelling was less in the ORR specimens, the radiation-induced changes in yield strength were much larger in ORR than in EBR-II, as shown in Figure 2.

2. Experimental Details

The AD-1 experiment was conducted to doses of 9.5 to 11.3 dpa in EBR-II at relatively low helium/dpa ratios typical of fast reactors. The ratios range from 0.66 to 1.2 appm/dpa for nickel levels of 25 to 45%. The MFE-4 experiment was conducted at a lower displacement rate in ORR to doses of 12.2 to 14.3 dpa at calculated helium/dpa ratios ranging from 27 to 58 appm/dpa[2]. A recent measurement on the 34.5% Ni alloy from the ORR experiment shows that the actual helium levels compare well with the calculated values, being only 4-8% lower[3]. For a given irradiation temperature the displacement level was relatively independent of nickel content in EBR-II, but this was not the case in ORR, where the $^{58}Ni(n,\gamma)^{59}Ni(n,\alpha)^{56}Fe$ reaction that produces the large helium levels in this reactor also makes a measurable contribution to the displacement rate (see Table 1).

The microstructural origins of the behavior shown in Figures 1 and 2 were sought using transmission electron microscopy. This paper presents the results of the annealed Fe-15Cr-XNi (x = 20 to 45) alloys only. Analysis of the interactive effects of helium generation rate with cold working and chromium variations is still in progress and will be published later.

3. Results

The analysis effort has concentrated on specimens irradiated at 330, 400 and 500°C in ORR and at 395°C in EBR-II. The EBR-II microstructural data at 450°C were reported in an earlier paper.[4] A comparison is shown in Figure 3 of the

PDF-XChange Viewer Click to buy NOW! www.docu-track.com

swelling at 500°C in ORR as determined by both immersion density and microscopy, showing relatively good agreement. No precipitates were observed in any alloy. As can be seen in Figure 4 the major difference between the two sets of irradiations is reflected in the cavity density. Whereas the EBR-II cavity densities exhibit the usual trends, decreasing both with irradiation temperature and nickel content, the densities reached in ORR not only increase with nickel content but reach levels that are two to three orders of magnitude larger than in EBR-II. Cavity densities are in excess of 10^{23} m^{-3} at 400°C and they are even larger at 330°C. These are some of the largest ever observed in reactor irradiation studies. The cavity densities at 400°C increase with nickel level, but show saturation at higher nickel contents.

The width of the size distribution of the cavities in the ORR experiment at 500°C was observed to become progressively smaller as the nickel content increased, as shown in Figure 5. This contrasts with the behavior observed in EBR-II, where cavities are in general larger but whose sizes are relatively independent of nickel content at a given irradiation temperature.

In contrast to the behavior observed at 500°C, the cavity sizes at 330 and 400°C are very small, but increase with nickel content as shown in Figure 6. Note that the cavity sizes for the most part do not change significantly between 330 and 400°C.

Figure 7 compares loop densities as a function of nickel content from the 500°C ORR specimens with those of another comparable experiment conducted in EBR-II[5]. The dislocation structure in the ORR experiment was dominated by a high density of loops at all nickel levels, while the EBR-II experiment contained mostly tangled dislocation segments, especially at lower nickel levels.

4. <u>Discussion</u>

In the range of 400-500°C the swelling of Fe-Cr-Ni ternaries in EBR-II is known to exhibit a transient regime of ~10 dpa prior to swelling at a rate of ~1%/dpa.[6] If this trend also pertains to swelling at the lower displacement rate of ORR then these alloys would have accumulated 280-580 appm helium prior to reaching the 10 dpa level. The large decrease in swelling at lower temperatures at all nickel levels relative to that in EBR-II may reflect the impact of the very large and almost unprecedented density of cavities arising from the high helium levels.

The swelling in ORR at 500°C increases with declining nickel content as the cavity density decreases from the 10^{23} to the 10^{22} m^{-3} level, but at this temperature in EBR-II it is known that swelling also increases as the nickel level falls.[6] Note in Figure 1 that swelling levels in the 24.4Ni alloy at 500°C were comparable in the two reactors. Thus the behavior observed at 500°C reflects a lesser impact of the helium-induced refinement, while still responding to the nickel's influence on swelling via its effect on vacancy diffusivity and dislocation bias.[7-9]

It is significant that the cavity sizes at 45% Ni and 500°C, as well as at all nickel levels at 330 and 400°C, are small enough that most of them are probably helium bubbles rather than voids. At these sizes, the cavities may have been driven below the critical radius for bubble-to-void conversion.[10] Cavity densities and sizes increased with nickel content at these temperatures, but were not proportional to either the nickel level or the helium content. This suggests that the critical cavity radius for bubble-to-void conversion increases with nickel content. This experimental evidence is consistent with the results of other studies, in which a variety of nickel-dependent mechanisms of swelling behavior were proposed.[5,7-9,11] The extensive refinement of the cavity microstructure is probably the major reason for the much larger level of strengthening observed in the tensile tests on specimens irradiated in ORR. The microstructural results indicate that, at all irradiation temperatures studied, the refinement of cavities (along with a smaller refinement of loop microstructure) is sufficient to account for the higher levels of strengthening observed in the ORR experiment.

It is tempting at this point to attribute the observed refinement in ORR solely to the higher helium levels, but there appears to be other contributing factors. Hamilton and Garner have recently irradiated the 25 and 45% Ni alloys in FFTF using ^{59}Ni isotopic doping to study the effect of helium in a truly single variable manner[12,13]. In the absence of displacement rate variations, a helium/dpa ratio of ~15 appm/dpa yielded no significant variations in swelling or tensile properties at 365°C. Yet in the ORR experiment a helium/dpa ratio roughly twice this level (34 appm/dpa for the 24.4Ni alloy) caused near-total suppression of swelling at 330 and 400°C. This change in behavior is too abrupt to explain confidently in terms of helium only, and would require invoking some very sensitive threshold behavior in which a factor of only two increase could provoke such an exaggerated response.

However, there are two important differences between the manner in which the ORR and EBR-II experiments were conducted. First, the ORR experiment proceeded at roughly an order of magnitude lower displacement rate and microstructural development and its macroscopic consequences are known to be very sensitive to displacement rate [14]. If the lower displacement rate and higher helium generation rate in ORR retard the growth and infaulting of loops at lower nickel levels, as shown in Figure 7, this could cause the lower swelling observed. At higher nickel levels, however, it appears that the higher helium production rate is the main reason for loop refinement.

More recently, however, it has been shown that details of temperature history in fission reactors can have a very large impact on microstructural evolution, especially when irradiation proceeds, even for very brief periods, at lower temperatures [15-19]. The ORR reactor had a low inlet temperature and a high gamma heating rate, and thus irradiation temperatures were closely tied to reactor power level. One facet of ORR operation was the automatic step-down of the power level periodically for periods of 10-30 min, resulting in periodic temperature decreases on the order of 100-150°C. Garner, Sekimura and their coworkers recently showed experimentally that only 0.01 dpa accumulated under such conditions can cause a significant refinement of microstructure.[18,19] It may be possible that helium bubble nucleation at higher nickel levels is particularly sensitive to refinement under such conditions.

5. Conclusions

Comparison of microstructures developed in Fe-Cr-XNi alloys irradiated in EBR-II and ORR shows that very large amounts of helium can significantly strengthen alloys via their effect on refinement of cavity microstructure. In addition, this refinement may have as one of its consequences a reduction of individual cavity sizes below the critical radius of bubble-to-void conversion for some alloys and irradiation conditions. It also was found that the critical radius appears to increase with nickel content. However, there also appears to be a possibility that the refinement in ORR was assisted by significant differences in displacement rate and temperature history compared to that found in EBR-II.

ACKNOWLEDGMENTS

The ORR portion of this work was sponsored by the Japan/U.S. FFTF-MOTA Collaboration of U.S. Department Of Energy and Monbusho, the Japanese Ministry for Education, Science and Culture. The AD-1 portion was conducted under the auspices of the Northwest College and University Association for Science at Pacific Northwest Laboratory.

REFERENCES

1. M. L. Hamilton, A. Okada and F. A. Garner, J. Nucl. Mater., 179-181 (1991) 558-562.

2. L. R. Greenwood, in Fusion Reactor Materials Semiannual Progress Report DOE/ER-0313/6 (1989) pp. 29-35.

3. F. A. Garner and B. M.Oliver, in Fusion Reactor Materials Semiannual Progress Report, DOE/ER-0313/9, (1991) pp. 58-60.

4. H. R. Brager, F. A. Garner and M. L. Hamilton, J. Nucl. Mater., 133 and 134 (1985) 594-598.

5. T. Muroga, F. A. Garner and S. Ohnuki, J. Nucl. Mater., 179-181 (1991) 546-549.

6. F. A. Garner and H. R. Brager, ASTM STP 870, 1985, pp. 187-201.

7. F. A. Garner and A. S. Kumar, ASTM STP 955, pp. 289-314.

8. J. J. Hoyt and F. A. Garner, J. Nucl. Mater. 179-181 (1991) 1096-1099.

9. W. A. Coghlan and F. A. Garner, ASTM STP 870 (1985) 289.

10. G. R. Odette, et al., J. Nucl Mater., 85 and 86 (1979) 533-545, also 103 and 104 (1981) 1361-1366 and also 122 and 123 (1984) 514-519.

11. E. H. Lee and L. K. Mansur, Philos. Mag. A52 (1985) 493.

12. F. A. Garner, M. L. Hamilton, R. L. Simons and M. K. Maxon, J. Nucl. Mater., (1991) 554-557.

13. M. L. Hamilton and F. A. Garner, this conference.

14. F. A. Garner, H. L. Heinisch, R. L. Simons and F. M. Mann, Rad. Eff. Def. Sol., 113 (1990) 229-255.

15. M. Kiritani, J. Nucl. Mater., 160 (1988) 135-141.

16. M. Kiritani, I. Yoshiie, S. Kojima, Y. Satoh and K. Hamada, J. Nucl. Mater., 174 (1990) 327-351.

17. N. Yoshida, J. Nucl. Mater., 174 (1990) 220-228.

18. N. Sekimura and S. Ishino, J. Nucl. Mater., 179-181 (1991) 542-545.

19. F. A. Garner, A. M. Ermi, N. Sekimura and J. W. Newkirk, Fusion Reactor Materials Semiannual Progress Report DOE/ER-0313/11 (1992) in press.

Table 1

Displacement and Helium Levels[a] in the MFE-4 Experiment in ORR

	330 and 400°C			500 and 600°C		
Composition, wt%	dpa	He, appm	appm/dpa	dpa	He, appm	appm/dpa
Fe-19.7Ni-14.7Cr	13.4	371	27.7	12.2	332	27.2
Fe-24.4Ni-14.9Cr	13.6	463	34.0	12.4	414	33.4
Fe-30.1Ni-15.1Cr	13.8	555	40.2	12.6	495	39.3
Fe-34.5Ni-15.1Cr	14.0	647[b]	46.2	12.7	573[b]	45.1
Fe-45.3Ni-15.0Cr	14.3	832	58.2	13.1	740	56.5

(a) These values were calculated using dosimetry calculations and measurements provided in Reference 2 for individual elements.

(b) Measured values of helium at 34.5 Ni and 330, 400, 500 and 600°C were 4-8% lower, at 597 ± 4, 611 ± 7, 533 ± 3 and 552 ± 7 respectively.

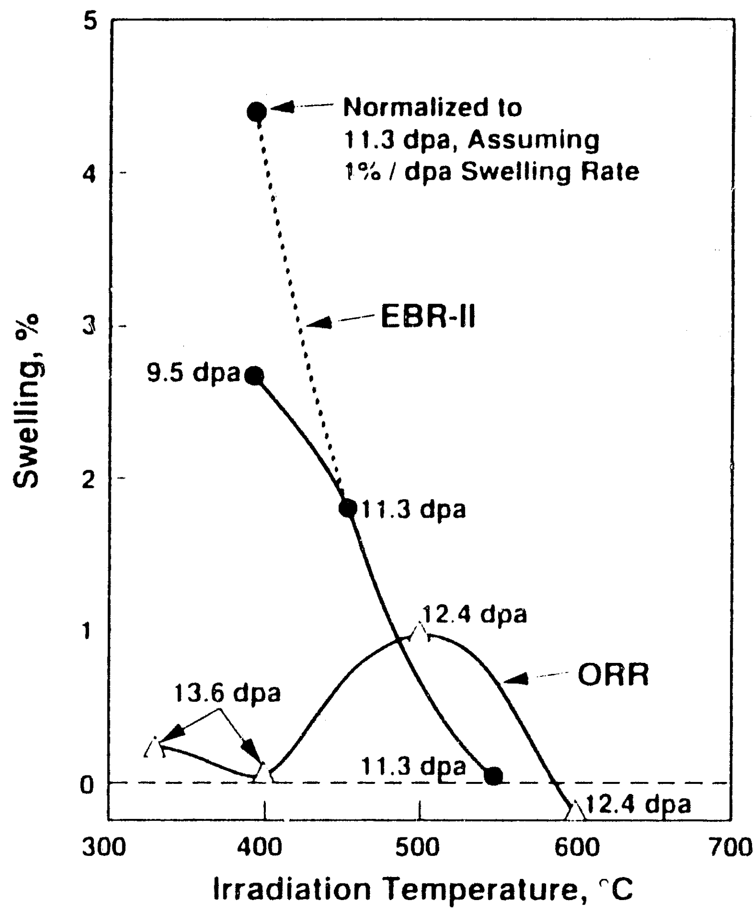

Figure 1. Comparison of swelling behavior of Fe-14.9Cr-24.4Ni in ORR and EBR-II reactors, showing strong suppression in ORR at lower irradiation temperatures[1] (displacement levels are shown beside each datum.) Similar behavior was observed at other nickel levels.

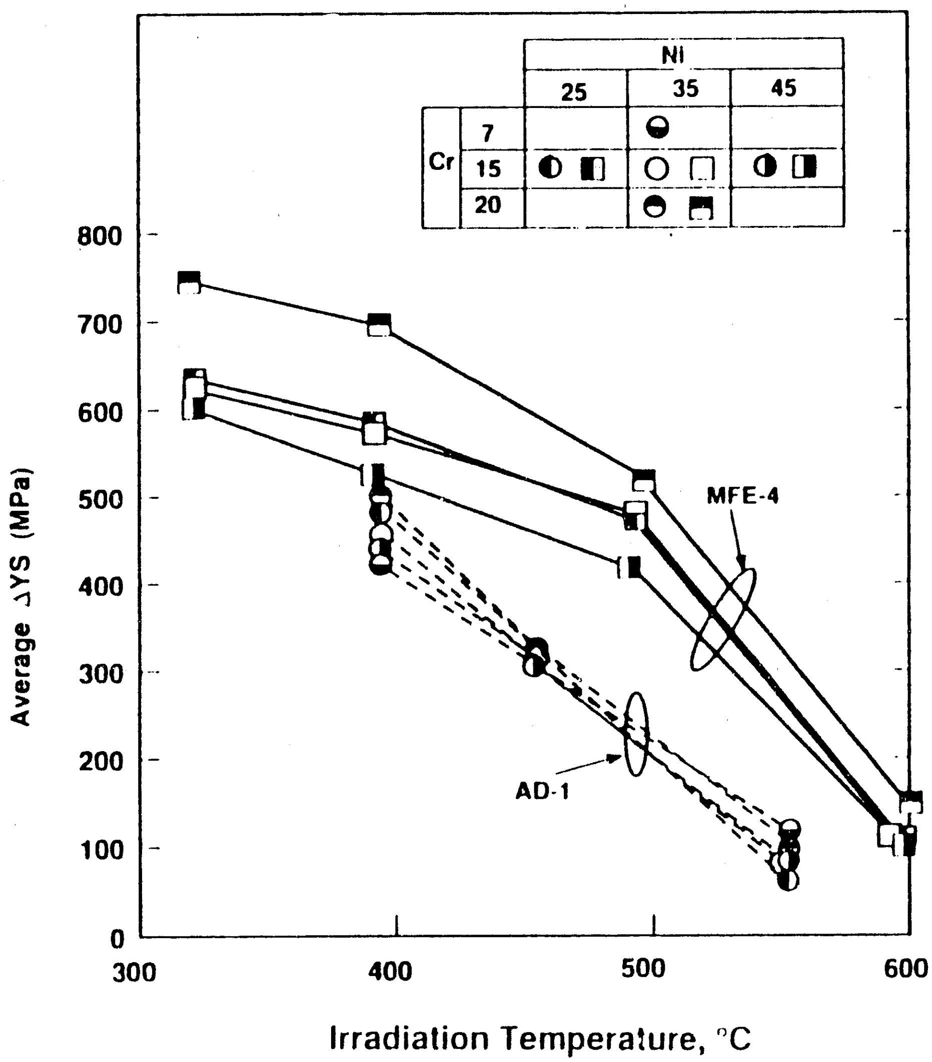

Figure 2. Comparison of radiation-induced changes in yield strengths of Fe-15Cr-XNi and Fe-YCr-35Ni alloys irradiated in the AD-1 experiment in EBR-II and the MFE-4 experiment in ORR.[1]

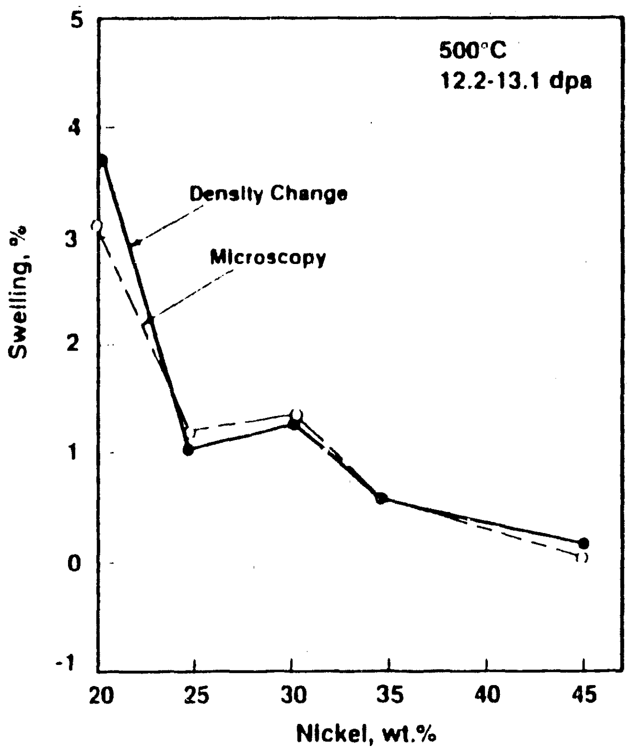

Figure 3. Excellent agreement between two different types of measurement of swelling in Fe-15Cr-XNi alloys irradiated at 500°C in ORR.

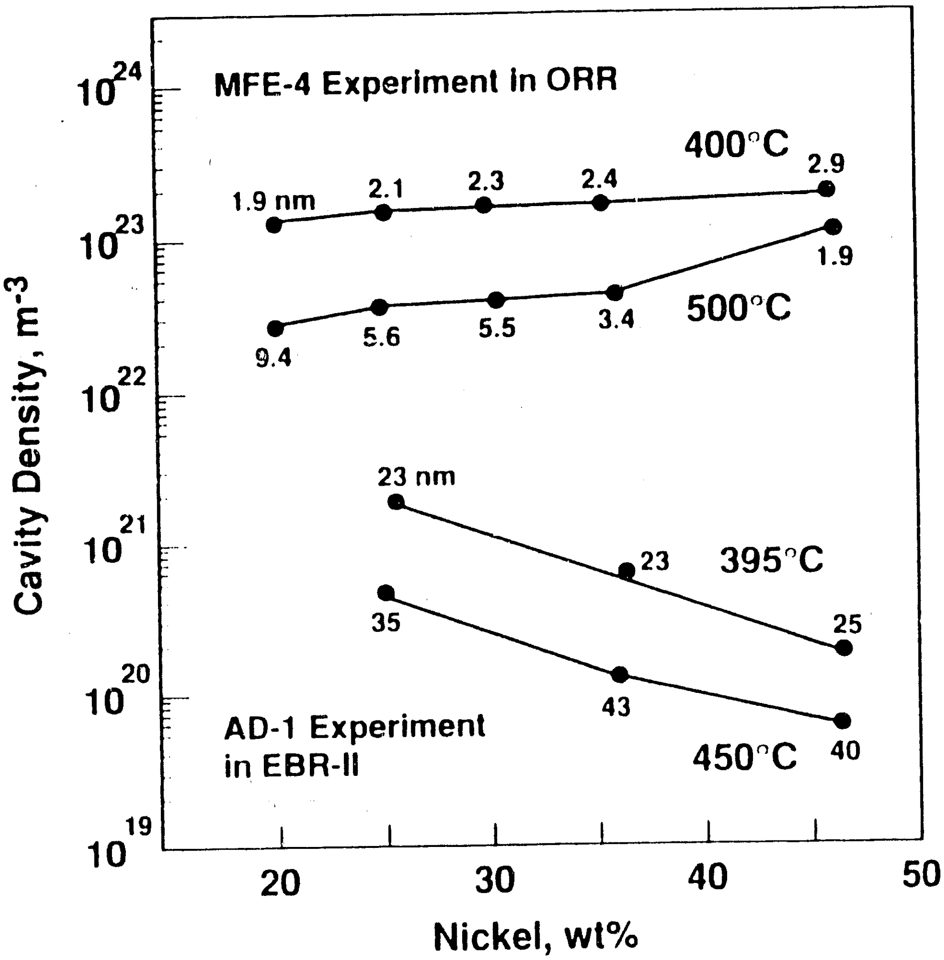

Figure 4. Cavity densities observed in the Fe-15Cr-XNi alloys irradiated in EBR-II and ORR. Mean cavity sizes are shown in nm.

PDF-XChange Viewer
Click to buy NOW!
www.docu-track.com

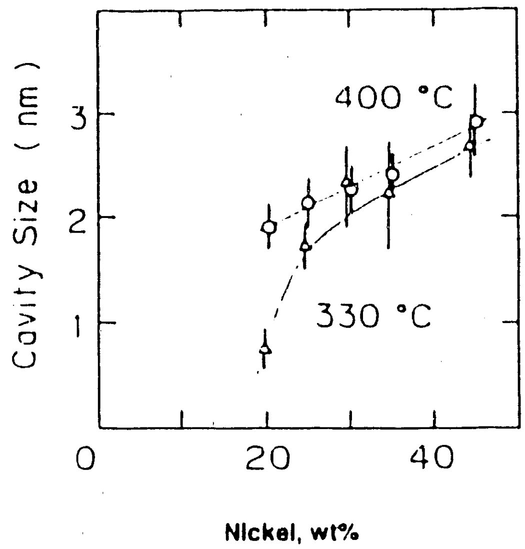

Figure 5. Cavity sizes observed in Fe-15Cr-XNi alloys irradiated in ORR at 330 and 400°C.

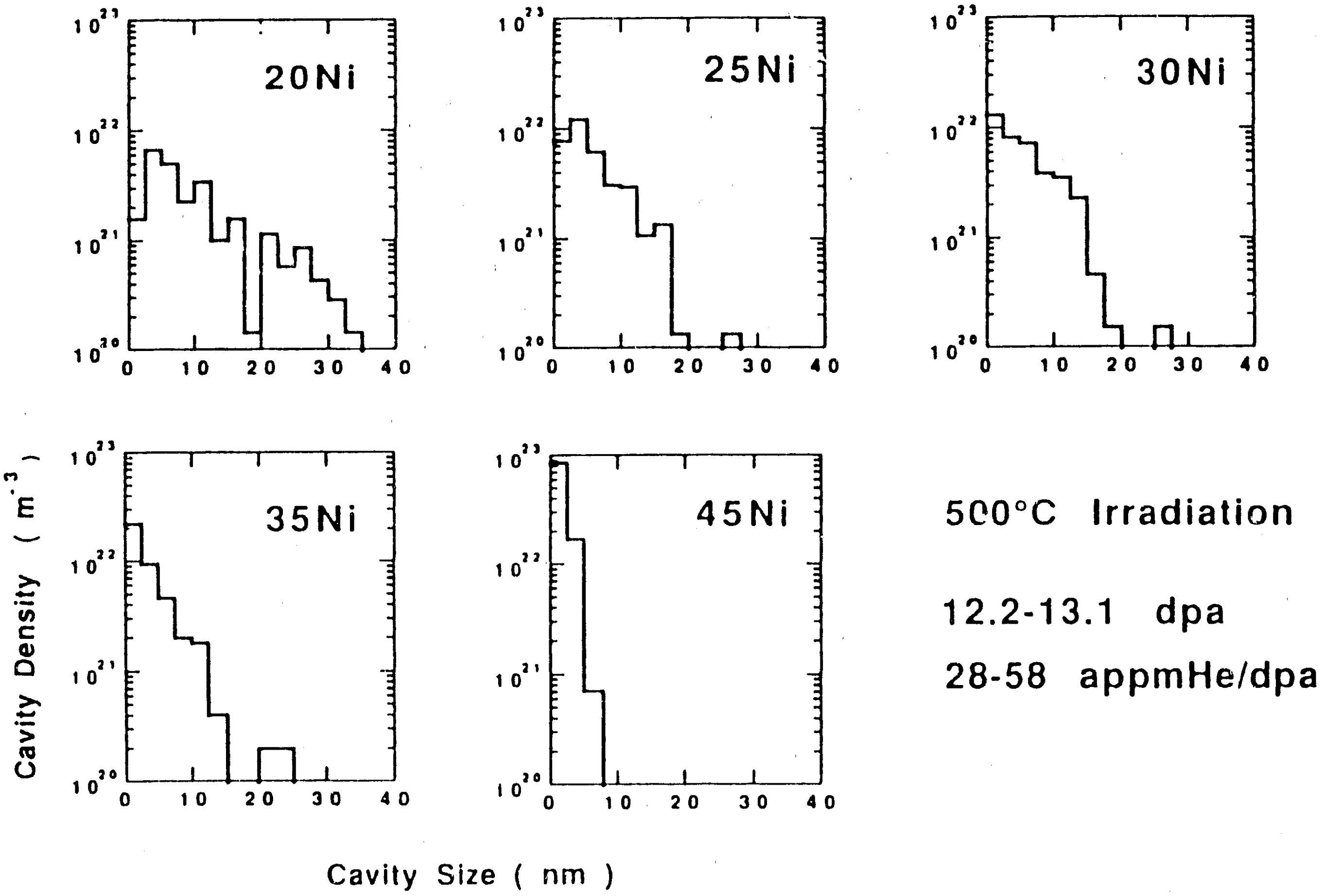

Figure 6. Cavity size distributions observed in Fe-15Cr-XNi alloys irradiated in ORR at 500°C.

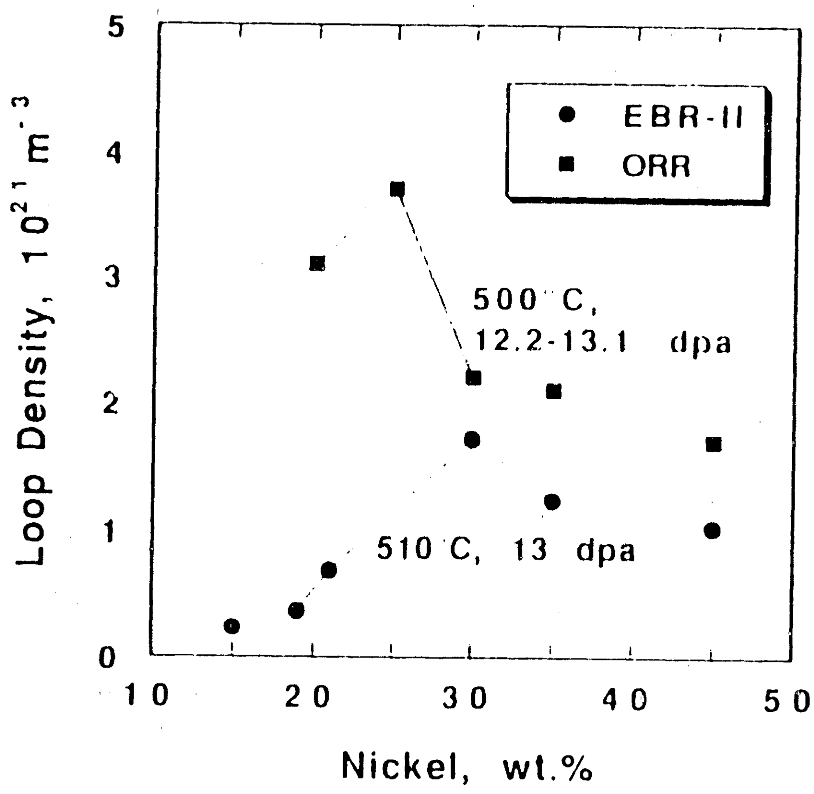

Figure 7. Comparison of loop densities observed at ~500°C in the MFE-4/ORR experiment and in the AA-7/EBR-II experiment.[5]

END

DATE
FILMED
2 / 26 / 92

PDF-XChange Viewer
Click to buy NOW!
www.docu-track.com