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THE RELATIVE INFLUENCE OF HELIUM/dpa RATIO AND OTHER VARIABLES ON NEUTRON-INDUCED SWELLING OF Fe-Ni-Cr ALLOYS AT 495 DEGREES CELCIUS AND 14 dpa

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The Relative Influence of Helium/dpa Ratio and Other Variables on Neutron-Induced Swelling of Fe-Ni-Cr Alloys at 495°C and 14 dpa

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Abstract

An isotopic tailoring experiment utilizing ^{59}Ni has been conducted in the Fast Flux Test Facility (FFTF) to study the influence of helium/dpa ratio on neutron-induced microstructural evolution. The influence of other variables, namely composition (nickel and phosphorous variations) and cold work on swelling was also examined. The alloys Fe-25Ni-15Cr, Fe-25Ni-15Cr-0.04P, and Fe-45Ni-15Cr were each irradiated in four conditions: solution annealed or 20% cold worked both with and without the ^{59}Ni additions yielding He/dpa ratios of 0.35 and 4.7 at 25Ni and 0.57 and 4.9 at 45Ni. Increases in helium were found to alter the details of microstructural evolution somewhat at 495°C, but the effect of helium was small compared to the effects of other variables studied.

KEYWORDS: Fe-Ni-Cr Alloys, Swelling, Helium, He/dpa Ratio, Nickel Level, Phosphorus, Cold Work, Microstructural Evolution

Introduction

The role of helium in void nucleation and growth is of concern for fusion applications where helium generation is higher than in fast and thermal fission reactors. While the potential for significant helium effects has long been recognized, a method to investigate the effect without introducing other variables has only recently become available.

In fusion reactor systems, the helium/dpa ratio will remain relatively constant during operation. In the present approach, the ^{59}Ni isotope is used as a dopant to produce a nearly constant helium/dpa ratio during irradiation in a fast fission spectrum [1]. This is usually impossible since ^{59}Ni is only produced by a (n,γ) reaction with ^{58}Ni. The subsequent $^{59}Ni(n,\alpha)^{56}Fe$ reaction does not reach significant proportions until sufficient ^{58}Ni is converted to ^{59}Ni. Thus the helium/dpa ratio increases with irradiation and is very low at the critical, initial stages of exposure. This experiment used ^{59}Ni isotopically tailored alloys to study the effects of helium and also explored the separate and synergistic effects of nickel, phosphorus and cold work on swelling and microstructural development.

Experimental Details

Microscopy discs were fabricated from the Fe-15Cr-XNi alloys shown in Table 1. Half were fabricated using nickel enriched in ^{59}Ni, the other half with natural nickel. Two nickel levels were used, 25 and 45 wt%, and 0.04 wt% phosphorus was also added as a variant in the 25% nickel alloy. Specimens were prepared in both solution annealed and 20% cold worked states.

These alloys were irradiated to 14 dpa at ~3 x 10^{-6} dpa/s in the Fast Flux Test Facility (FFTF) at 495±5°C. One temperature excursion to 600°C for

10 minutes was experienced after ~2 dpa had been accrued. Further details are given elsewhere [2,3].

Immersion density measurements, accurate to ±0.16% change in volume, were made and the averages of three separate measurements on each specimen are reported. Microstructural information was obtained using a JEM JEOL 100CX scanning transmission electron microscope (STEM) operated at 100 kV. Post-irradiation measurement established He/dpa ratios of 0.35 and 4.7 for the low and high helium cases of Fe-25Ni-15Cr and 0.57 and 4.9 for Fe-45Ni-15Cr. The increases at 45Ni are due to the (n,α) contribution of natural nickel.

Results

As shown in Figure 1, the void swelling data determined by microscopy follow the same trends as the immersion density data, but are consistently lower, apart from the cases where density measurements indicated little volume change or densification. The persistent difference between microscopy and density measurements of swelling indicates that some other non-void contribution to volume change exists. As yet, this other contribution has not been identified.

The data clearly show the effects of all of the major experimental variables. Higher nickel content and cold work both decrease swelling in a consistent manner. Phosphorus doping also decreases swelling. The effect of higher helium was, in all cases but one (solution annealed 25Ni with no P), to enhance the swelling slightly. The densification shown in the immersion density measurements is within the range of uncertainty of the measurement technique for the 45Ni case, but a substantial net densification is clearly evident in the 25Ni-0.04P case.

Microstructural analysis showed that voids were present in every case, even where immersion density indicated a net densification.

Figures 2 and 3 show a comparison of the swelling, void number densities and average void sizes for the solution annealed and cold worked cases, respectively. Figure 2 indicates that the strongest effect in inhibiting swelling in the solution annealed alloys is the nickel content, followed by the addition of phosphorus. The effect of higher helium is secondary to alloying effects at these irradiation conditions. The one anomaly in the data is the high helium 25Ni alloy without phosphorus. This alloy is the only high helium alloy which showed reduced swelling over its low helium counterpart.

Figure 3 shows a monotonic increase in both the void number densities and average void sizes with swelling. (The single exception is that the low helium phosphorus-free 25Ni alloy has nearly the same number density as the high helium 25Ni alloy with phosphorus.) Again, high nickel content correlates with reduced swelling. In all of the cases here, the high helium alloys swelled slightly more than their counterparts. This was due both to higher void number densities and to larger average void sizes.

Discussion

Both the immersion density and the void data indicated in all cases but one that the higher helium content results in larger values of swelling. However, the absolute values of the differences are relatively small.

Our results agree with modeling studies of the role of helium in cavity formation [4,5]. These models indicate that helium can reduce the critical diameter for void formation. Nickel was found to be the predominant variable controlling swelling in this experiment. This behavior is well known [6,7] and can also be related to an argument for larger critical radius [8,9].

The cold worked specimens showed lower swelling than the comparable solution annealed material conditions, presumably due to a higher initial point defect sink density. In the two alloy conditions (25 and 45% Ni) without phosphorus, the cold-work dislocation structure had largely disappeared during irradiation. A dislocation and loop structure similar to the solution annealed material was present. Figure 4 shows the relative contributions of line dislocations and loops to the total dislocation density. The materials irradiated in the cold worked condition possess similar dislocation densities to those irradiated in the annealed state. However, the loop population generally represents a slightly larger fraction of the total for the cold worked alloys, indicating that the stable dislocation structure may still be evolving. It has been shown earlier that the saturation loop and dislocation densities should reach values which are independent of starting levels [4,10].

The lack of recovery of the cold work dislocation density and the retardation of swelling in phosphorus-containing alloys have been observed in other studies [11]. Refinement of the loop structures in annealed specimens containing phosphorus has also been reported [12,13]. The role of phosphorus has been shown in previous studies to have varying effects on swelling depending on irradiation conditions, phosphorus levels, and whether or not precipitates form [6]. In the present study, no phosphide precipitates were found. This is consistent with another recent study [14] at 510°C and 13.2 dpa where phosphides were found to form only at 0.055% phosphorus and above. The results of this study are also in good agreement with a similar study at 365 and 600°C [15].

Conclusions

A simple model alloy experiment designed to study the separate and synergistic effects of helium/dpa ratio, nickel level, cold work, and phosphorus on microstructural evolution at 14 dpa and 495°C used ^{59}Ni isotopic tailoring to avoid variations in other important variables. While helium affected the details of the microstructure, its influence was secondary to that of the other variables studied.

Acknowledgments

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Table 1. Composition of Alloys*

Element	Weight Percent					
Fe	60.0	60.00	60.0	60.0	40.0	40.0
Ni, total	25.0	25.00	25.0	25.0	45.0	45.0
Cr	15.0	15.0	15.0	15.0	15.0	15.0
P	0.0	0.0	0.04	0.04	0.0	0.0
^{59}Ni	0.434	0.0	0.412	0.0	0.420	0.0

*all alloys irradiated in both the solution annealed (1030°C for 30 min.) and 20% cold worked conditions

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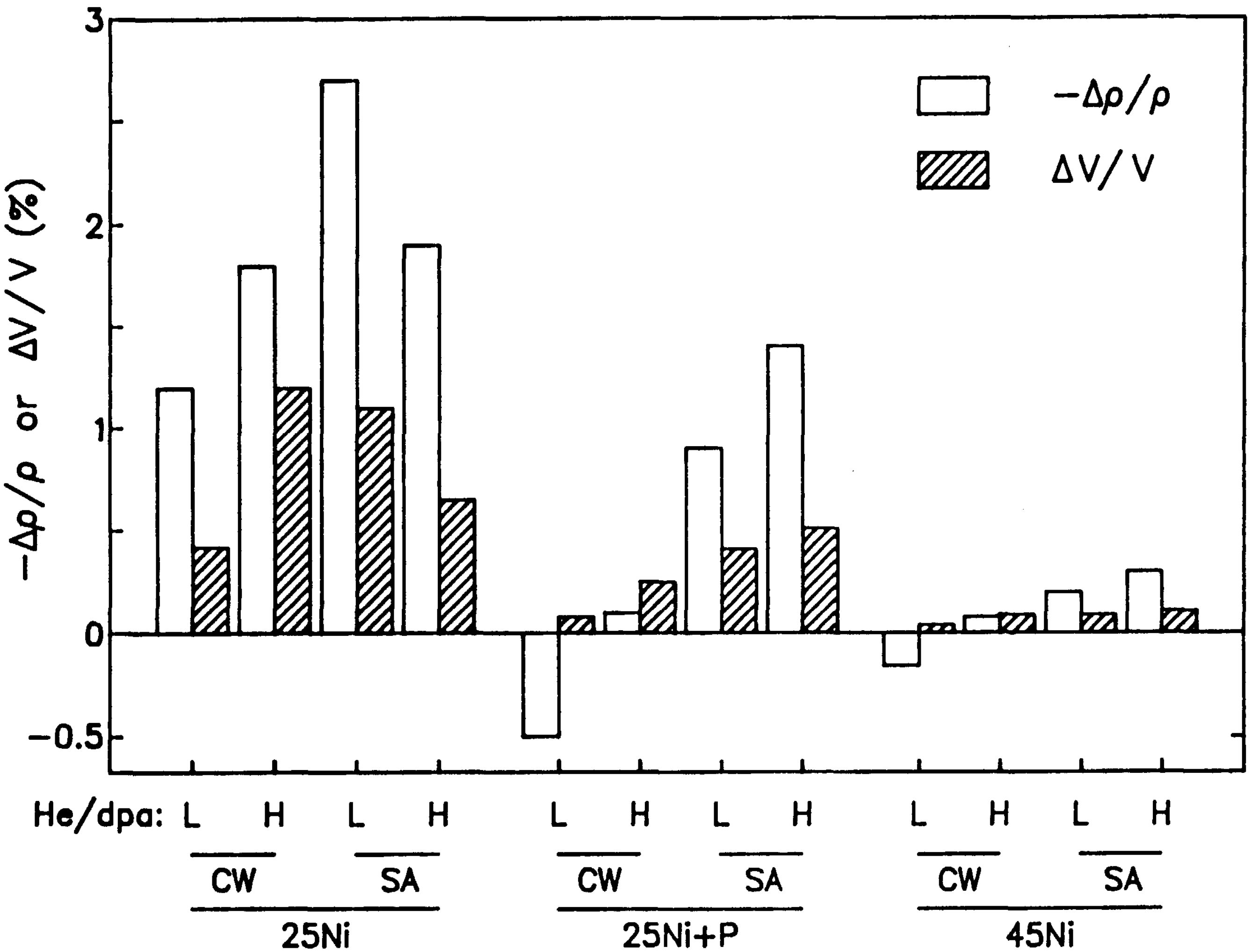
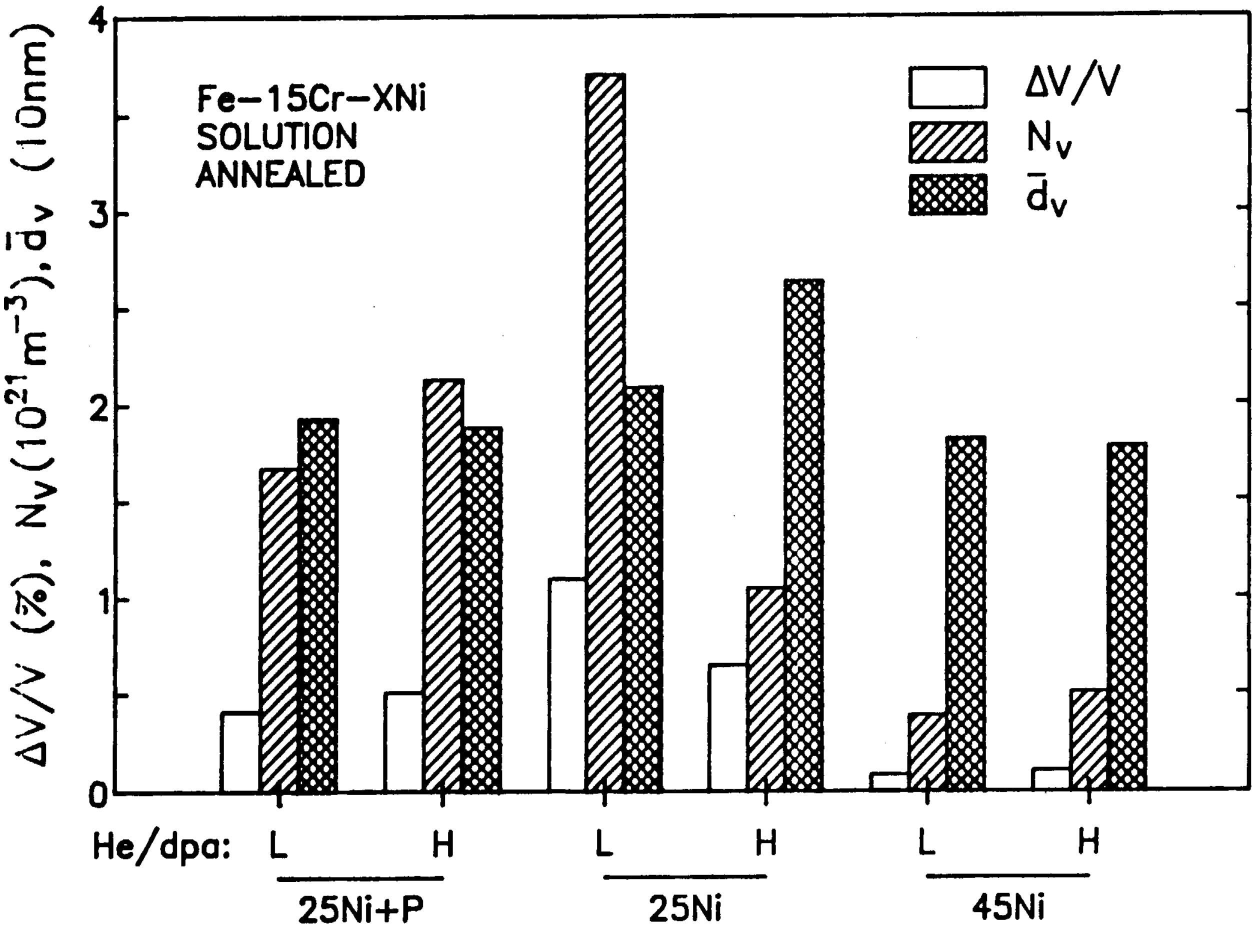
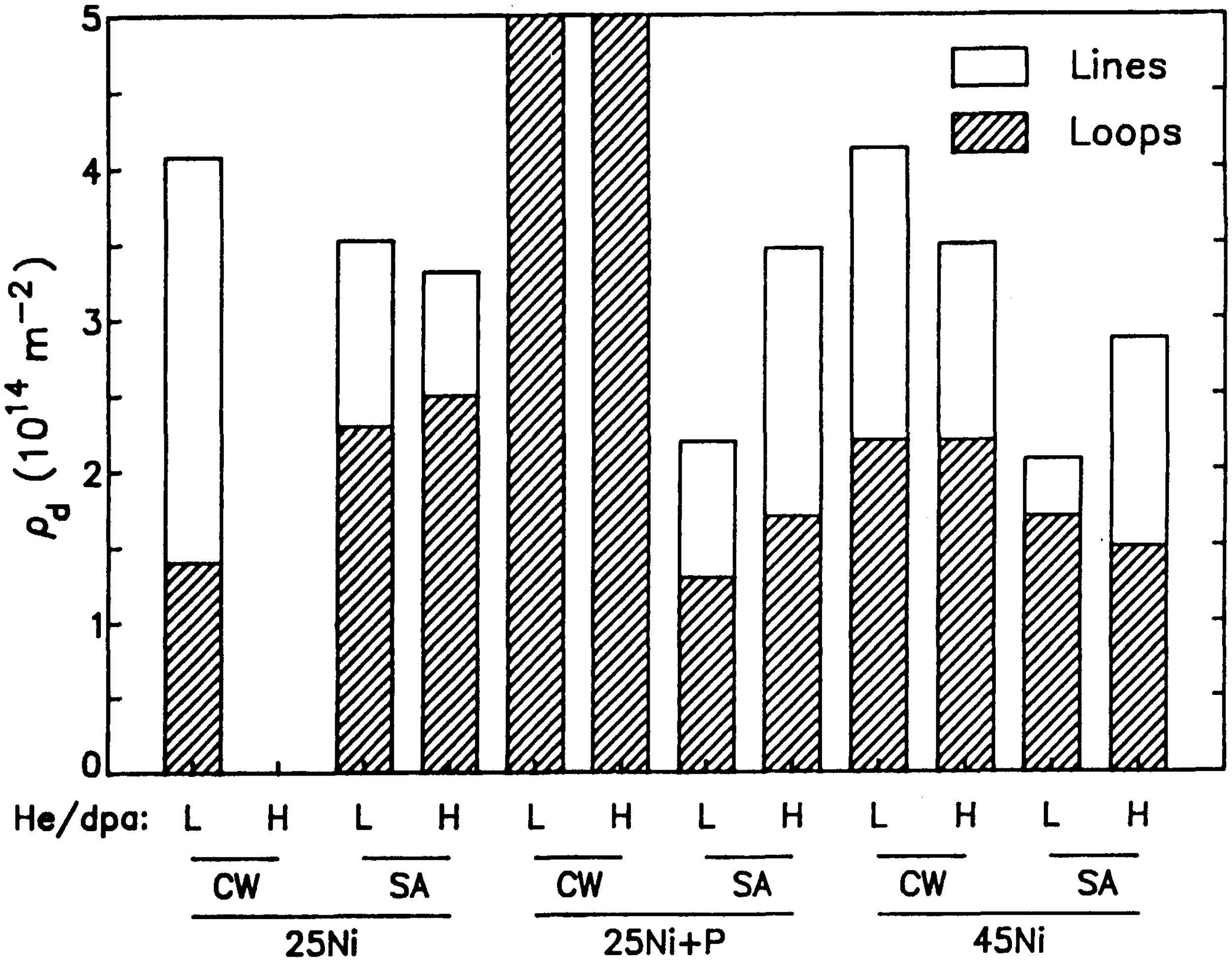


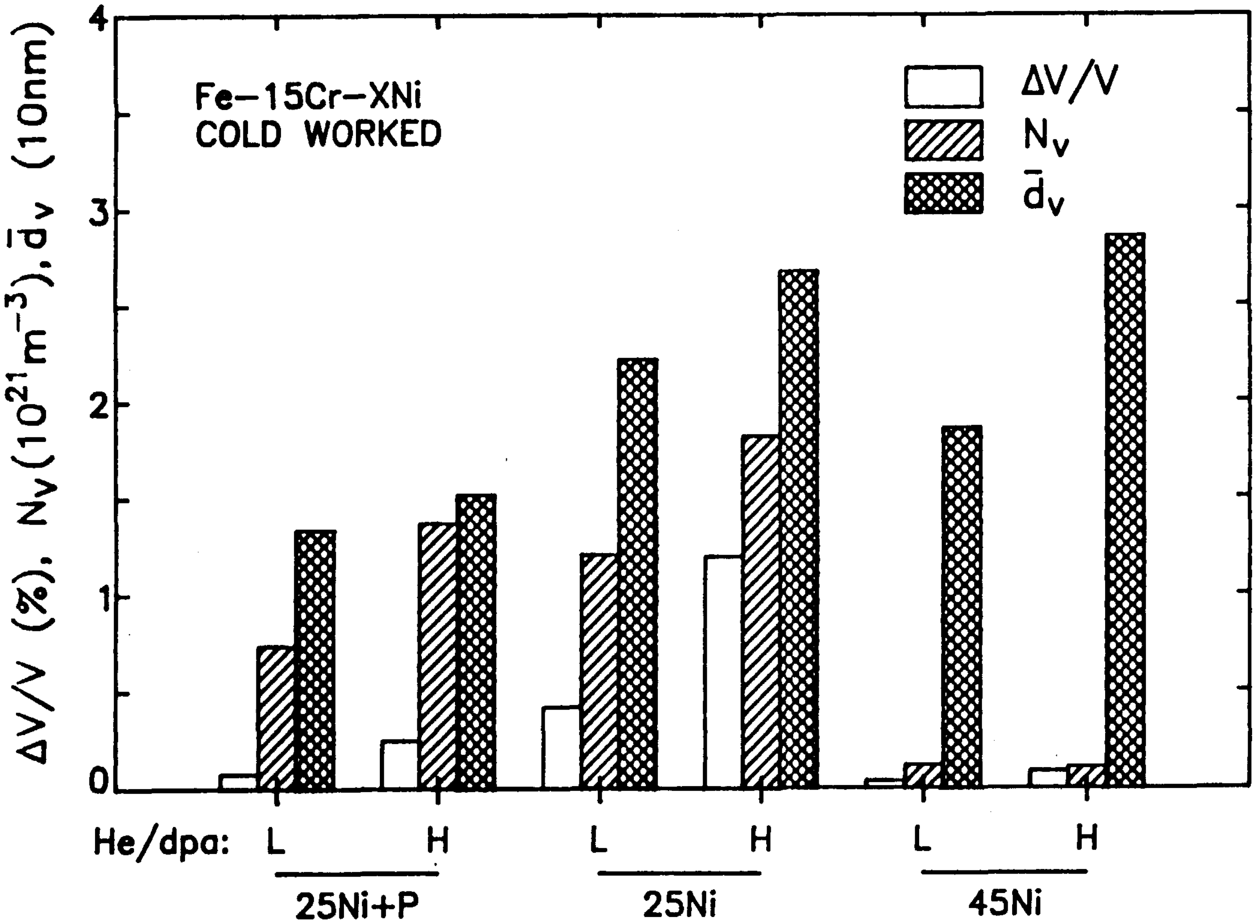
3
2
1
0
-0.5
$-\Delta\rho/\rho$ or $\Delta V/V$ (%)
$-\Delta\rho/\rho$
$\Delta V/V$
He/dpa: L H L H L H L H L H L H
CW SA CW SA CW SA
25Ni
25Ni+P
45Ni



Fe–15Cr–XNi
SOLUTION
ANNEALED
ΔV/V
N_v
d̄_v
ΔV/V (%), N_v (10^21 m^-3), d̄_v (10nm)
0
1
2
3
4
He/dpa:
L
H
L
H
L
H
25Ni+P
25Ni
45Ni



5
4
3
2
1
0
ρ_d (10^{14} m^{-2})
Lines
Loops
He/dpa:
L
H
L
H
L
H
L
H
L
H
L
H
CW
SA
CW
SA
CW
SA
25Ni
25Ni+P
45Ni



Fe−15Cr−XNi
COLD WORKED
ΔV/V
N_v
$\bar{d}_v$
ΔV/V (%), N_v (10^{21} m^{-3}), $\bar{d}_v$ (10nm)
0
1
2
3
4
He/dpa: L H L H L H
25Ni+P
25Ni
45Ni