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By

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ABSTRACT

Ni-Be alloys containing 0.1, 0.3, and 0.7 at.% beryllium have been irradiated with 3.2-MeV $^{58}Ni^{+}$ ions at 425, 525, and 625°C. Dose levels between 1 and 30 dpa have been obtained at a calculated dose rate of $\sim 3 \times 10^{-3}$ dpa/sec. Preliminary transmission-electron microscopy examination of samples irradiated to ~17 dpa show that at 525 and 625°C the void volume fraction in the Ni-0.1 at. % Be alloy is quite similar to that in pure Ni. However, between 0.1 and 0.7 at. % Be, the void volume fraction decreases by a factor of ~40 at 525°C and ~3 at 625°C.

INTRODUCTION

It has been recognized for some time that the fraction of vacancies and interstitials mutually annihilated by recombination can be increased, if substitutional solute atoms strongly trap self-interstitial atoms. As originally suggested by Harkness and Li,[1] this would provide a means of limiting void swelling, since fewer excess interstitials and vacancies would exist for the formation of dislocation loops and voids. However, to be effective, the solute-interstitial binding energy must be comparable to or greater than the vacancy migration energy.[2] Consequently, Blewitt and co-workers[3] made a search for doping elements in Ni that would decrease stage I recovery and increase stage III recovery, since this implies a

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solute-interstitial binding energy of the necessary magnitude. Both undersized and oversized solutes were investigated, and it was found that Be, which has the smallest atomic volume of all the metals, was the only dopant which caused a substantial decrease in stage I recovery and a corresponding increase in stage III recovery.

To demonstrate the swelling suppression predicted for Be-doped Ni, Ryan[4] irradiated pure Ni and Ni-1 at. % Be alloys at 520°C with $^{58}Ni^{+}$ ions at a dose rate of $\sim 4 \times 10^{-3}$ dpa/sec. Ryan found that at 45 dpa the void volume fraction in the Ni-1 at. % Be sample was ∿50 times less than in pure Ni. However, a large number density of plate-shape precipitates was also observed, which introduced an ambiguity as to whether the swelling suppression was associated with the presence of the precipitates or to enhanced recombination due to the trapping of self-interstitials by the Be atoms in solution. The appearance of precipitates was unexpected, since 1 at. % Be is below the solubility limit of ∿2.5 at. % Be at 520°C. A second series of experiments using lower Be concentrations was therefore initiated in an attempt to minimize the formation of precipitates.

PRELIMINARY RESULTS

Alloys doped with 0.1, 0.3, and 0.7 at. % Be were irradiated with 3.2-MeV $^{58}Ni^{+}$ ions at 425, 525, and 625°C. Dose levels between 1 and 30 dpa were obtained using a dose rate of $\sim 3 \times 10^{-3}$ dpa/sec. In these experiments, the specimens were covered with 200-mesh molybdenum grids to allow, during subsequent postirradiation examinations, a direct comparison of irradiated and unirradiated areas that have otherwise identical histories within the same electron microscope specimen.

Postirradiation examination of the bombarded specimens by transmission-electron microscopy are still in progress. To date, only those specimens irradiated to ∿17 dpa have been analyzed. Nevertheless, several points of interest have emerged from these examinations. The first is that decomposition of these dilute and normally single-phase solid solutions could not be suppressed at any of the irradiation temperatures investigated. The second point is that in all cases decomposition occurs only in the irradiated areas. An example illustrating this behavior is shown in Fig. 1 for the case of a Ni-0.1 at. % Be alloy irradiated at 525°C. The absence of precipitates in the unirradiated areas clearly indicates that decomposition of these alloys is a radiation-induced phenomenon and is not the result of the thermal history associated with the irradiation.

Figure 2 shows the void microstructures that develop after 17 dpa as a function of temperature and Be concentration. The corresponding swelling parameters are summarized in Table 1. For purposes of comparison, Ryan's 525 and 600°C data for pure Ni are also included.

Table 1. Comparison of Void Formation in Ni-Be Alloys Irradiated to 17 dpa at 525 and 625°C

	Ni*	0.1 Be	0.3 Be	0.7 Be
Mean Dia. (A)	310	442	210	220
Void Conc. (cm^{-3})	14×10^{14}	8×10^{14}	6×10^{14}	2×10^{14}
Vol. Frac. (%)	3	4	0.4	0.1
Mean Dia. (A)	575	587	457	519
Void Conc. (cm^{-3})	3×10^{14}	6×10^{14}	5×10^{14}	1×10^{14}
Vol. Frac. (%)	7	7	4	2

*Values interpolated from data at 525 and 600°C. Ryan[4]

The low-temperature form of the radiation-induced microstructure observed at 425°C was characterized by a high density of loop-like defects that are aligned along <100> directions. Selected area diffraction patterns exhibit weak precipitate reflections as well as streaking of the matrix reflections, which are characteristic of Frank loops. The

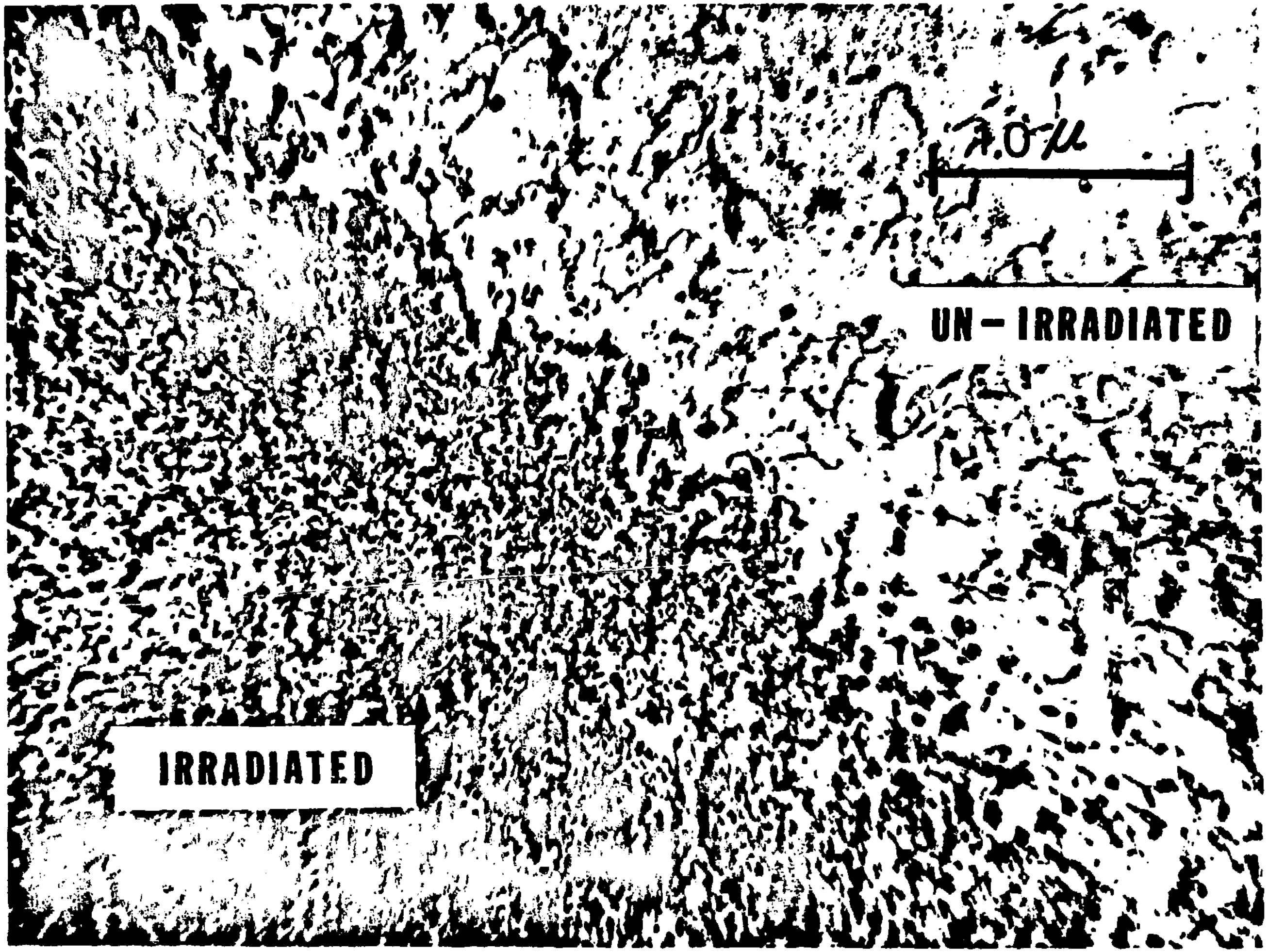

Fig. 1. Electron micrograph showing irradiated and unirradiated areas in a Ni-0.1 at. % Be alloy irradiated with 3.2-MeV $^{58}Ni^{+}$ ions at 525°C to ~17 dpa.

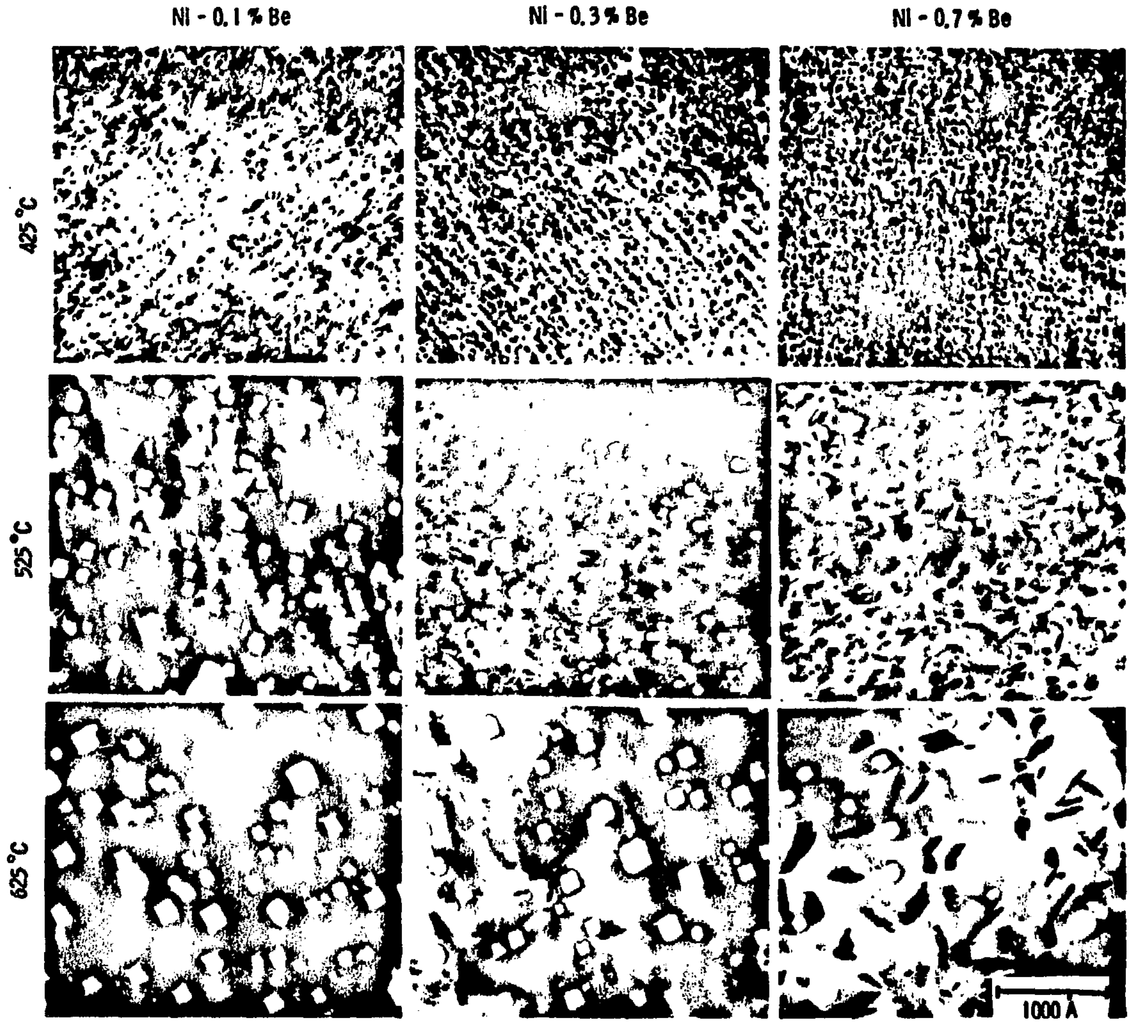

Fig. 2. Void microstructure in Ni-Be alloys irradiated to ∿17 dpa.

aligned defect structure may therefore be composed of loops as well as precipitates. However, more extensive selective dark-field imaging will be required before firm conclusions can be reached. Also, no voids were observed in any of the alloys irradiated at 425°C.

At the higher temperatures, the precipitates do not show any tendency toward alignment and appear to be randomly distributed. The precipitate distribution occurs on a much coarser scale at 625°C than at 525°C, but at both temperatures the density of the precipitates and their average size increases and decreases, respectively, on going from 0.1 to 0.7 at. % Be.

Taking into account the slightly different temperatures and dose levels used by Ryan, Table 1 shows that at 525 and 625°C the void volume fraction in the Ni-0.1 at. % Be alloy is quite similar to that in pure Ni. This indicates that Be additions >0.1 at. % Be are required to obtain significant reduction in swelling.

The last point of interest is that at 525°C the void volume fraction decreases by a factor of ∿40 on going from 0.1 to 0.7 at. % Be but only by a factor of ∿3 at 625°C. Consequently, Be doping appears to be much more effective at 525°C than at 625°C in suppressing void formation in Ni. However, since the final precipitate distribution is sensitive to both temperature and Be concentration, the question arises as to whether the suppression is the result of enhanced recombination induced by solute-interstitial trapping effects or to variations in the precipitate distribution.

DISCUSSION

Although the limited amount of data analyzed to date prevents an unambiguous interpretation of the role played by the precipitates in the suppression of swelling, it has raised an equally important question as to why phase decomposition occurs at all in these dilute solid solution alloys. Since the Be levels investigated are well below the solubility limits at the irradiation temperatures used in this study (~2, 2.5, and 4 at. % Be at 425, 525, and 625°C, respectively), the formation of precipitates cannot be solely due to radiation-enhanced diffusion.

We would like to speculate that precipitation is induced by the interstitial-solute drag mechanism recently proposed by Okamoto and Wiedersich.[5] They noted that, in a number of irradiated metals and alloys, undersized and oversized solutes tend to be enriched and depleted, respectively, around point-defect sinks. To account for this correlation, they suggested that undersized solutes interact preferentially with self-interstitial atoms and undergo long-range migration due to the formation of <100> mixed dumbbells. Consequently, undersized solutes tend to be dragged toward defect sinks by the interstitial flux, whereas oversized solutes tend to be depleted around defect sinks by the vacancy flux. This mechanism is believed to play an important role in the formation of precipitates on sinks such as voids and Frank loops in irradiated austenitic stainless steels doped with silicon.[5]

Frank loops are frequently observed to form ordered arrays, particularly at lower temperatures. The segregation of Be to Frank loops would therefore be consistent with the aligned precipitate structure observed at 425°C in the Ni-Be alloys. The decrease in precipitate density and increase in precipitate size with an increase in temperature is also consistent with the behavior expected of Frank loops. Furthermore, the recent Johnson-Lam[6] time-dependent, rate formulation of the solute drag mechanism predicts quite large segregation effects when the solute-interstitial binding energy is large, as is the case with Be in Ni. Moreover, for dose rates comparable to those used in this study, i.e., $\sim 10^{-3}$ dpa/sec, the theory predicts significant segregation to sinks between 0.2 T_m and 0.6 T_m, with maximum segregation occurring at $\sim$0.45 T_m. For Ni, this would correspond to 525°C where Be doping was observed to be most effective in suppressing void formation. Since the temperature dependence of the impurity-trapping effect is reflected in the segregation phenomenon, the optimum temperature at which the vacancy-interstitial recombination due to trapping of self-interstitials is maximized should coincide with the "peak" segregation temperature.[7] Therefore, our results suggest that trapping of self-interstitials by Be atoms in the matrix contributes, at least in part, to the reduction in void swelling.

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REFERENCES

1. S. D. Harkness and Che-Yu Li, Radiation-Induced Voids in Metals and Alloys, USAEC Symposium Series, CONF-7010601 (1972), p. 798.
2. F. A. Smidt, Jr., J. A. Sprague, J. E. Westmoreland, and P. R. Malmberg, J. Nucl. Mater. 18, 302 (1972).
3. T. H. Blewitt, C. Sartain, T. L. Scott, and R. A. Conner, Jr., Proc. of the Am. Phys. Soc. Mtg., Denver, Colorado, March 31-April 3, 1975.
4. T. D. Ryan, Ph.D. Thesis, University of Michigan, 1975.
5. P. R. Okamoto and H. Wiedersich, J. Nucl. Mater. 53, 336 (1974).
6. R. A. Johnson and N. Q. Lam, to be published.
7. N. Q. Lam, private communication.