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THE INFLUENCE OF RADIATION-INDUCED SEGREGATION
ON DUCTILITY OF A NICKEL-SILICON ALLOY*

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Flat tensile specimens 60 μm thick of Ni-8 at. % Si were irradiated to bulk damage levels of 0.1 to 0.3 dpa with either 7 MeV protons or 28 MeV alpha particles at 750 K. The alpha bombarded specimens incurred 750 at. ppm He per 0.1 dpa in the course of their damage-generating irradiation. Radiation-induced silicon segregation gave rise to Ni_3Si layers at internal and external surfaces. Postirradiation tensile tests conducted either at 300 K or 720 K revealed fully ductile (chisel-edged) transgranular fracture profiles. There were no significant differences between the proton-bombarded specimens and the unbombarded controls, both exhibiting >25% total elongations, while the alpha-bombarded specimens showed ductile fractures with somewhat lower (17-18%) elongation values probably due to hardening caused by small helium bubbles. Certain specimens that were preimplanted with 250-1000 at. ppm He at 970 K to encourage intergranular failure and expose grain boundaries did fail intergranularly. It is concluded that radiation-induced silicon segregation does not cause intrinsic embrittlement.

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BACKGROUND

Radiation-Induced Segregation (RIS) is a phenomenon of increasing significance because it threatens to undo some of the hard-won gains in resistance to radiation damage obtained from increasingly sophisticated, "finely-tuned" alloys. When RIS is taking place, the alloy matrix may be depleted of critical constituents while any coarse precipitates that form can serve as vacancy collection points and promote swelling. If the matrix is thereby hardened while segregation or precipitation occurs at the grain boundaries, the material could be gravely embrittled. Neutron irradiations of austenitic superalloys such as PE-16 followed by tensile testing at the irradiation temperature T_{irr} or particularly at about one hundred degrees above T_{irr} have produced major reductions in ductility [1-4]. Microstructural examination of such specimens suggested that the brittle intergranular fractures were caused by cleavage of RIS-generated grain boundary γ' phase [2,4].

Elements vary in their propensity to segregate under irradiation. One of the most active species is silicon which, being undersized in a matrix of nickel or iron atoms, couples readily to the interstitial flux and accumulates at interstitial sinks such as dislocation loops, internal interfaces and the external surface [5]. Various experiments have been conducted using techniques such as Auger Electron Spectroscopy [6,7], Secondary Ion Mass Spectrometry [8], or Rutherford Backscattering Spectroscopy [9] to analyze the extent of silicon segregation to the external surfaces in heavy-ion bombarded alloys of nickel with 1 to 12 at. % Si.

The consistent findings have been that silicon is enriched to the point of developing an Ni_3Si (γ' structure) surface layer at doses well below 1 dpa. The effects of these active silicon segregation processes on mechanical behavior have yet to be explored, except for several studies of irradiation creep in pure nickel and nickel with 4-5 at. % Si [10,11]. The minor effects found were attributed to silicon-enhanced dislocation loop/ γ' nucleation within the grains. The present work endeavors to find out whether radiation-induced silicon segregation does or does not promote embrittlement in an alloy of Ni-8 at. % Si. Characterization of the nature of the segregation to internal interfaces using analytical Transmission Electron Microscopy (TEM) and sputter depth profiling in a Scanning Auger Microprobe (SAM) are published elsewhere [12,13].

EXPERIMENT

The material used was a binary alloy of nickel with 8 atomic per cent silicon (about 4 wt % Si) prepared from 99.99% Ni and 99.999% Si stock. At this concentration silicon is completely soluble in nickel so no precipitation is observed after homogenization. The ingot was cold rolled to a thickness of 60 μm with intermediate anneals at 1075 K in argon. Flat tensile specimens were then spark cut to dimensions of 8 mm long $\times$ 2 mm wide gage section and 8 mm square grip regions. The final anneal, 500 s at 1075 K in 10^{-4} Pa vacuum, resulted in an annealed structure with grains ranging in size from small (<10 μm , often in clusters) to large, 40 to 60 μm .

The tensile specimens were irradiated with light ions from the compact cyclotron at KFA Jülich. The bombardment chamber is usually used for "in-beam" creep testing and has been described in detail previously [14,15]. Three different bombardment modes were used to produce displacement damage: a) 7 MeV protons passing through the specimen, b) 7 MeV protons degraded and stopped homogeneously within the specimen, and c) degraded 28 MeV alpha particles stopped homogeneously within the specimen. The dose rates for the proton and alpha particle irradiations were 3×10^{-6} dpa/s (3×10^{14} protons/cm².s) and 6×10^{-6} dpa/s (2×10^{13} He⁺⁺/cm².s) respectively and the accumulated doses varied from 0.1 to 0.3 dpa. In the use of mode c) about 750 at. ppm helium per 0.1 dpa was introduced in conjunction with the displacement damage, which may affect the mechanical properties because helium is known to induce grain boundary embrittlement [16,17]. The irradiation temperature was 750 K at which the RIS effect should be a maximum for the dose rate used --based on the observations of Okamoto and Rehn [18] that maximum RIS occurs about 40° below the peak swelling temperature for pure nickel. Some of the specimens were preimplanted with alpha-particles at 970 K to intentionally induce grain boundary helium embrittlement during subsequent mechanical testing in order to study RIS at grain boundaries by Auger electron spectroscopy (AES). This helium concentration was varied between 250 and 1000 at. ppm He. After irradiation most of the specimens as well as unirradiated controls were tensile tested either at room temperature or at 720 K. The tensile machine was especially designed for foil specimens. The strain rate was

$5 \times 10^{-6} \text{ s}^{-1}$ up to the yield point and then about $3 \times 10^{-4} \text{ s}^{-1}$ until failure. The fracture surfaces were examined in a scanning electron microscope (SEM) to ascertain the type of fracture.

RESULTS

The results of all the tensile tests are summarized in Table 1. In addition, some of the stress-strain curves are plotted in Figs. 1a and 1b for the different test temperatures, 300 and 720 K respectively, to show the influence of the different irradiation modes compared to unirradiated controls. The main effects of the irradiation are a moderate hardening and the consequent reduction of the strains to rupture; this was most pronounced in the alpha-particle irradiated specimens. But all the irradiated specimens still exhibited good ductility and they failed with transgranular, chisel-like fractures as shown in Fig. 2. An exception was specimen No. 12. This specimen exhibited pronounced (helium) embrittlement and a fully intergranular failure (see Fig. 3) as a result of its particular irradiation and test conditions. These were: a high concentration of helium (1000 at. ppm) preimplanted at high temperature (970 K), the maximum irradiation dose at 750 K accompanied by an additional 2300 at. ppm He, and tensile testing at 720 K. Similarly treated specimens (with similar-appearing fractures) have been broken at 970 K "in-situ" in a Scanning Auger Microprobe allowing silicon segregation profiles on individual grain surfaces to be determined [13].

The effects of the irradiation and RIS on the microstructure have been studied also using TEM. The irradiations produced mainly faulted dislocation loops on {111} planes. The use of alpha particles introduced large numbers of small bubbles (1.5 to 2 nm diameter) which were nearly exclusively situated in the planes of the loops and at grain boundaries. The loops and the bubbles are thought to be responsible for the observed hardening.

The high-temperature preimplantation of helium created additional, much larger (10-20 nm) bubbles with lower concentrations. Such bubbles, most often observed in grain boundaries (Fig. 4a), are believed (and were intended) to induce the observed high temperature intergranular fractures. Fig. 4b illustrates the radiation-induced precipitation which has been observed in all irradiated specimens regardless of the irradiation mode. Many types of sinks for point defects are decorated with gamma prime precipitates (Ni_3Si): grain boundaries, twin boundaries, loops, helium bubbles, and the outer surfaces. At grain boundaries the gamma prime layer tends to be about 20-30 nm thick and consistently lies entirely on one side or the other of the boundary (sometimes being observed to change sides). The detailed analysis of the microstructure is given elsewhere [12].

DISCUSSION

The purpose of these experiments was to ascertain if radiation-induced silicon segregation in a binary nickel-silicon alloy influences fracture behavior. As mentioned at the beginning, there have been reports [1-4] of serious degradation in tensile ductility for Fe-Ni-base alloys

after neutron irradiation. The alloys were all precipitation-hardened (γ' -forming), including both commercial (PE-16, Inconel 706) and developmental (D21, D25, D66, D68) types. Detailed microchemical analyses using energy-dispersive x-ray (EDX) microanalysis of the PE-16 and the D-series alloys [2,4] identified continuous grain boundary layers of γ' structure which were held responsible for the embrittlement. These layers were characteristically depleted in Fe and Cr and enriched in Ni, Ti, Al, Si, and Nb — presumably the product of RIS processes. There were other grain boundary precipitates as well, including (depending on the alloy and temperature) $M_{23}C_6$, Laves, and G phases. Embrittlement was most pronounced at T_{irr} and $T_{irr} + 110^\circ$.

The same behavior does not seem to be maintained when the experiment is simplified to the model binary alloy of the present investigation. Even though RIS processes have been active, building up γ' precipitate layers on boundaries throughout the specimens, intergranular failure did not occur except where it was deliberately helium-induced. The fractures were basically ductile whether the tests were conducted at room temperature or near T_{irr} (see Fig. 2 for which the test temperature was 720 K). The composition of the γ' precipitate formed here was simple Ni_3Si as opposed to the complex composition of the earlier neutron studies, and there is some evidence that Ni_3Si (unlike Ni_5Si_2) is not an inherently brittle phase [19]. It should also be acknowledged that the relatively large grains and thin specimen sections of this experiment (frequently only two or three grains through the thickness) may not have fully reproduced the strain

constraints that would be experienced by grains in a true bulk environment and which might enhance to some degree the prospects for intergranular failure.

It is worth noting that ductility was maintained for alpha particle as well as proton-irradiated material which means that the effect of RIS plus helium (if not implanted at high temperature) was not substantially different from that of RIS alone (i.e., no synergistic degradations). Further light may be shed on the causes of the observed behavior by detailed TEM study (in progress) of the interfacial precipitation [12].

SUMMARY

Experimental tests of the effect of radiation-induced segregation of silicon on mechanical behavior have been carried out in Ni-8 at. % Si irradiated up to 0.3 dpa with light ions and subsequently tensile tested at either room temperature or near the irradiation temperature of 750 K. Excluding specimens intentionally subjected to a helium embrittlement treatment, the fracture behavior of either proton or alpha-particle-bombarded specimens remained ductile and transgranular despite the occurrence of substantial silicon segregation and γ' phase formation at internal interfaces.

REFERENCES

- [1] R. Bajaj, R. P. Shogan, C. DeFlitch, R. L. Fish, M. M. Paxton, and M. L. Bleiberg, in Effects of Radiation on Materials: Tenth Conf., ASTM STP 725, D. Kramer, H. R. Brager, and J. S. Perrin, Eds., (Amer. Soc. for Testing and Mater., 1981) pp. 326-351.
- [2] W. J. S. Yang, J. Nucl. Mater. 108 & 109 (1982) 339.
- [3] S. Vaidyanathan, T. Lauritzen, and W. L. Bell, in Effects of Radiation on Materials: Eleventh Conf., ASTM STP 782, H. R. Brager and J. S. Perrin, Eds., (Amer. Soc. for Testing and Mater., 1982) pp. 619-635.
- [4] W. J. S. Yang, D. S. Gelles, J. L. Straalsund, and R. Bajaj, J. Nucl. Mater., 132 (1985) 249.
- [5] P. R. Okamoto and H. Wiedersich, J. Nucl. Mater. 53 (1974) 336.
- [6] L. E. Rehn, P. R. Okamoto, D. I. Potter, and H. Wiedersich, J. Nucl. Mater. 74 (1978) 242.
- [7] L. E. Rehn, P. R. Okamoto, and H. Wiedersich, J. Nucl. Mater. 80 (1979) 172.
- [8] R. C. Pillar and A. D. Marwick, J. Nucl. Mater. 71 (1978) 309.
- [9] W. Wagner, L. E. Rehn, and H. Wiedersich, Phil. Mag. A 45, 6 (1982) 957.
- [10] J. Nagakawa, V. K. Sethi, and A. P. L. Turner, in Phase Stability During Irradiation, J. R. Holland, L. K. Mansur and D. I. Potter, Eds., (Met. Soc. AIME, 1981) pp. 445-459.
- [11] T. Atkins and R. J. McElroy, in Effects of Radiation on Materials: Eleventh Conf. ASTM STP 782, H. R. Brager and J. S. Perrin, Eds., (Amer. Soc. for Testing and Mater., 1982) pp. 841-855.

- [12] W. Kesternich, N. H. Packan, and H. Schroeder, "Radiation-Induced Segregation to Grain Boundaries in a Ni-Si Alloy Studied by Transmission Electron Microscopy", to be published.
- [13] N. H. Packan, L. Heatherly, W. Kesternich, and H. Schroeder, "Radiation-Induced Segregation in Light-Ion Bombarded Ni-8% Si", paper presented at Effects of Radiation on Materials: Thirteenth Conf., Seattle, June 1986, Proceedings to be published by ASTM.
- [14] H. Schroeder and P. Batfalsky, J. Nucl. Mater. 103 & 104 (1981) 839.
- [15] P. Batfalsky, doctoral thesis, KFA Jülich report JUL-1923, 1984.
- [16] H. Schroeder, W. Kesternich, and H. Ullmaier, Nucl. Eng. and Design/Fusion 2 (1985) 65.
- [17] F. W. Wiffen in Effects of Radiation on Structural Materials: Ninth Conference, ASTM STP 683, J. A. Sprague and D. Kramer, Eds., (Amer. Soc. for Testing and Mater., 1979) pp. 88-106.
- [18] P. R. Okamoto and L. E. Rehn, J. Nucl. Mater. 83 (1979) 2.
- [19] K. J. Williams, J. Inst. of Metals 97 (1969) 112.

Table 1. Irradiation and Tensile Data

Spec. No.	Preimpl. He at 970 K (ppm)	Dam. Bomb. 750 K		<u>Mechanical Properties</u>				Other Studies
		dpa	He (ppm)	Test Temp (K)	σ_y (MPa)	σ_u (MPa)	total elong. (%)	
CONTROLS								
6	--	--	--	300	133	432	39	
8	--	--	--	300	133	452	35	TEM
10	--	--	--	720	99	334	25	
BOMBARDED by 28 MeV ALPHAS								
2	--	0.2	1500	300	328	561	17	TEM
7	260	0.3	2300	300	336	611	18	Fract.in SAM;TEM
11	1000	0.15	1100	--				TEM
12	1000	0.3	2300	720	270	397	2.5	TEM
BOMBARDED by 7 MeV PROTONS*								
3	525	0.3	--	--				Fract.in SAM;TEM
4	--	0.13	--	300	198	504	28	Fract.in SAM (ductile); TEM
5	--	0.13	--	--				TEM
9	--	0.1	--	720	151	391	24	

* protons stopped within specimens except for spec. no. 5

FIGURE CAPTIONS

Fig. 1. Stress-strain curves for 0.060-mm-thick Ni-8 at. % Si specimens tested at (a) 300 K and (b) 720 K (450°C). Specimens 6 and 10 are controls, 4 and 9 were proton-bombarded to 0.1 dpa, while 2 and 12 were irradiated to 0.2-0.3 dpa with alpha particles; No. 12 also received an additional 1000 at. ppm of helium preimplanted at 970 K.

Fig. 2. Fracture surface of proton-bombarded specimen No. 9 tested at 720 K illustrating the ductile, chisel-type failure typical of most irradiated and control specimens.

Fig. 3. Brittle, fully-intergranular fracture resulting from high-temperature helium preimplantation; shown is specimen No. 12 tested at 720 K.

Fig. 4. Transmission electron micrographs of Ni_3Si phase (γ' , L1_2 structure) formed in (a) specimen No. 12 preimplanted with 1000 at. ppm He at 970 K and α -particle-irradiated to 0.3 dpa at 750 K and (b) specimen No. 5 proton-irradiated to 0.13 dpa. The bright field TEM of (a) illustrates large helium bubbles on the grain boundaries (from the 970 K preimplantation), small bubbles from the 750 K alpha irradiation, and Ni_3Si precipitate (gray band) predominantly to one side of the boundary. Dark field TEM (b) reveals the occurrence of Ni_3Si at a grain boundary and on dislocation loops in the grain interiors.

Fig. 1

a

b

Fig 2

Fig. 3

50 μ m

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