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Two Ni alloys with 4 at.% Si and 12.7 at.% Si in solution have been irradiated at 4.2K with 14 MeV fusion neutrons. The resistivity damage rate of well annealed Ni-4%Si alloy showed an initial transient in the plot of $d\Delta\rho/d\Delta\phi t$ versus $\Delta\rho$. A high dislocation density appeared to reduce this transient. The resistivity damage rate of Ni-12.7%Si alloy showed an unusual behavior; $d\Delta\rho/d\Delta\phi t$ increases with $\Delta\rho$ after the initial transient period. This behavior is attributed to precipitation and growth of Ni_3Si during irradiation.

Post-irradiation isochronal annealing results showed significant effects of cold work and composition on recovery. Ni-4%Si recovered slower than pure nickel and a high dislocation density enhanced its recovery. For the Ni-12.7% Si alloy, recovery ended after being annealed to 38K, after which the resistivity increased with annealing temperature. This is attributed to further precipitation and growth of Ni_3Si.

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1. Introduction

In general, irradiation of a metal with high energy particles produces both isolated Frenkel pair defects and their clusters. The rate of defect production is a function of temperature, alloying content, particle energy, fluence, etc. At very low temperatures where defects are immobile, their production rate can be measured by electrical resistivity. At high temperatures, where defects are mobile, their survival is reduced by recombination and by annihilation at sinks. The remaining defects in the form of stable clusters, loops, etc., are more difficult to measure due to complicating concurrent phenomena such as radiation induced precipitation, which also affect measured physical properties. Our intent is to use information gained in studying defect production at low temperatures to help in interpreting high temperature experiments. Reported in this paper are results on the measurement of defect production in two Ni(Si) alloys at 4.2K. High temperature irradiations, which are in progress, will be reported in the future. Ni(Si) alloys have attracted much attention in the field of irradiation effects research due to the strong binding between Ni interstitials and Si solute atoms [1]. Several studies [2-5] had shown that radiation induced precipitation occurred in alloys containing 1-13% Si at elevated temperatures. The relative efficiency of defect production as a function of irradiating ion energy has also been investigated [6].

2. Experimental procedures

Two Ni(Si) alloys were studied: A Ni-4%Si sample was supplied by G. Martin of CEN de SACLAY. The received sample was 150 um thick and had been annealed at 1000°C for 4 hours in vacuum. A portion of the sample was sheared into wire and reannealed at 1000°C for 4 hours before irradiation. Another portion of the sample was cold rolled to 100 um, sheared into wire, then annealed at 625°C for one week in vacuum before irradiation. The Si solutes are fully in solution for both samples since 4% Si is less than the solubility limit in Ni even at room temperature. The resistivities of the samples at 4.2K were 109.941 nΩm and 105.970 nΩm respectively. Transmission electron microscopy was used to investigate the microstructure of both samples. No second phases were detected. The dislocation density was determined as $10^{12}/m^2$ - $10^{13}/m^2$ for the 1000°C annealed sample and $10^{14}/m^2$ - $10^{15}/m^2$ for the 625°C annealed sample.

The Ni-12.7%Si sample was supplied by A. Ardell of UCLA. A sheared resistivity sample, 25 um thick, was solution annealed at 900°C for two hours in vacuum, followed by water quench. The solvus of this alloy is 756°C [7]. The solution treatment dissolved all Si solutes before quenching. The initial resistivity of this sample was 256.792 nΩm at 4.2K.

The 14.8 MeV neutron irradiation was performed at the Rotating Target Neutron Source (RTNS-II) of Lawrence Livermore National Laboratory with a helium cooled cryostat. The samples were mounted at the tip of the cryostat with epoxy. Niobium foils were spaced on both sides of each sample to monitor neutron fluences. The temperature of samples were monitored with two carbon glass resistors located near the samples. A proton recoil counter was used to monitor total neutron production rate. This information and the total fluence determined from foils at each sample were used to derive irradiation histories. During the experiment, the flux varied from 1.0×10^{16} to 1.4×10^{16} neutrons/m^2-sec.

Electrical resistivity was monitored using a four point measurement technique with a computerized data acquisition system. The system was programmed to measure in both current directions and average the results.

3. Results and discussion

3.1 Resistivity data and defect production

The irradiation results are summarized in Table 1. The total resistivity change, fluence, average and initial resistivity damage rates are given for each sample except the Ni-12.7%Si, for which the change in initial rate was so rapid that extrapolation to zero was not possible. The resistivity damage rates are also plotted versus irradiation induced resistivity in Figures 1 and 2. For comparison, the damage rate of nickel [8] irradiated with 14 MeV neutrons at 4.2K is also included in Figure 1.

The resistivity damage rate of Ni-4%Si is about 10% lower than nickel, except in the initial stage, in which the well-annealed Ni-4%Si shows a transient. The high Si content might contribute to the disruption of focussed energy transport out of the cacade, resulting in slower cascade cooling, leading to more defect recombination.

If the resistivity damage is related to the defect concentration linearly, the initial resistivity damage rate can be related to the damage energy cross section by the modified Kinchin-Pease expression [9] as

$$\left(\frac{d\Delta\rho}{d\Delta\phi t}\right)_{\Delta\rho = 0} = \frac{K\sigma_{DE}\rho_F}{2T_A} \quad (1)$$

where ρ_F is the resistivity per atom fraction of Frenkel pairs, T_A is the effective displacement energy average over crystallographic directions. σ_{DE} is the damage energy cross section, and K is an efficiency factor. The values of these parameters for pure nickel are used for the current evaluation. These are 6 uΩm and 33 ev. for ρ_F and T_A respectively [10], and 301 kev-b [8] for σ_{DE}.

Using the experimentally determined initial damage rate in Equation 1, the defect production efficiency factor K was determined as .34 for the 1000°C annealed Ni-4%Si, .22 for the 625°C annealed Ni-4%Si, and >.60 for the Ni-12.7%Si, while the value for nickel was previously [8] determined as .23.

3.2 Effect of silicon and dislocation density on defect production

The addition of silicon produces large increases in the initial damage rate in nickel, but the enhancement is short-lived and disappears well before any significant cascade overlap occurs. The usual magnetoresistive explanation [11] cannot apply here since the radiation induced resistivity is only a few percent of the initial resistivity. We believe the effect arises from the stabilization of close pairs by silicon. In general, cascade damage in metals [8,11] exhibits a lack of the close pair recovery stages which are so prominent in electron irradiated samples. These defects anneal during the cascade cooling phase [12]. Since the silicon-stabilized close pairs are only marginally stable, they may be easily annealed by the energy overlap of a nearby cascade.

We note that the energy overlap volume is hundreds of times larger than the cascade volume itself. This then accounts for the rapid decrease in damage rate to values comparable to pure nickel.

The difference between the 1000°C and 625°C annealed samples indicates that the presence of a high dislocation density nearly completely suppresses the enhanced defect production. Even at these densities, the direct interaction of cascades with dislocations will be a small fraction of the total events. The suppression must arise from the influence of the long range strain fields on close pair stability.

Certainly a more complicated experiment, such as a cryo-transfer experiment, which allows the direct observation of cascade structure by TEM before annealing, will have to be done to elucidate the effects of dislocations on cascade defect production.

3.3 Defection production in the non-equilibrium Ni-12.7%Si alloy

The results for the Ni-12.7%Si alloy in Figure 2 show a very interesting resistivity damage rate behavior. This alloy has a low dislocation density comparable to the 1000°C annealed Ni-4%Si. Initially, the resistivity damage rate curve shows a transient similar but of much larger amplitude than in Ni-4%Si. As before, the decreasing damage rate in this transient can be attributed to the recombination of Si stabilized close Frenkel pairs due to energy overlap of nearby cascades. The larger rate is probably due to the increased numbers of Si stabilized close pairs.

In the later stage of irradiation, the resistivity damage rate increases with total resistivity damage. This can be attributed to the precipitation and coarsening of Ni_3Si precipitates [13]. As the density of cascades increases with irradiation, the overlap of cascade cores with existing precipitates can contribute to their growth.

3.4 Post-irradiation annealing behavior

Figure 3 shows the results of annealing after irradiation for the Ni-4%Si alloy. For comparison, the results of 14 MeV neutron irradiated Ni [8] are also included. The two Ni-4%Si samples show similar behavior up to 200K, above which the 625°C annealed sample has a more rapid recovery rate and shows an additional peak near 210K. This difference results from its high dislocation density, which provides sinks for the annihilation of interstitials and their complexes, thus enhancing recovery.

The annealed peak at 58K is close but somewhat higher in temperature than the first peak in annealed Ni. It is believed that this peak is due to the recombination of free interstitials and vacancies. The fact that its amplitude for the Ni-4%Si alloy was lower than that for the Ni suggests that a larger fraction of interstitials is trapped (presumably with Si) in the Ni-4%Si alloy.

The annealing peak at 152K in Ni-4%Si can be attributed to the migration of Ni-Si complexes to vacancies. It is not likely this peak is due to detrapping of Ni-Si complexes, since it is well known that the complexes exist even at very high temperatures [2-5]. It is also not likely that this peak is due to the long range migration of Ni-Si complexes to dislocations, as there is no significant difference in recovery for the two Ni-4%Si samples.

In contrast to the Ni-4%Si, the previously studied Ni shows three second stage peaks near 100, 160 and 210K. Since this nickel had a low resistivity ratio of 17, these peaks are most likely due to the migration or detrapping of interstitials at different impurities.

The annealing results for the non-equilibrium Ni-12.7%Si alloy are shown in Figure 4. After the first annealing peak near 38K, the resistivity goes through a negative recovery, i.e. resistivity increases with annealing. This phenomenon could be due to the precipitation and growth of small Ni-Si precipitates such as Ni_3Si. As discussed above, the growth of these precipitates induced by cascade mixing are possibly also responsible for the increasing resistivity damage rate with total resistivity damage during irradiaiton.

The first annealing peak of this alloy is lower than that of Ni-4%Si, 38K vs. 52K. It suggests that numbers of Si stabilized close pairs still remain and recover at this temperature. In this alloy, free interstitials will not exist since each lattice site will have one or more Si nearest neighbors.

4. Conclusions

We have irradiated two Ni(Si) alloys with fusion neutrons at 4.2K and conducted post-irradiation annealing. We observed that dislocations eliminated an initial transient in the defect production rate of Ni-4%Si. Except in the transient region, the defect production rate in Ni-4%Si was about 10% lower than in pure nickel. Post-irradiation annealing of Ni-4%Si showed that Ni-Si interstitial complexes started to migrate near 150K. Dislocations enhanced the recovery rate in the second stage of annealing above 200K. Resistivity damage rate and recovery in Ni-12.7%Si are complicated by the nucleation and growth of stable precipitates during both irradiation and annealing.

5. Acknowledgement

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TABLE 1. Summary of 4.2K Irradiation Results for Ni(Si) Alloys

Sample	$\Delta\rho_{max}$ (nΩm)	$\Delta\phi t$ ($10^{21} m^{-2}$)	$\left(\frac{\Delta\rho}{\Delta\phi t}\right)_{average}$ ($10^{-31} \Omega m^3$)	$\left(\frac{d\Delta\rho}{d\Delta\phi t}\right)_{\Delta\rho = 0}$ ($10^{-31} \Omega m^3$)
Ni-4%Si (annealed at 1000°C)	1.725	2.992	5.8	9.2
Ni-4%Si (annealed at 625°C)	2.149	4.088	5.3	6.0
Ni-12.7%Si	2.766	1.706	16.2	>16.2

Figure Captions

Figure 1. Resistivity damage rate versus the resistivity increase in Ni-4%Si irradiated at 4.2K.

Figure 2. Resistivity damage rate versus the resistivity increase in Ni-12.7%Si irradiated at 4.2K.

Figure 3. Post-irradiation annealing results in Ni-4%Si and Ni.
(a) Showing percent of resistivity unrecovered versus temperature;
(b) Showing percent of resistivity recovered per degree Kelvin versus temperature.

Figure 4. Post-irradiation annealing results in Ni-12.7%Si after 14.8 MeV neutron irradiation.

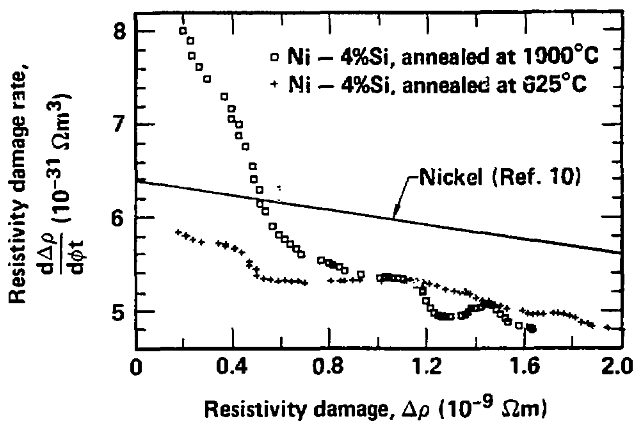
Resistivity damage rate,
$\frac{d\Delta\rho}{d\phi t}$ (10^{-31} Ωm³)
8
7
6
5
0
0.4
0.8
1.2
1.6
2.0
Ni – 4%Si, annealed at 1000°C
Ni – 4%Si, annealed at 625°C
Nickel (Ref. 10)
Resistivity damage, Δρ (10^{-9} Ωm)

Figure 1

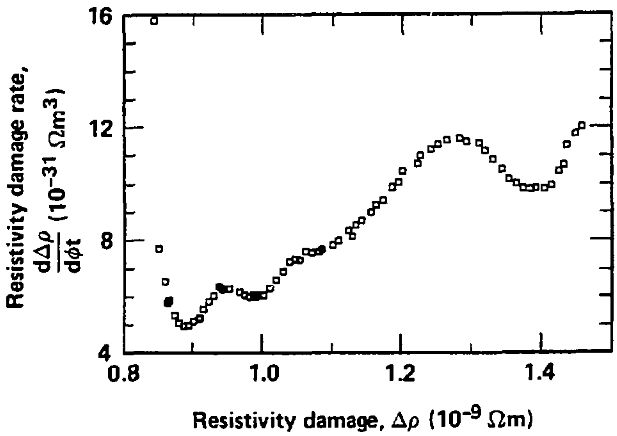
Resistivity damage rate,
dΔρ/dφt (10^-31 Ωm^3)
16
12
8
4
0.8
1.0
1.2
1.4
Resistivity damage, Δρ (10^-9 Ωm)

Figure 2

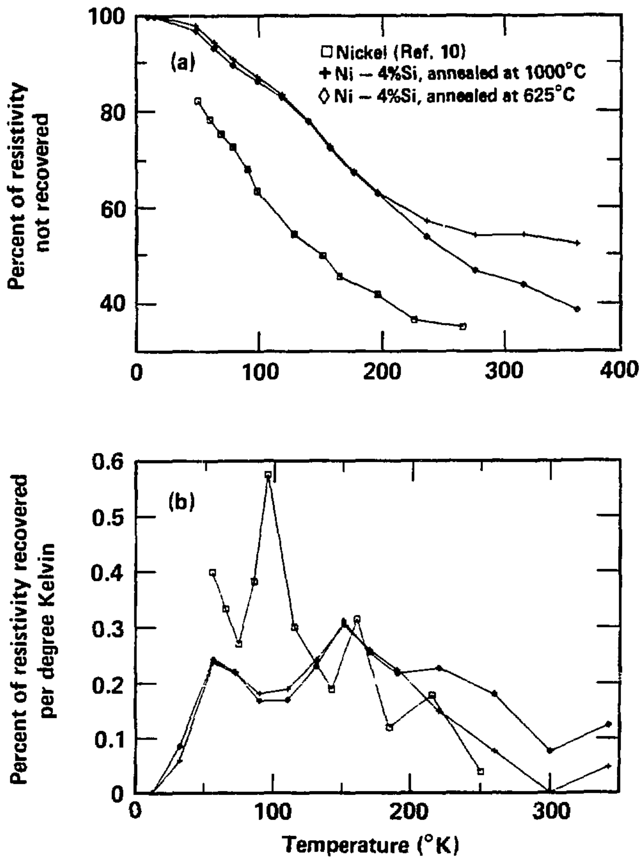

Figure 3a and 3b

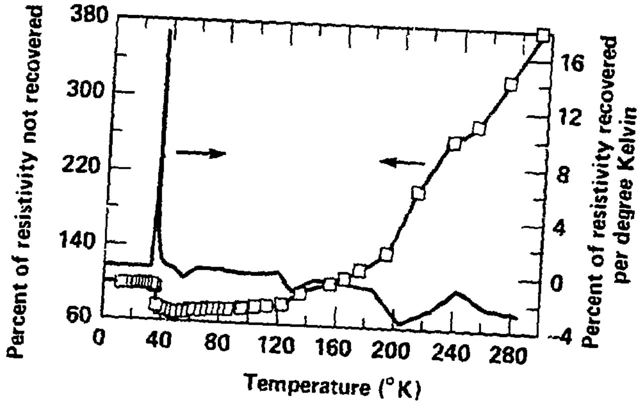

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Figure 4