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J. S. Huang, M. W. Guinan and P. A. Hahn

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Fusion Neutron Irradiation of Ni(Si) Alloys at High Temperature*

J. S. Huang, M. W. Guinan and P. A. Hahn

Lawrence Livermore National Laboratory
P.O. Box 808
Livermore, CA 94550, U. S. A.

ABSTRACT

Two Ni-4% Si alloys, with different cold work levels, are irradiated with 14 MeV fusion neutrons at 623 K, and their Curie temperatures are monitored during irradiation. The results are compared to those of an identical alloy irradiated by 2 MeV electrons. The results show that increasing dislocation density increases the Curie temperature change rate. At the same damage rate, the Curie temperature change rate for the alloy irradiated by 14 MeV fusion neutrons is only 6-7% of that for an identical alloy irradiated by 2 MeV electrons. It is well known that the migration of radiation induced defects contributes to segregation of silicon atoms at sinks in this alloy, causing the Curie temperature changes. The current results imply that the relative free defect production efficiency decreases from one for the electron irradiated sample to 5-7% for the fusion neutron irradiated sample.

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1. Introduction

Irradiation of materials by high-energy particles produces vacancy and interstitial defects which are responsible for many types of microstructural evolution. Some examples are void swelling, precipitation of second phases, and disordering of ordered alloys. Extensive efforts have been made in the past decades to study defect production by different kinds of particle irradiation. Through both computer simulation[1-3] and transmission electron microscope observation[4-7], it is now recognized that one of the most important variables in determining defect production is the mass and energy of the irradiating particle. As this energy increases, the primary recoil energy of the irradiated material increases, and the defect configuration changes from the one with randomly distributed Frenkel pairs to a configuration of defects in close proximity to each other within cascades. The result of this non-random distribution of defects is the reduction of the probability for defects to freely escape from cascades and to participate in microstructural evolution processes. An accurate understanding of free defect production under cascade-formation situations is particularly important for materials irradiated by fusion neutrons since at the high neutron energy nearly all defects are produced in cascades. There are several studies on defect production by fusion neutrons[8-9], conducted at the liquid helium temperature using 14 MeV neutron irradiation. For most materials, the defect production rate for irradiation by high energy fusion neutrons is approximately one-quarter to one-third of that for irradiation by electrons or low energy particles which do not produce cascade defects. There are very few studies on defect production in materials irradiated by fusion neutrons at elevated temperatures. The intent of this work is to compare the efficiency of free

defect production by 14 Mev fusion neutrons in a model alloy, Ni-4% Si, with that of other type of irradiation such as electron irradiation. The technique used is to measure the radiation induced Curie temperature change. It has been known that particle irradiations on Ni(Si) alloys produce strong segregation of Si atoms at sinks[10-13] due to the coupling of Si atoms with free migrating nickel interstitials. The irradiation induced segregation(RIS) of Si atoms leads to large changes of the Curie temperatures of the alloys. For one atomic percent of Si atom depleted from the nickel matrix of a Ni(Si) alloy, its Curie temperature increases by about 37 K. Therefore, by comparing irradiation induced Curie temperature changes, the relative efficiency of free defect(interstitial) production can be found.

2. Experimental Details

The Ni-4% Si alloy studied in this work was supplied by G. Martin of CEN de SACLAY. A study of electron irradiation on this alloy had been published by Barbu and Martin[10]. The received foil had been originally annealed at 1273 K for four hours. The thickness of the foil is 150 μm . A portion of the foil was sheared into a wire sample(15 mm long and 0.25 mm wide) and reannealed at 1273 K for 4 hours before irradiation. Another portion of the foil was cold rolled to 100 μm , sheared into a wire sample(same dimensions as above), then annealed at 898 K for one week in vacuum before irradiation. For both the heat treatments, the silicon atoms are fully in solution in the nickel matrix before irradiation. The dislocation densities and grain sizes of these samples were determined using transmission electron microscopy. Results were based on averages generated from 15 micrographs for each sample. The density is $3 \times 10^{12}/\text{m}^2$ for the 1273 K annealed sample with a grain

size of $130\text{ }\mu\text{m}$, and $2 \times 10^{13}/\text{m}^2$ for the 898 K annealed sample with a grain size of $60\text{ }\mu\text{m}$.

The samples are irradiated in vacuum. The initial vacuum is 5×10^{-6} mbar at the ambient temperature before irradiation, and 1.0×10^{-4} mbar at 623 K during irradiation. The samples are attached onto the front end of a cylindrical copper block which is located within the vacuum chamber and maintained at the irradiation temperature (623 K) by a cylindrical-shaped heater embedded in the center of the copper block. The copper block has many internal cooling channels to allow the passage of helium gas to facilitate control of temperature. To insulate the samples, electrical wires (used for four-point resistance measurement), and thermocouples wires from electrical ground, a liquid B_4C compound was used to coat the samples, wires and the copper block. The whole assembly was baked at 623 K before irradiation, the additional outgassing of this compound during irradiation accounts for the poor vacuum.

The experiment was conducted in the following sequence. The samples were irradiated at 623 K to a fluence level which was usually determined by operating convenience. Then the irradiation was turned off, and the samples were cooled to the ambient temperature at a fixed cooling rate. During the cooling, the resistance and temperature of samples were monitored with a computerized data acquisition system. Once the samples were cooled to the room temperature, they were reheated to 623 K and the resistance and temperature were monitored again. As soon as the temperature was at 623 K, the irradiation was continued. The resistances of samples were measured by a

four-point measurement technique. The potential and current leads were spot-welded on the samples. The temperature of samples was monitored by two Pt-Pt(10%Rh) thermocouples which were also spot-welded on the samples. For a ferromagnetic material such as the current nickel alloy, its electrical resistivity shows a change of temperature coefficient at its Curie temperature. A typical plot of resistance versus temperature is shown in Fig. 1 for the sample annealed at 1273 K. Note that the temperature coefficient of resistivity drastically decreases at a temperature near 483 K. To define the Curie temperature of a sample, the resistivity data between 473 and 481 K were first fitted by the least-square method to a linear function. The differences between the actual resistivity data and the values calculated by the function at different temperatures were then plotted. The Curie temperature is then defined by visual determination of the temperature where the calculated difference first deviates from zero. For example, Fig. 2 shows the calculated differences for Fig. 1, and the Curie temperature is determined as 483.51 K.

Due to irradiation damage of the thermocouples, the measured Curie temperature has to be corrected. Right after irradiation, a new thermocouple was installed near a sample. The sample was heated to 483 K, which is near the Curie temperatures of the samples. The difference of temperature indicated by his new thermocouple and that by an old one was measured. After this, the samples were cooled to room temperature, and the wires for the junction of the new thermocouple(unirradiated) were attached to the junction for the old(irradiated) thermocouple and vice versa. The samples were then heated to 483 K and the temperature difference between these two sets of

thermocouples was measured. The difference measured from the first set-up reflects the sum of errors induced from irradiation on a thermocouple junction and its wires, subtracted with the actual temperature difference between the two junctions due to any non-uniformity of temperature. The difference measured from the second set-up reflects the error induced by irradiation on a thermocouple junction, subtracted with the error due to irradiation on the wires of a thermocouple and the actual temperature difference between the two junctions due to non-uniformity of temperature. From these two differences, the overall error due to irradiation damage on a thermocouple(junction plus wires) is estimated. The estimated value is $-0.50 (\pm 0.30 \text{ K})$, where the uncertainty is caused by the unprecise information on the actual temperature difference between the two thermocouple junctions due to non-uniformity of temperature. This estimated error is used for the correction of Curie temperature at final fluence. For a fluence less than this, the correction is assumed linearly proportional to fluence.

The irradiation was performed at the Rotating Target Neutron Source (RTNS-II) of Lawrence Livermore National Laboratory. Niobium foils were mounted side by side with samples to monitor neutron fluences. A proton recoil counter was used to monitor the total neutron production rate. This information and the total fluence determined from the niobium foils were used to derive irradiation flux. The average of flux is determined as 3.0×10^{15} neutrons/m²-s, and the variation of flux through the whole irradiation history is less than 10%. The total fluence is 2.96×10^{21} neutrons/m² for the sample annealed at 1273 K, and 2.86×10^{21} neutrons/m² for the sample annealed at 898 K.

3. Results and Discussions

3.1 Curie temperature change versus fluence

The measured Curie temperature changes versus irradiation fluence for the two samples are summarized in Table 1. The damage energy corresponding to each fluence is also shown in the same table. The damage energy is calculated by using a damage energy cross section for 14.8 MeV neutrons of 296 KeV-barn[14]. The Curie temperature changes versus the calculated damage energy are plotted in Fig. 3. These data are fitted to linear equations as shown in the figure. For the sample annealed at 1273 K, the effects of current irradiation doses on the Curie temperature is very small. For the sample annealed at 898 K, the change is much more significant, and is about 9 times that of the sample annealed at 1273 K. Since, under steady state conditions, we expect the silicon segregation rate to be proportional to sink density[17] the results for the sample annealed at 898 K should be 7 times larger based on measured sink (dislocation) density. This is in good agreement with the ratio of observed changes in Curie temperature.

3.2 Comparison with the result of 2 MeV electron irradiation

The results of the study by Barbu and Martin[10] on 2 MeV electron irradiation of the same alloy are compared with our results here. For the 2 MeV electron irradiation, we calculated the damage energy cross section as 3.06 KeV-barn by assuming the threshold energy for displacement as 23 eV (for nickel). Figure 4 shows a plot of the Curie temperature change from their work versus the calculated damage energy. The alloy used for this work is annealed at 1273 K for 4 hours, which is the same heat treatment as that of

our sample with low dislocation density. Comparing the results in Fig. 3 with those in Fig. 4, it can be seen that the rate of Curie temperature increase in our 1273 K annealed sample is one-tenth of that of the 2 MeV electron-irradiated sample. However the effect of different dose rate has to be accounted for a better comparison. Johnson and Lam[15] developed a model to describe solute segregation during irradiation. Their model shows that decreasing dose rate increases RIS at intermediate temperatures where the thermal equilibrium vacancy concentration is not the dominant component of the total vacancy concentration and the mutual recombination of radiation induced vacancies and interstitials contributes significantly to defect annihilation. This can be simply reasoned as that at lower dose rate, point defects have a greater chance of making long-range migration to sinks and contributing to RIS. Rehn, Okamoto and Averback[12] conducted measurement of RIS in ion-irradiated Ni-12.7% Si and Ni-10% Ge alloys. Their results confirmed this phenomenon. Rehn and Okamoto[16] further estimated that the rate of RIS varied inversely as the fourth-root of the dose rate. For the electron irradiation experiment[10] on Ni-4% Si alloy, the flux is about $1.44 \times 10^{18} / \text{m}^2\text{-s}$, which corresponds to a damage energy rate of $4.5 \times 10^{-7} \text{ eV/s}$. For our experiment, the flux is $3.0 \times 10^{21} / \text{m}^2\text{-s}$, which corresponds to a damage energy rate of $8.9 \times 10^{-8} \text{ eV/s}$. The dose rate of our experiment is a factor of five lower than that of the electron irradiation experiment. Using the dose rate dependence of RIS given by Rehn and Okamoto[16], the rate of the Curie temperature increase for our 1273 K annealed sample would be a factor of 1.50 lower if the dose rate were the same as that used in the 2 MeV electron irradiation experiment. Including the original factor of one-tenth, it can be concluded that at the same dose rate, the free defect production rate for the

14 MeV neutron irradiated alloy would only be 6-7% of that of the 2 MeV electron irradiated alloy.

The effect of recoil energy on RIS in a Ni-12.7% Si alloy during ion irradiation has been studied by Rehn, Okamoto and Averback[13]. It was shown that increasing the recoil energy from 730 eV to 740 KeV, the relative RIS efficiency decreased from 100% to about 2%. In the current results, the relative RIS efficiency decreases from 100% for the 2 MeV electron irradiated sample (average recoil energy = 60 eV) to 6-7% for the 14 MeV neutron irradiated sample(average recoil energy = 500 KeV). Considering the data scatter involved in this work, the agreement between these two results is good. Furthermore, in the present comparison, segregation occurred in the bulk to internal sinks as contrasted with the surface segregation measured in the ion studies.

It is important to note that the decrease of relative free defect production efficiency from electron irradiation to 14 MeV neutron irradiation at elevated temperatures is much lower than that for total defect production at very low temperatures. At very low temperatures, this decrease is from one to about one-quarter for most FCC and one to one-third for most BCC metals and alloys[8,9]. This difference suggests that at elevated temperatures, after the cascade cooling phase(10^{-11} s), there is additional recombination of defects within a cascade or in-between cascades.

4. Conclusions

We have measured the rate of Curie temperature increase as a function of irradiation dose in two Ni-4% Si alloys, irradiated by 14 MeV fusion neutrons at 523 K, and compare the results with those by 2 MeV electron irradiation.

The following conclusions can be made from the results of this work:

1. Increasing dislocation density in the alloy increases the rate of radiation induced segregation proportionally.

2. For the same alloy and under the same damage energy rate condition, the free defect production rate by 14 MeV neutron irradiation is only 6-7% of that by 2 MeV electron irradiation.

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Table 1 Summary of the effect of 14-Mev neutron irradiation on the Curie temperature of Ni-4% Si alloy

<u>Sample (1273 K annealed)</u>			<u>Sample (898 K annealed)</u>		
<u>Fluence</u>	<u>Damage-Energy</u>	<u>ΔT</u>	<u>Fluence</u>	<u>Damage-Energy</u>	<u>ΔT</u>
<u>(10^{13}m^{-2})</u>	<u>(eV/atom)</u>	<u>(K)</u>	<u>(10^{13}m^{-2})</u>	<u>(eV/atom)</u>	<u>(K)</u>
0.75	0.0223	-0.10	0.73	0.0216	-0.13
2.02	0.0597	0.03	1.95	0.0577	0.22
2.02	0.0597	0.09	1.95	0.0577	0.55
2.96	0.0876	0.25	2.86	0.0847	0.81
2.96	0.0876	-0.06	2.86	0.0847	0.40
2.96	0.0876	-0.06	2.86	0.0847	0.91

Figure Captions

- Fig. 1 A typical plot of the electrical resistivity versus temperature for the Ni-4% Si sample annealed at 1273 K.
- Fig. 2 The temperature dependence of the electrical resistivity difference between the actual data and the linearly fitted equation which is obtained from least-square fitting of the resisting data (Fig. 1) between 473 K and 481 K.
- Fig. 3 Curie temperature change versus damage energy for the Ni-4% Si alloys irradiated by RTNS-II fusion neutrons at 623 K.
- Fig. 4 Curie temperature change versus damage energy for the Ni-4% Si alloy irradiated by 2 MeV electron at 623 K (from Ref. 10).

Figure 1

A-067-U-6856-3

Figure 2

A-067-U-6858-4

Figure 3

A-067-U-6859-2

Figure 4

A-667-U-6858-1