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**MICROSTRUCTURAL CHANGES IN DILUTE NI-Be ALLOYS
DURING HVEM SUB-THRESHOLD IRRADIATIONS***

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ABSTRACT

The microstructural sensitivity of Ni-0.7 at.% Be alloys to HVEM sub-threshold irradiations was investigated. Several aspects were examined: (i) dose dependence of the microstructure change during irradiation with 350-keV electrons at 350°C; (ii) energy dependence of the incubation dose for the first appearance of black dots in the alloy films under irradiation at 350°C along various crystallographic directions; and (iii) temperature dependence of the microstructural evolution during 350-keV electron irradiation. It was found that sub-threshold irradiations were capable of inducing nonequilibrium solute segregation. Below ~400°C, segregation-induced homogeneous precipitation of the γ -phase occurred in the alloy matrix, whereas at higher temperatures, only heterogeneous precipitation was observed at defect sinks. From the information about the energy dependence of the incubation dose for precipitation, the displacement threshold energy for Ni and point-defect production rate by secondary Be-Ni collisions were estimated.

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1. Introduction

Most of the component materials for future fusion reactors will contain low-atomic-mass elements (H, D, T, He, Li, Be...) either by controlled alloying or because of unavoidable contamination. For example, coating of the first walls and limiters with replenishable light elements is believed to drastically reduce the sputtering effects and, hence, prevent plasma contamination by sputtered particles. However, the presence of low-Z elements in these component materials results in a number of new problems related to microstructural changes and alloy phase stability. One of the problems will be discussed here, involving the effect of light solutes on the production of point defects. As demonstrated experimentally in our previous work [1-3], only small additions of light interstitial solutes can significantly enhance the defect production in heavy-host matrix under sub-threshold irradiation. This effect may have important implications in the assessment of damage processes in fusion reactor materials.

Since our previous study was concerned with interstitial solutes [1-3], a question has recently been raised: could the unexpectedly-high sensitivity of interstitial alloys (such as Pt-C and Ni-C) to sub-threshold irradiations be generalized to substitutional alloys containing low-Z solutes? To find an answer for this question, we have performed new experiments with a Ni-0.7 at.%

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Be solid solution. Some interesting aspects of sub-threshold electron irradiation-induced microstructural changes in this alloy system are reported in the present paper.

2. Experimental Procedure

Thin films of a Ni-0.7 at.% Be alloy used in the present work were prepared and characterized in previous investigations of radiation-induced void swelling [4]. The sample-preparation procedure was already described therein. Irradiations were carried out in the high-voltage electron microscopes (HVEM) at both Argonne National Laboratory and Centre d'Etudes Nucléaires de Saclay, in the energy range 250-1000 keV and at temperatures between 25 and 500°C. Both microscopes are equipped with an ion beam trap, ensuring that there was no ion-bombardment damage of specimens from foreign heavy ions during electron irradiation. Changes in the alloy microstructure were monitored in situ during irradiation, using sufficiently thick regions in the specimens to minimize surface effects. However, thin regions were also irradiated in order to perform a detailed contrast analysis of the final microstructure.

3. Results

3.1 Dose dependence

During irradiation of Ni-0.7 at.% Be alloy films with electrons of energy between 250 and 1000 keV at 350°C, small black dots appeared after a certain incubation dose. These black dots multiplied, grew rapidly and, when large in size, exhibited the strong strain contrast typical of Be-rich precipitates (fig. 1). Extra diffraction spots corresponding to the γ phase were also observed at this stage, which confirmed that these clusters were indeed

precipitate particles. This precipitation was radiation-induced because the alloy was irradiated well above the solvus temperature; the solubility limit of Se in Ni at 350°C is ~1.5 at.% [5]. Stereo electron microscopy revealed that the precipitates were disseminated throughout the irradiated region. At higher doses, dislocation loops were nucleated on the precipitates, grew and eventually tangled, while a number of precipitates divided themselves into small pieces. It is noted that precipitates were observed before dislocation loops, indicating that radiation-induced precipitation may have been homogeneous in nature. In fact, the precipitation sequence was similar to that observed previously by Mukai and Mitchell [6] in Ni-1 at.% Se alloys irradiated with above-threshold, 650-keV electrons at higher temperatures. Since we have not been able to obtain good dark-field images, we could not make any statement about whether radiation-induced solute segregation occurred at dislocation loops at high doses, after these loops had been formed.

3.2 Energy dependence

The effect of the incident-electron energy on the incubation dose required for the appearance of first black dots in the irradiated zone was systematically studied for various principal crystallographic orientations at 350°C. The results are plotted in fig. 2.

There was no published experimental determination of the threshold energy for pure Ni at 350°C. However, according to Bourret [7], the electron energies corresponding to threshold irradiations of pure Ni at room temperature along the $\langle 011 \rangle$, $\langle 001 \rangle$ and $\langle 111 \rangle$ directions are 400, 620 and > 870 keV, respectively. In view of the temperature dependence of the threshold energy measured by Urban and Yoshida [8] for Cu, the neighboring FCC metal, we can assume a reduction of ~50 keV in the threshold energy for Ni between room

temperature and 350°C. Thus, it is obvious that our irradiations were sub-threshold, especially those carried out along the $\langle 001 \rangle$ and $\langle 111 \rangle$ directions. This indicates that Be solutes are very efficient in producing point defects via secondary collisions with Ni atoms. In fact, for a given electron energy, Ni atoms can receive as much as 3 times more energy from secondary Be-Ni collisions than from primary e-Ni collisions. In addition, for the lowest energy used, a simple calculation shows that a Ni atom can receive up to 35 eV in a secondary collision with a Be atom, a value which is comparable to the displacement threshold (Bourret's values are 21, 38 and > 60 eV for the $\langle 011 \rangle$, $\langle 001 \rangle$ and $\langle 111 \rangle$ directions, respectively [7]). The strong contribution of secondary solute-solvent collisions to defect production observed here is thus similar to the effect found previously in Pt-C and Ni-C alloys [1-3].

3.3 Temperature dependence

The dependence of the microstructural evolution on irradiation temperature was investigated with 350-keV electrons impinging along the $\langle 011 \rangle$ direction. It was found that below $\sim 250^\circ\text{C}$, radiation-induced precipitates were so numerous that they could not be resolved individually. At room temperature, especially, the irradiated area only showed a greyish orange-peel at high doses. In the temperature range 300-400°C, γ -precipitates were clearly observed in the matrix, as described in section 3.1. The lower the temperature, the higher the number density of radiation-induced precipitates and the smaller their size. At higher temperatures, however, the scenario was different. For example, after a short irradiation time at 450°C, only some precipitate particles appeared in the matrix, but a lot of them were formed on pre-existing dislocations. Precipitation on dislocations was quite abundant,

anchoring the dislocations and preventing them from climbing (fig. 3). Thus for sub-threshold irradiations of Ni-0.7 at.% Be alloys, $\sim 400^{\circ}\text{C}$ seems to be the transition temperature, below which only radiation-induced homogeneous precipitation [9-11] took place. Above this temperature, radiation-induced heterogeneous precipitation at defect sinks [12-17] was dominant, as shown in fig. 4 for, e.g., 500°C . Within about one minute of irradiation, precipitates appeared on pre-existing dislocations. They acted as anchoring sites for these fast-climbing dislocations. Hence, Bardeen-Herring sources for dislocation regenerative multiplication were formed. New precipitates were also observed to nucleate on fresh dislocation bows, serving as new anchoring sites, and thus giving rise to copious dislocation multiplication. No precipitates were observed in the dislocation-free matrix at this temperature.

4. Discussion

The present study demonstrates that HVEM irradiations of Ni-0.7 at.% Be alloy with electrons of energies lower than the minimum required to displace Ni atoms by direct collisions were able to redistribute light Be solutes, giving rise to radiation-induced precipitation, just as during above-threshold irradiation. The two situations are nearly the same, except that, during sub-threshold irradiation, Ni atoms are only displaced by secondary collisions with Be as an intermediate projectile. As a consequence, for a given electron flux, the rate of point-defect production by this process is considerably lower than that obtained in above-threshold irradiation. In fact, we can make a quantitative comparison by calculating the defect-production rate for sub-threshold irradiation. Using the tabulated cross section for e-Be collisions [18], it can be easily shown that, for the lowest energy used (250 keV) and a flux of $2.8 \times 10^{19} \text{ e/cm}^2\text{-s}$, the displacement rate of Be is $8 \times 10^{-5} \text{ dpa/s}$

(assuming a reasonable Be displacement threshold energy in the range 40-50 eV). If we assume that each displaced Be atom is capable of inducing a short replacement collision sequence in the Ni matrix, resulting in the formation of a stable Frenkel pair, then we obtain a defect-production rate of $\sim 6 \times 10^{-7}$ dpa/s. Under this condition, radiation-induced precipitation was observed after ~ 70 minutes of irradiation. On the other hand, by systematic irradiations with electron fluxes ranging from 10^{17} to 10^{20} e/cm²·s, we determined that a flux of $\sim 2 \times 10^{16}$ e/cm²·s was necessary to induce precipitation with 1-MeV electrons at 350°C in the same time period. For a realistic displacement threshold energy, between 20 and 24 eV, we get a defect-production rate of $\sim 10^{-6}$ dpa/s, in remarkable agreement with our estimate for 250-keV electron irradiation. Thus, for the same electron flux, the defect-production rate during 250-keV irradiation is about three orders of magnitude smaller than that obtained during 1-MeV irradiation.

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Figure Captions

- Fig. 1.** Microstructural evolution in a Ni-0.7 at.% Be alloy during 350-keV electron irradiation at 350°C. Electron doses: (a) 2.05×10^{22} e/cm², (b) 8.22×10^{22} e/cm², and (c) 1.43×10^{23} e/cm². $\hat{n} \sim [011]$ and $\hat{g} = [200]$.
- Fig. 2.** Energy dependence of the incubation dose for radiation-induced precipitation of a new phase in Ni-0.7 at.% Be alloys irradiated at 350°C.
- Fig. 3.** Radiation-induced segregation to dislocations during 350-keV electron irradiation at 450°C. Electron doses: (a) before irradiation, (b) 1.24×10^{22} e/cm² and (c) 7.09×10^{22} e/cm². $\hat{n} \sim [011]$ and $\hat{g} = [200]$.
- Fig. 4.** Radiation-induced segregation to dislocations giving rise to dislocation multiplication during 350-keV electron irradiation at 500°C. Electron doses: (a) 0.3×10^{22} e/cm², (b) 0.8×10^{22} e/cm² and (c) 2.12×10^{22} e/cm². $\hat{n} \sim [011]$ and $\hat{g} = [200]$.