

CONF-841157--4

RADIATION-INDUCED COMPOSITIONAL REDISTRIBUTION AND LOCAL PHASE TRANSFORMATION IN IRRADIATED ALLOYS*

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*This work was supported by the U.S. Department of Energy.

To be presented at the Materials Research Society Symposium, **ALLOY PHASE DIAGRAMS**, November 26-30, 1984, Boston, MA.

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RADIATION-INDUCED COMPOSITIONAL REDISTRIBUTION AND LOCAL PHASE TRANSFORMATION IN IRRADIATED ALLOYS*

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The relevance of radiation-induced phase instability to the engineering of materials for irradiation environments and the potential application of radiation-assisted modifications of subsurface alloy compositions have recently stimulated the search for a fundamental understanding of mechanisms and processes by which alloy phases are transformed during high-temperature irradiation.

Under thermal conditions, without irradiation, the stable state of an alloy is generally known with the aid of the existing phase diagrams or may, in principle, be predicted theoretically from thermodynamic criteria [1]. During irradiation, the situation is more complex; several processes can give rise to the appearance of unexpected phases: (a) radiation-induced disordering of ordered phases, (b) recoil dissolution of thermodynamically stable precipitate phase, (c) radiation-induced changes in free energies and, hence, in relative stability of phases, (d) radiation-enhanced diffusion which greatly increases the rate at which phase equilibria can be achieved in alloys, and (e) radiation-induced segregation (RIS) of alloying elements. Among these different processes, RIS has been found to have the most profound effect on the phase microstructure. This phenomenon gives rise to significant compositional redistribution, and can thus effectively translate local alloy compositions into different regions of the phase diagram.

In order for RIS to occur, it is required that there exists a preferential coupling of certain alloy elements to radiation-induced defect fluxes that persist in time. Since the migration of point defects, interstitials and vacancies, is always associated with the motion of atoms in the alloy, the total fluxes of point defects can be viewed as the sums of contributions from partial fluxes occurring via different alloy constituents. In general, the defect fluxes are not partitioned in the same proportions as the atom fractions of the alloy, i.e., interstitials may migrate preferentially via, say, A-atoms and vacancies may migrate by preferential exchange with, say, B-atoms in an alloy A-B. This is the physical origin of a preferential coupling between a particular alloying element and defect fluxes. If these fluxes are persistent, local concentration gradients will be set up and maintained during irradiation. Strong local solute enrichment or depletion can induce precipitation of a new phase whenever the solute solubility limit is exceeded or dissolution of a pre-existing phase, respectively. There are many causes for persistent defect fluxes. The most obvious one is the local elimination of radiation-induced defects at extended sinks such as dislocations, voids, grain boundaries and surfaces. Precipitation of a new phase at these sinks in a number of irradiated alloys has been observed; several reviews can be found in ref. 2. Defect trapping at local inhomogeneities, such as solute clusters and coherent interfaces that increase defect recombination, can also give rise to persistent defect fluxes [3]. Another cause is the spatial nonuniformity in defect production, e.g., during charged-particle irradiations [4]. Some recent observations of local phase changes in Ni-based alloys during ion bombardment and high-voltage electron-microscope (HVEM) irradiation are reported in the following.

*This work was supported by the U.S. Department of Energy.

In the case of ion bombardment, the rate of defect production by ions is spatially dependent and varies rapidly near the end of their range. Consequently, defect fluxes that provide the basic driving forces for RIS are produced not only in the vicinity of defect sinks, but also near the peak-damage depth where steep gradients of defect concentrations exist. The combination of defect fluxes near the surface and in the peak-damage region modifies the alloy phase microstructure in a peculiar fashion, as shown in Fig. 1 for 250-keV H^+ bombardment of a Ni-12.7 at.% Si alloy at 550°C [5]. The damage-rate distribution is indicated in the upper insert. After irradiation, pure Ni was electroplated onto the bombarded alloy surface, and transverse sections were cut out of the sample for TEM examinations (right insert). Persistent defect fluxes in the alloy generate net fluxes of Si atoms (Si has a preferential association with interstitials in Ni-Si alloys) to the bombarded surface and out of the peak-damage region into the mid-range and beyond-range regions, leading to strong Si enrichment at these locations. As a result, a γ' -Ni₃Si precipitate film was formed on the surface, and coherent γ' particles precipitated out in the mid-range and beyond-range regions at the expense of severe Si depletion in the peak-damage region. The same observations were also made in undersaturated Ni-Si solid solutions by Janghorban and Ardell [6].

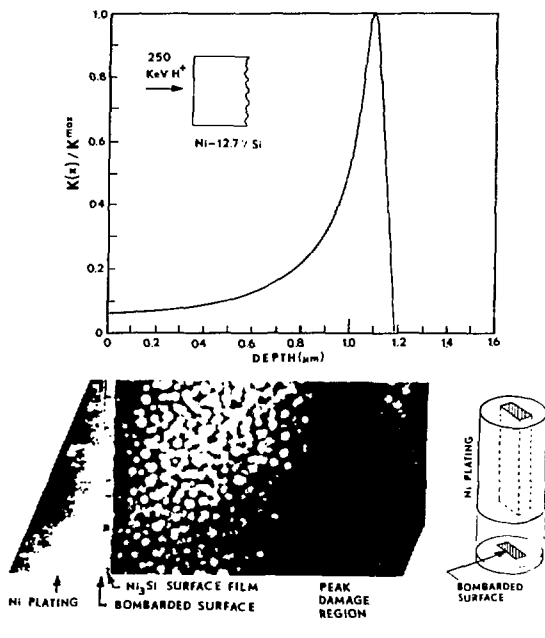


Fig. 1

Spatial variation of the phase microstructure in a Ni-12.7 at.% Si alloy bombarded with 250-keV protons at 550°C [5].

During in-situ HVEM irradiation, the radial nonuniformity in the point-defect production rate generated by highly-focused electron beams can also induce large defect fluxes in the radial direction, in addition to axial defect fluxes caused by defect annihilation at the film surfaces [7]. The radial defect fluxes can have a significant effect on the kinetics of RIS in alloy films. Depending on the coupling of solute atoms to defect fluxes, a net flux of solute into or out of the irradiated zone will be created, thus altering the average alloy composition in this region. For example, long HVEM irradiation will lead to severe Si depletion in the irradiated zone and Si enrichment in the surrounding unirradiated regions in Ni-Si alloys, and to strong Al enrichment in the irradiated volume at the expense of Al depletion in the surrounding regions in Ni-Al alloys in which the Al solutes segregate via a preferential exchange with mobile

vacancies. This Al enrichment can be extremely large, surpassing the Al solubility limit of Ni-Al solid solutions, leading to precipitation of coherent γ' -Ni₃Al particles in the irradiated zone. In two-phase Ni-12.7 at.% Al alloys, strong Al enrichment gives rise to segregation-induced coarsening of the γ' -precipitate phase in the irradiated region and dissolution of pre-existing γ' particles in the surrounding matrix, as shown in Fig. 2. The coarsened γ' precipitates were not distributed uniformly in the axial direction, but were concentrated near the midplane of the foil where the Al enrichment is greatest.

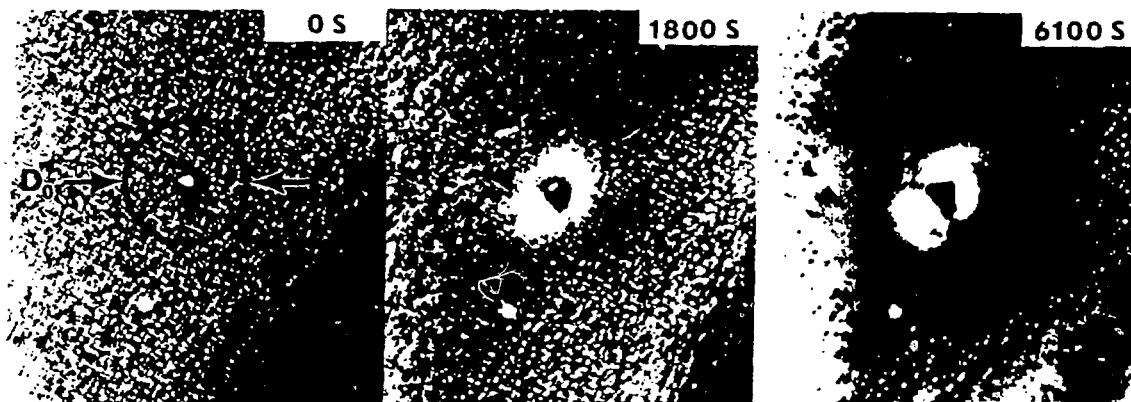


Fig. 2. Dark-field images showing the evolution of the phase microstructure in a two-phase Ni-12.7 at.% Al alloy irradiated with 1-MeV electrons at 700°C. The effective electron beam size is $D_0 = 0.64 \mu\text{m}$ (shown by the circle), and the irradiation times are indicated.

The drastic modifications of the alloy composition and, hence, local phase transformations during charged-particle irradiation, as experimentally demonstrated in the present work, must be taken into account in any assessment of radiation-induced property changes that are composition-dependent. Conversely, the ability and flexibility in locally modifying the nature, density and distribution of precipitates by irradiation will have potential applications in the development of novel materials.

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