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TITLE IRRADIATION-INDUCED AMORPHIZATION AND ELASTIC SHEAR INSTABILITY IN INTERMETALLIC COMPOUNDS

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SUBMITTED TO Proceedings of Symposium A, Beam-Solid Interactions: Physical Phenomena, MRS Fall Meeting November 27 - December 2, 1989, Boston, MA

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IRRADIATION-INDUCED AMORPHIZATION AND ELASTIC SHEAR INSTABILITY IN INTERMETALLIC COMPOUNDS *

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ABSTRACT

Previously we reported a substantial (~ 50 %) decrease in shear modulus prior to amorphization in Kr irradiated Zr_3Al , and proposed that amorphization is triggered when the crystalline lattice becomes unstable against shear stress. In the present work, the relation between amorphization and shear elastic instability has been investigated in two additional compounds (FeTi and NiAl) during room temperature irradiation with 1.7-MeV Kr^+ . A shear modulus was measured using Brillouin scattering; structural information was obtained in situ in a high voltage electron microscope interfaced to a tandem accelerator.

During irradiation of FeTi, chemical disordering and a large (~40 %) decrease of shear modulus were observed, and an amorphous phase developed subsequently. In contrast, NiAl remained crystalline and chemically ordered during irradiation, and exhibited only a ~ 10 % decrease in shear modulus. Hence, these two results provide further support that a shear instability triggers irradiation-induced amorphization. The shear instability mechanism may also apply to other solid-state amorphization techniques, e.g. hydrogen charging and mechanical deformation.

INTRODUCTION

Amorphous phases can be produced from crystalline materials by means of various techniques such as, for example, high energy particle irradiation, ion-beam mixing, ion implantation, hydrogenation, annealing of diffusion couples, and mechanical alloying. These solid-state techniques provide the possibility to produce bulk amorphous material. Amorphous phases have been demonstrated to improve surface and interface properties in metallic compounds and semiconductors. We have been studying irradiation-induced amorphization in intermetallic compounds to understand the driving force and underlying mechanisms [1-3].

In Zr_3Al irradiated with 1-MeV Kr^+ , a large decrease in shear modulus, chemical disordering and lattice dilation were all observed prior to amorphization. A linear relation [1] was found between the measured shear modulus and lattice dilation, similar to the Grüneisen relation due to the anharmonic thermal vibration of lattice atoms with increasing temperature [4]. Based upon several observed analogies to melting [5,6], the amorphization mechanism was suggested [1] to be a shear elastic instability of the disordered crystalline lattice.

In the present work, FeTi and NiAl are examined to see if the similar correlation exists. Both compounds have the B2 (ordered b.c.c.) structure, and exhibit a strong tendency to chemical order, as indicated by the fact that the ordered phases are stable up to the melting point in both materials [7]. However, it has been reported that FeTi is susceptible to amorphization under heavy ion irradiation at room temperature, while NiAl remains crystalline and chemically ordered under the same conditions [8]. The phase stability of these compounds therefore appears similar in equilibrium, but different under irradiation. This difference is examined by measuring chemical disordering, lattice dilation, and shear modulus as a function of irradiation

* The work supported by DOE W-31-109-ENG-38 and NSF DMR-8411178.

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dose. This comparison between FeTi and NiAl will be seen to provide further evidence that an elastic shear instability triggers solid-state amorphization.

EXPERIMENTAL PROCEDURE

Nominal alloy compositions of FeTi and NiAl were arc-melted, and TEM samples were prepared by jet-polishing of the thin disks spark-cut from the annealed alloy button. Samples were irradiated at room temperature with 1.7-MeV Kr ions in the Argonne High Voltage Electron Microscope (HVEM) interfaced to a tandem accelerator. The dose rate was 3.4×10^{11} ions/cm²/sec, which corresponds to a calculated displacement rate of 6.3×10^{-4} dpa/sec and 5.1×10^{-4} dpa/sec for FeTi and NiAl, respectively, assuming a sample thickness of 100 nm and a displacement threshold energy of 25 eV for both compounds. The calculated average projected range is 443 nm and 444 nm for FeTi and NiAl, respectively.

During irradiation in the HVEM, negatives of diffraction patterns were taken at appropriate dose intervals, and subsequently digitized by an optical microdensitometer. When diffraction patterns were taken, the diffraction condition suggested by Urban [9] was employed to minimize any error in the measurement of the chemical long-range order and lattice parameters. The sample orientation was adjusted in such a way that the diffraction pattern has a single row of diffraction spots, h00, with one side (200 spot) strongly excited, and the other (200 spot) in the kinematical condition. For both FeTi and NiAl, the lattice parameter was determined by measuring the distance between the transmitted beam and the 200 spot. The chemical long-range order parameter was taken to be the intensity ratio of the superlattice 100 spot and the fundamental 200 spot.

For the shear modulus measurement, samples of ~1mm thickness were irradiated to various doses (outside of the HVEM) under the same irradiation conditions. A shear modulus of the irradiated samples was measured using Brillouin scattering spectroscopy (see for example Ref.10). The samples were probed with a polarized Ar laser ($\lambda=514.5$ nm). The outgoing beam, which contains an inelastically scattered beam due to interaction with transverse lattice vibrations in the sample, was filtered with a 5+2 pass Fabry-Perot interferometer, and the energy spectrum was obtained with a multichannel analyzer. The velocity of the transverse wave was derived from the energy shift of the inelastically scattered beam with respect to the nonscattered beam. The shear modulus can then be calculated by multiplying the squared transverse velocity by the specimen density. The depth probed by this technique is < 300 nm, which is within the calculated average projected range. The calculated displacement rate for the Brillouin scattering experiment, averaged over the thickness of 300 nm, was 7.5×10^{-4} dpa/s and 6.2×10^{-4} dpa/s for FeTi and NiAl, respectively.

RESULTS

Fig.1 shows the selected area diffraction patterns from FeTi and NiAl irradiated at room temperature to 0.6 dpa and 0.3 dpa, respectively. FeTi is amorphized by the 1.7-MeV Kr⁺ irradiation, as evidenced by the appearance of a diffuse ring. As shown later (Fig.3(a)), the relative intensity of the superlattice spot to that of the fundamental spot decreases significantly before a diffuse ring appears, indicating that chemical disordering precedes amorphization. In contrast, no diffuse ring is observed in NiAl. The strong fundamental and superlattice spots (Fig.1 b) indicate that NiAl remains chemically ordered and crystalline. These results are consistent with previous observations for room temperature irradiation with 2.5-MeV Ni ions [8]. A corresponding bright-field image of NiAl is shown in Fig.2. The black contrast for areas which satisfy the Bragg condition is not continuous, as is usually observed in unirradiated samples. This indicates that the crystalline plane orientations vary locally on a fine scale, possibly due to defect aggregation or dislocation loop formation.

Fig.3 (a) shows the measured chemical long-range order parameter, S, plotted on a logarithmic scale as a function of dose. Only a slight decrease of S is observed in NiAl. On the other hand S of FeTi decreases very rapidly to 0.15, followed by amorphization at ~ 0.1 dpa. In the figure, only S values before the appearance of the diffuse ring are shown. Once the diffuse

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Fig.1 Diffraction patterns of (a) FeTi irradiated to 0.6 dpa, and (b) NiAl irradiated to 0.3 dpa.

Fig.2 Bright field image of NiAl irradiated to 0.3 dpa.

ring appears, significant scatter in S is observed, probably due to the overlapping effect of the diffuse ring with the crystalline spots.

Fig. 3(b) shows the measured lattice dilation, $3\Delta a/a_0$, plotted as a function of dose. Here a_0 is the lattice parameter before irradiation, and Δa is the measured change of the lattice parameter. In both compounds, lattice dilation increases rapidly with irradiation, and appears to saturate around 0.1 dpa. The maximum lattice dilation in FeTi is large ($\sim 6\%$) compared to NiAl ($\sim 2\%$). In the case of Zr_3Al , lattice dilation reached a maximum value of $\sim 2.4\%$ at 0.15 dpa, and subsequently decreased by a small amount (0.3%) when the onset of amorphization was observed. More accurate measurements would be necessary in the present work to examine this subtle behavior, although there is some evidence for an abrupt change in lattice parameter in both materials near 0.1 dpa.

Fig. 3(c) shows the variation of shear modulus with increasing dose. In FeTi, the shear modulus decreases by $\sim 40\%$ with irradiation. The dose at which the shear modulus saturates (~ 0.1 dpa) coincides with the onset dose for amorphization (indicated by an arrow on the abscissa). NiAl shows a decrease in shear modulus of $\leq 10\%$. A plot (not shown) of the shear modulus v.s. lattice dilation for both FeTi and NiAl reveals a linear decrease of modulus with increasing lattice dilation, as observed in Zr_3Al during irradiation, and in pure metals during heating to melting.

DISCUSSION

A strong similarity between the temperature dependence of amorphization and that of chemical disordering has been reported in Cu-Ti intermetallic compounds irradiated by electrons [11,12]. On this basis, chemical disordering was suggested as a major driving force for amorphization. In NiAl irradiated with Kr ions, a large number of secondary defects are formed as shown in the bright field image in Fig.2, but significant chemical disordering is not observed. FeTi undergoes chemical disordering, followed by amorphization. The differences

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Fig.3 Variation of the measured parameters of FeTi and NiAl as a function of irradiation dose; (a) chemical long-range order parameter, (b) the lattice dilation, and (c) shear modulus.

between FeTi and NiAl in amorphization tendency and chemical disordering are consistent with the previous results in Cu-Ti compounds, confirming that chemical disordering is a major driving force for irradiation-induced amorphization.

As originally suggested by Johnson [5] and Cahn and Johnson [6], the observed linear relationship between the increase in lattice parameter and decrease in shear modulus during irradiation is analogous to what occurs during heating to melting. Lattice dilation is observed during heating of most materials, and it is accompanied by a decrease in elastic moduli. The linear dependence of the bulk modulus on lattice dilation is known as the Grüneisen relation, and can be understood as the elastic energy increase generated by lattice dilation being compensated for by decreased elastic moduli [13]. Tallon [4] experimentally showed in many pure elements that the linear relation between lattice dilation and shear moduli during heating extrapolates to zero at a dilation less than that characteristic of the liquid. Tallon's results support the hypothesis proposed by Born [14] and Durand [15] that crystal melting is caused by an elastic shear instability. Ida [16] demonstrated theoretically that transverse atomic vibrations induce a large softening of the shear modulus, typically 40 to 70 % during heating to melting.

In FeTi as well as Zr_3Al , we have observed several parallels between heating-to-melting and irradiation-induced amorphization. That is, in both cases, a large decrease of shear modulus accompanied by lattice dilation is observed prior to the loss of long range crystalline order (amorphization). Recent molecular dynamics computer simulations of amorphization also showed similar behavior. Hsieh and Yip [17] calculated such parameters as, for example, the radial distribution function, coordination number, and mean square displacement of lattice atoms by interstitial insertion and the subsequent relaxation to simulate irradiation-induced amorphization. With increasing number of interstitials inserted, the mean square displacement increased, and the radial distribution function and coordination number became similar to those of an amorphous structure. Massobrio et al. [18] calculated the radial distribution function and the mean square atomic displacement during chemical disordering. They also found an increase in mean square atomic displacement, and eventual amorphization, during chemical disordering. Since an increase of mean square displacement (lattice dilation) produces a decrease of shear modulus, these theoretical studies are consistent with the present experimental results.

The present results, and these molecular dynamics calculations, have an interesting implication for other solid-state amorphization techniques. The calculation method to simulate irradiation effects in an intermetallic compound is different in the two studies (interstitial insertion in one, and chemical disordering in the other). Yet amorphization accompanied by a significant increase of mean square displacement was found in both cases. This implies that mean square displacement, and the concomitant lattice dilation, is the key factor underlying amorphization. Interstitial insertion and chemical disordering seem simply to be independent methods to produce lattice dilation.

In fact, Zr_3Al was amorphized by hydrogen charging without any discernible chemical disordering [19]. The total lattice dilation at which the onset of amorphization was observed by hydrogen charging was the same as in the case of Kr^+ irradiation-induced amorphization [1]. Another straightforward method to induce additional atomic displacement is to apply an external stress. In NiTi [20] and in GaSb [21], external stress applied under pressure produced an amorphous phase. More recently, cold rolling of NiTi at room temperature was also found to be successful in producing an amorphous phase [22].

SUMMARY

Irradiation-induced amorphization was studied in FeTi and NiAl with electron microscopy and Brillouin scattering. In FeTi, a large lattice dilation and shear modulus softening were observed during chemical disordering, which was followed by amorphization. NiAl remained crystalline, and much smaller changes were observed in the chemical long-range order parameter, lattice dilation, and shear modulus. The strong correlation observed between chemical disordering, lattice dilation, shear modulus softening, and amorphization suggests that solid-state amorphization is triggered by an elastic shear instability driven by lattice dilation and chemical disordering. Examination of theoretical models and other experiments on solid-state amorphization suggests that the shear instability mechanism may be applicable to several different solid-state amorphization techniques.

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