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Grain growth in Zr–Fe thin films during in situ ion irradiation in a TEM

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Abstract

In situ ion-beam irradiation was used to study irradiation induced grain growth in co-sputter-deposited Zr/*x*Fe ($0\% \leqslant x \leqslant 4.5\%$) nanocrystalline thin films. Samples were irradiated with 500 keV Kr ions to fluences in excess of 10^{16} ions/cm^2 (on the order of 80–100 dpa), at irradiation temperatures ranging from 20 K to 573 K. The average grain size increased monotonically with ion fluence until it reached a saturation value which depends both on temperature and on the presence of Fe. Similarly to thermal grain growth, the ion irradiation induced grain growth curves could be best fitted with curves of the type: $L^n - L_0^n = K\Phi$. Grain growth at 20 K is similar to that which occurs at 298 K. Above 298 K, the rate of grain growth increases with irradiation temperature.

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Keywords: Ion-beam irradiation; Grain growth; Thin film; Ion dose

1. Introduction

The understanding and ultimately the control of the grain microstructure of polycrystalline materials is of great technological importance, since properties such as mechanical strength, magnetic susceptibility, electrical conductivity, electro-migrative properties in semiconductor interconnects, etc. are functions of porosity, the average grain size and the grain size distribution. Irradiation induced grain growth has been observed in bulk materials and in thin films [1–4]. In these experiments, an irradiation dose is applied to the material from which a TEM sample is subsequently prepared to study grain growth. In this work we examine in situ grain growth in co-sputter-deposited Zr–*x*Fe ($0\% \leqslant x \leqslant 4.5\%$) thin films under ion-beam irradiation in the intermediate voltage electron microscope (IVEM) (Hitachi H9000NAR) at Argonne National Laboratory [5]. The advantage of an in situ study is the possibility of directly observing the phenomenon and its kinetics so the mechanism and driving forces can be understood in greater detail. This work focuses on better determining the effects of temperature on ion irradiation induced grain growth in thin films, especially at low temperature.

2. Experimental methods

2.1. Sample preparation

Supersaturated polycrystalline Zr–*x*Fe films ($0\% \leqslant x \leqslant 4.5\%$) were co-sputter-deposited onto NaCl substrates and onto Si wafers (for Rutherford Backscattering Spectroscopy (RBS) characterization) using a dual gun system at a base pressure of 9.5×10^{-7} Torr at room temperature (at the Materials Research Laboratory (MRL), Pennsylvania State University). The coated NaCl substrates were cleaved into small pieces and the specimens were floated on a de-ionized water–ethanol solution onto TEM copper grids, cleaned in de-ionized water, and dried before use.

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2.2. Microstructure of as-deposited films

Fig. 1 shows an example of the as-deposited films; these were ~800 Å thick, laterally homogeneous, and nanocrystalline, with an initial mean grain size between 15 nm and 20 nm. The diffraction patterns of the as-deposited films were characteristic of the hexagonal α-Zr phase (Fig. 1). Because of the rapidity of the effective quench during thin film deposition, no second phase formed although the RBS analysis confirmed an iron content in the Zr–Fe films beyond the solubility limit of Fe in Zr. The grains were preferentially oriented with the basal plane perpendicular to the foil surface, as verified by the high (002) diffraction ring intensity in Fig. 1(b).

2.3. Irradiation experiment

The thin films were irradiated at the IVEM-TANDEM facility at Argonne National Laboratory where an ion accelerator is attached to an electron microscope operated at 300 keV [5]. Samples were irradiated with 500 keV and 600 keV Kr^+ ions at fluxes typically around 2.50×10^{12} ions/cm^2 s, at temperatures ranging from 20 K to 573 K. The ion-beam energy was chosen on the basis of computer simulations using SRIM-2003 [6] so that ion-implantation is minimized in the films. The total displacement damage, calculated by SRIM-2003 using a displacement energy of 25 eV, was about 80 displacements per atom (dpa) at 10^{16} ion/cm^2, for both 500 and 600 keV Kr ions. Evolution of the microstructure was followed by sequentially taking images and diffraction patterns (DP) of the films while they were being irradiated. Individual grain sizes were obtained from direct measurement on the images. Beam heating calculations show that in the given configuration, with the sample holder in contact with the grid, itself in contact with the sample, the temperature rise in the sample was no more than 10–20 K. This increase is negligible in terms of grain-growth thermal activation processes.

3. Results and discussion

Direct observation of films under irradiation revealed a gradual increase of the average grain size (i.e. average grain diameter), for all ion energies, at all irradiation temperatures, even at 20 K where thermal effects are negligible. Fig. 2 shows a sequence of bright field electron micrographs of a pure zirconium film irradiated with 500 keV Kr^+ ions at 20 K taken at different ion fluences. The increase of grain size is apparent on the micrographs. The grain growth could also be detected on the diffraction patterns which became spottier as the fluence increased, indicating grain size increase. For each grain, the grain size was calculated by taking the average of the smaller diameter and the larger diameter measured on the TEM micrographs.

The evolution of the measured average grain sizes with fluence is shown in Figs. 3 and 4. For all compositions

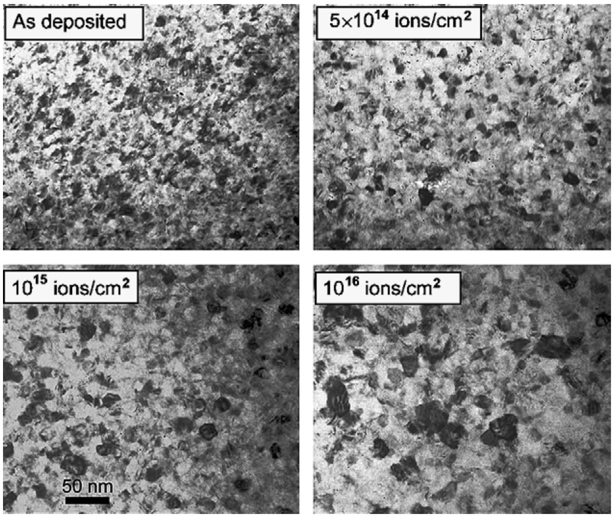

Fig. 2. Sequence of bright field TEM micrographs taken at different ion fluences (as indicated) during irradiation of a pure Zr thin film irradiated with 500 keV Kr ions at 20 K, showing grain growth induced by ion irradiation.

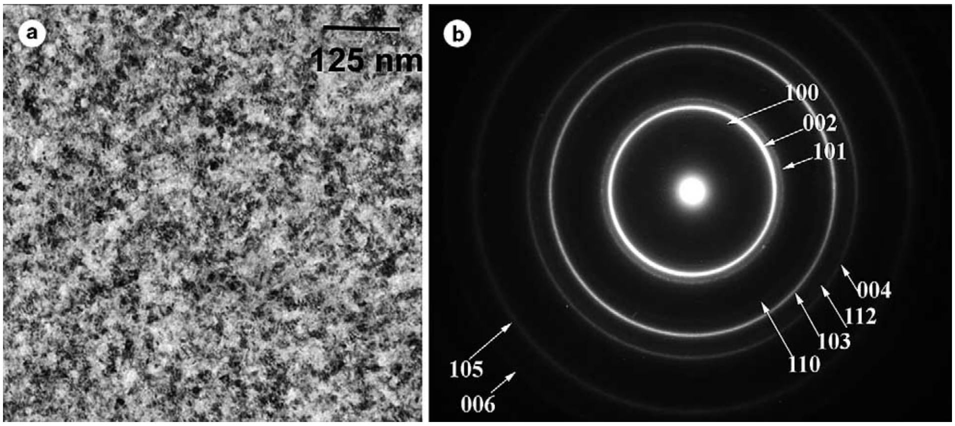

Fig. 1. Bright field (a) image of as-deposited Zr–Fe thin film and the corresponding diffraction pattern (b) showing well defined rings indexed as hcp Zr.

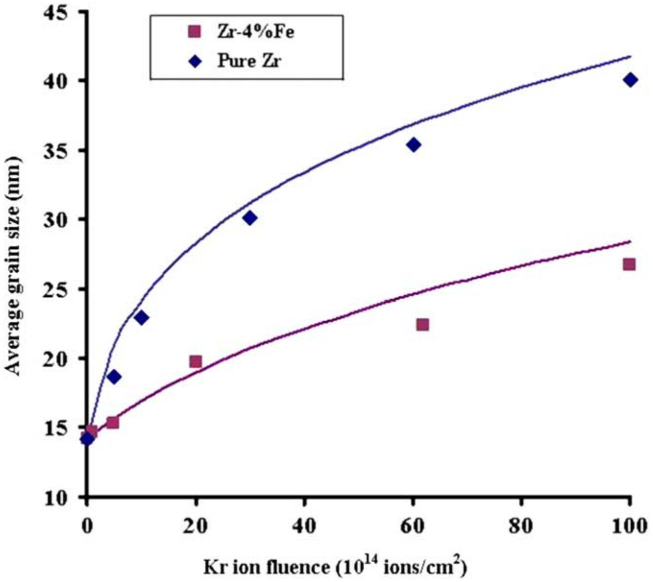

Fig. 3. Average grain size versus ion fluence for pure Zr and Zr–1.2%Fe irradiated with 500 keV Kr ions at 20 K.

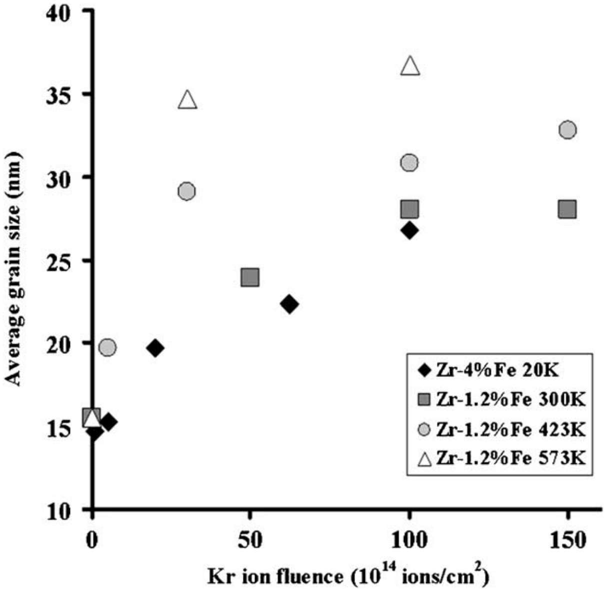

Fig. 4. Average grain size versus 500 keV Kr ion fluence in Zr–*x*Fe for irradiation temperatures between 20 K and 573 K.

considered and all irradiating temperatures, the average grain size increases monotonically with ion fluence until it reaches saturation. It is not clear what causes grain growth saturation. Possibilities evoked in the literature for grain growth saturation mechanisms include a solute or impurity effect, a cascade size effect, and the thickness of the thin foil [1,8]. Similarly to thermal grain growth [7], these curves could be fitted with curves of the type: $L^n - L_0^n = K\Phi$, where L_0 is the initial mean grain diameter, Φ is the ion fluence, and K and n are constants. Typical fits are shown in Fig. 3. The calculated values for n from the fitting curves are in the range of 3 to 4. Fig. 3 shows that irradiation induced grain growth occurs even at 20 K and both the rate of grain growth and the final saturation size are higher for pure Zr than for Zr–Fe.

The data in Fig. 4 shows that, below room temperature, the kinetics of ion-beam induced grain growth appear to be independent of temperature. This suggests that in this temperature range ballistic recoil effects of ion irradiation dominate the grain growth process. At temperatures above room temperature, the rate of grain growth and the final saturation grain size increase with temperature, suggesting that ballistic processes are supplemented by thermal processes. Other researchers have also seen temperature dependence of grain growth above room temperature, and explain the effect of irradiation in terms of increased defect concentrations which enhance normal thermal grain growth [2]. Also, in a study of Cu thin films, Liu found that the grain growth rates of the Ar^+ and Xe^{2+} irradiated thin films were independent of temperature for temperatures lower than 213 K and increased with temperature at higher irradiation temperatures [4]. These results are in agreement with the idea that there are two regimes of grain growth in thin-films under ion irradiation which is also supported by the present work. In the lower temperature regime, intra-cascade processes are likely responsible for the process, i.e. little thermal motion is required for grain growth. In the higher temperature regime thermal diffusion of irradiation-produced point defects influences the grain growth. The present work further supports the idea that, at lower temperatures, intra-cascade processes may be responsible for the process, i.e. little thermal motion is required for grain growth [4].

4. Conclusions

A study was performed of irradiation induced grain growth in Zr and Zr–Fe thin films by exposing free-standing thin film samples to an ion beam while observing grain growth in a transmission electron microscope. The results presented here indicate that grain growth occurs for all the irradiation temperatures studied. Grain growth was similar at 20 K and 298 K. This suggests that there are two temperature regimes of grain growth in thin-films under ion irradiation, the lower temperature regime being controlled by collisional processes and the higher temperature regime being affected by thermally activated processes.

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