ELSEVIER Nuclear Instruments and Methods in Physics Research B 175–177 (2001) 521–525

NIM B
Beam Interactions with Materials & Atoms

www.elsevier.nl/locate/nimb

Grain growth in Zr–Fe multilayers under in situ ion irradiation

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Abstract

Transmission electron microscopy (TEM) observations during in situ ion irradiation were used to study grain growth in free-standing Zr–Fe thin film multilayers at 25 and 300 K. Irradiations were performed with three different types of ions: 100 keV Ar, 300 keV Kr and 500 keV Xe ions to fluences of 3×10^{15} ion cm^{-2}. Grain growth during irradiation at 20 K occurs at a similar rate to that at 300 K. At both temperatures the grain growth rate was proportional to the total number of displacements, regardless of the ion used for the irradiation. We discuss these results in terms of two previous models for grain growth under irradiation.

PACS: 61.80.Jh; 61.82.Bg; 61.80.Az; 68.55.Jk
Keywords: Grain growth; Ion irradiation; Charged particles; Metallic multilayers

1. Introduction

Grain growth under irradiation is a topic of great interest, with applications to semiconductor processing, and to ion beam modifications of materials [1–4]. In general, irradiation acts to assist thermal processes. Models in the literature have focused on the role of thermal spikes and of point defects to explain the role of irradiation in assisting grain growth. Since experiments are usually conducted in bulk samples thinned after irradiation, it is difficult to determine directly how grains change with irradiation.

In this work we have examined grain growth in free-standing Zr–Fe metallic multilayers during in situ ion irradiation in the intermediate voltage electron microscope (IVEM) at Argonne National Laboratory, with the aim of better understanding the temperature and ion mass dependence of grain growth in metals. We discuss the results in terms of the previous work in the literature.

2. Experimental methods

Zr–Fe multilayer samples were prepared and characterized with Rutherford backscattering

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0168-583X/01/$ - see front matter
PII: S0168-583X(00)00644-3

(RBS) and Mossbauer spectroscopy (CEMS) at the Institute of Physics of the Federal University of Rio Grande do Sul, in Porto Alegre, Brazil. Metallic multilayers of Zr and Fe were prepared by vapor deposition onto a NaCl substrate (TEM samples), and oxidized Si wafers for the other characterization techniques in a Balzers UMS 500P dual source system at pressures of 5×10^{-9} Torr. Our most common multilayer composition was 64% Fe, with three Zr–Fe bilayers of respective thicknesses of 18 and 15 nm. The overall sample thickness was kept around 100 nm to make the multilayers electron-transparent at 300 keV. TEM samples were prepared by floating the multilayers in de-ionized water onto Cu grids. In the as-deposited state the metallic multilayers, both Fe and Zr, exhibit a grain size of 20–25 nm. The as-fabricated samples showed electron diffraction patterns characteristic of bcc-Fe and hcp-Zr.

Samples were irradiated in the IVEM at the Centre for Electron Microscopy at Argonne National Laboratory. This is a Hitachi 9000 microscope operated at 300 keV with an attachment that permits in situ ion irradiation of the sample [5]. The irradiation temperature can be controlled from 15 to 973 K. Samples were irradiated with three types of ions: Ar at 100 keV, Kr at 300 keV, and Xe at 500 keV, to fluences of up to 3×10^{15} cm^{-2} at currents ranging from 0.5 to 1×10^{12} ion cm^{-2} s^{-1}. Previous work has shown that after a dose of approximately 5×10^{15} ion cm^{-2} the individual grains start to be consumed by intermetallic compound phase formation [6]. In this work, we kept the ion fluences such that the

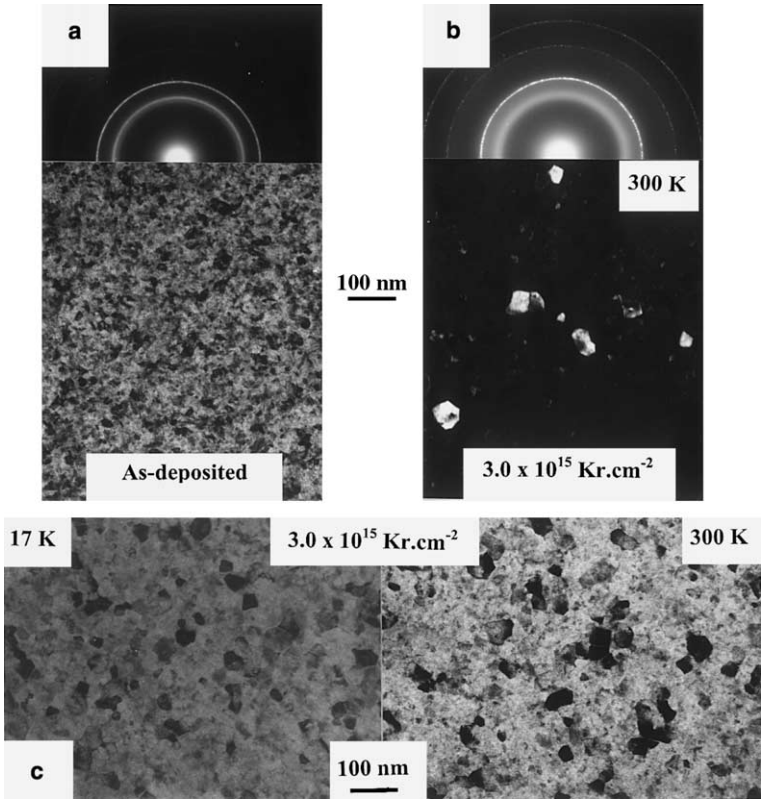

Fig. 1. (a) Bright-field micrograph and corresponding diffraction pattern for the as-deposited film. (b) Dark-field micrograph showing Fe grains after irradiation with 300 keV Kr ions at 300 K to a total fluence of 3×10^{15} ion cm^{-2}, with corresponding diffraction pattern showing grainy Fe rings. (c) Bright-field micrographs showing grain size after irradiation with 300 keV Kr ions at to a total fluence of 3×10^{15} ion cm^{-2}, at 17 K (left) and 300 K (right).

whole experiment was conducted in the pure growth region. Bright field (BF) and dark field (DF) micrographs were taken at regular intervals during the irradiation to monitor grain growth. Dark field micrographs taken from the Fe rings and from the Zr rings illuminate the respective sets of grains. The grain size distributions were then obtained by direct measurement from the images in the micrographs, for all the irradiation conditions conducted and the average grain sizes calculated.

3. Results

Grain growth occurred for the three types of ions used at both temperatures studied. Fig. 1(a) and (b) shows a pair of bright field and dark field micrographs from the Fe ring and corresponding diffraction patterns taken from films in the as-deposited state and after irradiation to 3×10^{15} ion cm^{-2} with 300 keV Kr ions at 17 K and 300 K. The increase in grain size is evident from the micrographs. As the fluence increases, the grain size increases continuously, up to the maximum fluences studied. It should also be noted that the bcc-Fe rings in the diffraction pattern grow increasingly more "spotty" as the fluence increases, (as can be seen in Fig. 1(b)), indicating that the Fe grain size is increasing. Fig. 2 shows typical grain size distributions for an irradiation conducted with Kr ions at 20 K, in the as-deposited state, after 6×10^{14} and 2.25×10^{15} ion cm^{-2}. Similar grain size distrubutions were obtained for the other ions and for the 300 K irradiation temperature..

Comparing the three types of ions, the highest rate of grain growth was seen in the multilayers irradiated with Xe, followed by those irradiated with Kr and then those irradiated with Ar. This is illustrated in Fig. 3, which shows the average grain size as a function of fluence for Xe, Kr and Ar ion irradiations conducted at 300 K. We fit the data with an empirical curve of the type

$$d^n - d_0^n = k\Phi t,$$

where d is the grain diameter, d_0 is the initial grain diameter, Φt is the irradiation fluence and k and n are constants. We found that the best fit was obtained with $n = 1$. We then derived values for the constant k for the three types of ions and the two temperatures studied. These are shown in Table 1. As expected, the rate of grain growth increased with ion mass, and the consequent availability of

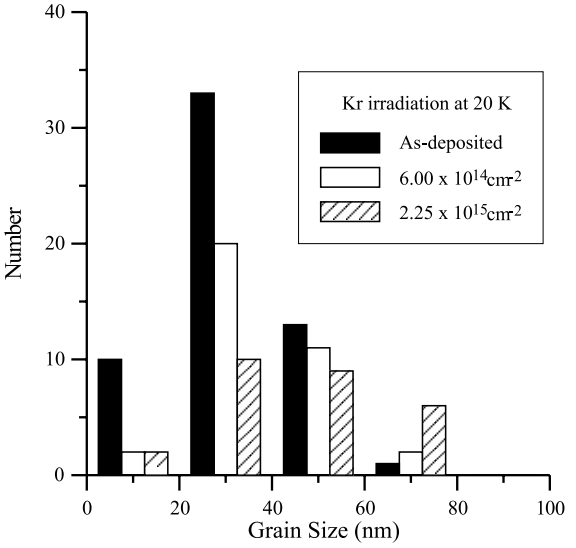

Fig. 2. Grain size distributions for an as-deposited Fe film and after irradiation with 300 keV Kr ions to 2.25×10^{15} ion cm^{-2}, conducted at 20 K.

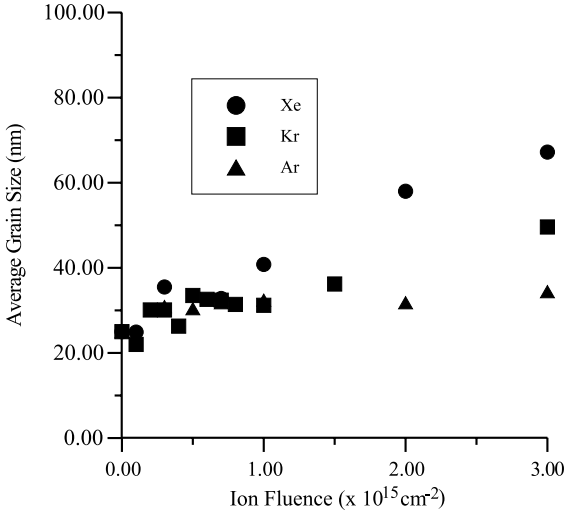

Fig. 3. Average Fe grain size versus ion fluence for irradiations conducted at 300 K for the three types of ions studied.

Table 1
Grain growth constant k for the irradiations conducted

	Irradiation temperature (K)	Growth constant k (nm^4)
500 keV Xe	300	1.50
	20	1.25
300 keV Kr	300	0.83
	20	0.69
100 keV Ar	300	0.38
	20	0.35

higher density cascades. Surprisingly, grain growth at 20 K was not noticeably slower than at room temperature, i.e., there was no noticeable temperature dependence of grain growth, as shown in Fig. 1(c). This suggests that there is no thermally activated process of grain growth in this system at room temperature and below.

In this work we adjusted the ion energies so that they would provide us with a relatively constant ion range, about equal to the size of the multilayers. Clearly, for a given ion fluence, as the ion mass increases, the amount of elastically deposited energy (and consequently the number of atomic displacements, dpa) increases. We used the TRIM 95 code with a displacement energy of 24 eV for both Fe and Zr to evaluate the number of displacements per ion/cm^2 for each of the ions used. We found that the displacement rates were $G_{Xe} = 7.4 \times 10^{-15}$ dpa/(ion/cm^2), $G_{Kr} = 4.2 \times 10^{-15}$ dpa/(ion/cm^2) and $G_{Ar} = 2.1 \times 10^{-15}$ dpa/(ion/cm^2), so that at 3×10^{15} ion cm^{-2} the total fluences in dpa were respectively 22.2, 12.7 and 6.2 dpa. These displacement rates are in almost exact proportion with the growth rates, so that when the grain sizes are plotted against dpa, they fall on the same line, as shown in Fig. 4, for the three irradiations plotted in Fig. 3. In summary the data show a clear proportionality to the elastically deposited energy fraction F_D.

4. Discussion

The results of this study indicate that the grain growth process in these multilayers has little

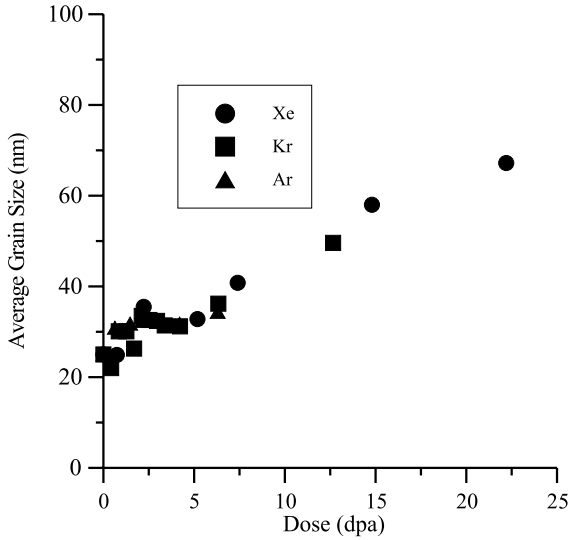

Fig. 4. Average Fe grain size versus dpa for irradiations conducted at 300 K for the three types of ions studied.

temperature dependence down to 20 K. Allen [7] observed a similar lack of temperature dependence of grain growth during Xe irradiation of Cu and Au films at 75 and 300 K. Since in that study grain growth was seen under both electron and ion irradiation at 75 K, this was attributed to the availability of mobile defects even at the lower temperature. If the results in the present study are to be explained by point defect mobility, there need to be mobile defects at 20 K. Other researchers have investigated the temperature dependence of grain growth in Cu films and found a strong dependence with temperature above 213 K [4], and little dependence below. The fact that in our study, grain growth was found to be essentially proportional to the number of displacements per atom, is in reasonable agreement with the lack of temperature dependence, since if the primary mechanism for grain growth is grain boundary motion effected by ballistic displacements across the interface, then the growth rate should be directly proportional to fluence.

It is interesting to briefly compare our observations with similar observations reported in the literature. Karpe et al. [8] conducted a study of Fe and Fe–5%Zr films, which they subjected to Xe and Ar ion irradiation at 300 K. They found that

the grain boundary mobility k was essentially proportional to F_D^2, in agreement with a thermal spike model [2], and in contrast to this study, where we found k proportional to F_D. They also found that the growth rates were higher in the Zr-containing Fe film than in the pure Fe film. Although it is difficult to compare the rates directly (particularly because they found that $n = 2$ was a better fit to their data), it is clear that the growth rates they found are much lower than the ones found in this study. It is possible that this is due to a chemical effect, i.e., as the Zr content increases, the growth rates in the Fe film increase. Alternatively, this could be due to the pronounced texture we observed in our films (especially in the Zr layers), which could cause the grain growth process to be easier, because of less grain misorientation in the more textured films.

5. Conclusions

We have conducted a study of grain growth in Zr–Fe multilayer films under in situ ion irradiation with three different types of ions and two temperatures. The main conclusions are as follows:

1. Grain growth was observed for the three types of ions studied at both temperatures used. The grain growth rate increased with ion mass, proportionally to the number of dpa caused, and was not very sensitive to irradiation temperature.

2. These results are in agreement with a grain boundary mobility model in which the grain growth is proportional to the fraction of elastically deposited energy F_D. The lack of temperature dependence suggests that the growth mechanism is primarily related to ballistic displacements across the grain boundaries.

Acknowledgements

The authors would like to thank Ed Ryan for his help in conducting the irradiations and Stan Ockers and Loren Funk of Argonne National Laboratory for their technical assistance. We would also like to thank S.R. Teixeira for furnishing the films used in this study. A. Paesano acknowledges a postdoctoral fellowship from the Brazilian National Research Council (CNPq) to work at Penn State. The authors gratefully acknowledge the support of this work by the National Science Foundation (NSF) and by the Brazilian National Research Council, CNPq.

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