IN SITU ION IRRADIATION/IMPLANTATION STUDIES IN THE HVEM-TANDEM FACILITY AT ARGONNE NATIONAL LABORATORY*

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The HVEM-Tandem User Facilty at Argonne National Laboratory interfaces two ion accelerators, a 2 MV tandem accelerator and a 650 kV ion implanter, to a 1.2 MV high voltage electron microscope. This combination allows experiments involving simultaneous ion irradiation/ion implantation, electron irradiation and electron microscopy/electron diffraction to be performed. In addition the availability of a variety of microscope sample holders permits these as well as other types of in situ experiments to be performed at temperatures ranging from 10–1300 K, with the sample in a stressed state or with simultaneous determination of electrical resistivity of the specimen. This paper summarizes the details of the Facility which are relavant to simultaneous ion beam material modification and electron microscopy, presents several current applications and briefly describes the straightforward mechanism for potential users to access this U. S. Department of Energy supported facility.

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1. Introduction

Established in 1981 by the U. S. Department of Energy as a User Facility, the High Voltage Electron Microscope–Tandem Ion Accelerator Facility (HVEM–Tandem) is a user-oriented resource for materials research, located at Argonne National Laboratory. The Facility has been described in detail at previous presentations in the Denton Conference series [1,2], and thus the emphasis of this paper will be on recent updates within the Facility . In brief the Facility consists of a modified Kratos/AEI HVEM with electron accelerating potentials ranging continuously from 0.1–1.2 MV, interfaced to a 2 MV tandem accelerator and to a 650 kV ion implanter-type accelerator, both manufactured by National Electrostatics Corporation. This combination of instruments offers capability, unique in the western hemisphere, for a wide range of in situ experiments involving electron irradiation, ion irradiation and ion implantation with simultaneous electron microscopy and electron diffraction. In fact during 1987 approximately 75% of microscope time was devoted to this type of experiment (Fig. 1) which included studies of solid state phase transformations such as amorphization, of radiation damage and defect structures, and of the effects of implantation of noble gas and metal ions.

In situ experiments of various types account for nearly 90% of usage of the HVEM. In addition to the radiation effects studies, this usage includes experiments performed in the microscope involving deformation, annealing and environmental effects. In this regard, the degree of versatility is determined largely by the availability of a variety of specimen stages which allow experiments to be performed involving application of high or low temperatures or of stress, reaction in controlled atmospheres (environmental cell) and simultaneous measurement of some physical property such as electrical resistivity. Specifications for the available sample stages are summarized in Table 1. The X-tilt limits listed arise from the stages themselves obscuring the electron beam; in most cases the manufacturer's specification is actually $\pm 60^{\circ}$ [1]. The deformation and environmental effects studies include environmental effects on fracture of metals and composites, oxidation of alloys, catalyst microstructure development, growth of carbon fibers, defect microstructures during plastic deformation and environment-induced phase transformations in ceramics. Unfortunately, however, design of the present environmental cell is not compatible with in situ ion irradiation/implantation, although there is strong interest in the materials community in development of such capability. Design of an environmental cell which would admit ion irradiation is under serious consideration.

The majority of users of the HVEM-Tandem Facility are faculty and graduate students of universities in North America, Europe and Japan. In some instances these users collaborate with Argonne staff scientists and engineers; such collaboration, however, is by no means a requirement for use of the Facility. The mechanism for access to the Facility is quite straight forward. Instrument time is assigned to all users on the basis of brief research proposals which are reviewed by a committee composed of Argonne and other scientists representing the materials community. Prof. John Silcox of Cornell University

currently chairs this committee. Use of the Facility is generally free of charge, provided studies are not proprietary in nature.

2. Ion beam studies in the HVEM

Currently the subjects of in situ ion and electron irradiation effect studies include the following:

(1) Solid state phase transformations in ceramics and intermetallic compounds, including amorphization.
(2). Radiation damage and defect structures in metals, including superconductors, and intermetallics.
(3). Irradiation-induced dimensional instability in intermetallics.
(4). Ion implantation-induced modification of grain boundary structures.
(5). Ion irradiation-induced grain growth.

The specific research projects constituting these areas involve research groups from eleven universities as well as from Argonne. We will attempt to summarize broadly the results from several of the areas.

Irradiation-induced grain growth

In general the phenomenon of ion irradiation-induced grain growth has been studied extensively in the past several years in a variety of pure metal thin films including Cu, Ni, Au, Pt, Ge and Pd and a few alloys including several in the Cu-Ni system. In situ grain growth experiments have been initiated at the HVEM-Tandem Facility by three research groups in 1988 and have concentrated on 1 MeV Kr and 1.5 MeV Xe on evaporated and sputtered thin films of Au and Pt. Temperatures in these experiments have spanned the range 10–450 K. The salient features of this irradiation-induced phenomenon include a very slight temperature dependence of growth rate over the entire temperature range and an apparent saturation grain size after doses typically in the 10^{19} m^{-2} range, the final grain size depending on material and on impinging ion mass. Theories which have been proposed assume that only the population of point defects created at grain boundaries cause boundary migration; the principal difference in the theories seems to be whether or not there is a saturation grain size after sufficient dose or just a continuously decreasing growth rate. Unfortunately, the theories imply that all grains grow.

The results of in situ HVEM observations indicate that there is a component of growth associated with grain boundary migration as predicted by the theories; however, the observations also demonstrate the important contribution of grain coallescence in the net growth process. That is, dynamic orientation fluctuations of grains in a given neighborhood eventually result in the elimination of a boundary between two neighbors as their orientations come into coincidence. This is demonstrated most dramatically by observing the same area of sample in dark field during ion irradiation. Furthermore the significant activity of the irradiation-induced dislocation population suggests that these orientation fluctuations probably involve dislocations as well as point defects being absorbed by boundaries. The dislocations resulting from collapse of point defect aggregates thus may achieve by glide a result at a boundary which is as

effective in the average grain growth process as the migration of the point defects might have been. The importance of the role of dislocations in this grain growth process is supported by the lack of a significant temperature dependence, and also by preliminary results of electron irradiation experiments which indicate a very different microstructure development with electron irradiation and, in fact, little or no net grain growth.

Fundamental amorphization studies

The subject of electron and ion irradiation induced amorphization of intermetallic compounds has been one of strong interest and activity for several years. Rather than emphasizing the purely structural aspects of a resultant amorphous or glassy state, the research in this area focusses on various aspects of the question, what are the fundamental reasons for the significant differences in behavior of various intermetallics when subjected to high energy electron and ion irradiation. Significant insight on this complex question has evolved especially over the past two years from studies correlating long range order parameter, lattice parameter and degree of amorphization as determined in the HVEM with determinations of shear elastic constant as determined from Brillouin scattering measurements on irradiated films. Typically when complete amorphization can be achieved, as in Zr_3Al under 1 MeV Kr irradiation for example [3], the effect of irradiation is to cause both the order parameter and adiabatic shear constant to continuously decrease with dose as lattice parameter increases until the appearance of the first vestage of amorphization corresponding to an order parameter around 0.2 and a shear constant approximately 50 percent of its initial value. The shear constant then increases rapidly by about 20 percent, as the order parameter goes to zero, and remains constant as the amorphization process procedes in the disordered matrix. For Zr_3Al extrapolation to zero of the shear elastic constant as a function of interatomic spacing yields a spacing typical of that for the supercooled liquid state. The decreasing resistance of the crystalline lattice to elastic shear preceeding amorphization is remaniscent of the mode softening phenomenon which is a common prelude to martensitic transformations.

Early stages of the amorphization process are under study in A15 superconductors by observation of general microstructure including cascade-induced defects at low temperatures. For example, in situ HVEM experiments have been conducted with Nb_3Sn at 12–30 K [4]. For 50 keV Xe irradiation at 13 K, at low doses (2×10^{15} m^{-2}) where individual cascades are well separated, features in black-white contast appear when imaged in fundamental reflections but in black dot contrast in superlattice reflections. Such features are consistent with disordered zones having a different intrinsic density. At higher ion doses (2×10^{16} m^{-2}) where significant cascade overlap is present, defect density and image contrast appear unexpectedly large; this is evidently associated with the formation of amorphous zones in the domains of cascade overlap. The effect of 1.5 MeV Kr at 15 K is to eliminate evidence of the martensitic microstructure by 5×10^{16} m^{-2}. Not until a dose of 3.5×10^{17} m^{-2} was the initiation of amorphization observed, however.

Consequences of amorphization and noble gas implantation

In addition to the strong interest in the basic origins of ion-induced amorphization of materials, several studies are underway in which phenomena are observed which are a direct consequence of amorphization or noble gas implantation. One of these is an in situ study involving ion beam simulation of the extraordinary swelling phenomenon which occurs in certain low enrichment reactor fuel candidates [5]. Uranium-based intermetallics are being developed as low-enrichment or high-density reactor fuel materials. However, the irradiation performance of several alloys with high uranium content, such as U_3Si and U_6Fe, suffers from the formation of extraordinarily large voids (on the order of several μm) at low or medium fuel burnup. This catastrophic swelling prohibits the use of the highest density material as reactor fuel. Similar alloys with somewhat smaller uranium concentration, such as U_3Si_2, develop a fine and stable dispersion of fission-gas bubbles (on the order of 10 nm), and do not exhibit catastrophic swelling. In order to understand this drastic difference in behavior, the effects of burnup have been simulated by irradiation and implantation with 1 MeV Kr ions in the HVEM.

Below 550 K, crystalline U_3Si becomes amorphous at very low Kr fluences. Subsequent irradiation of the amorphous U_3Si at 470 K results in the rapid growth of the initial specimen perforation in a manner similar to the rapid swelling observed during reactor burnup. Detailed examination during irradiation reveals that microvoids and void coalescence result, presumeably due to residual stresses resulting from differential damage . On the other hand, irradiation of the crystalline material at 620 K produces no growth. The rapid swelling at 470 K is not typical of growth due to fission-gas bubbles; rather, it is due to plastic flow of the amorphous material away from internal surfaces. Such flow does not occur in the crystalline phase. Furthermore the crystalline U_3Si may be stabilized against amorphization at the lower temperature by implantation of Kr into the crystalline phase at temperatures above the amorphization limit. The U_3Si initially exhibits a martensitic structure which is eliminated by 1 MeV electron irradiation to several dpa.

A second study involves several alloys in the Ti-Cr system [6,7]. The intermetallic $TiCr_2$ is amorphized by 0.4 MeV Kr irradiation at temperatures below 250 K. For ion doses of order 10^{18} m^{-2} the amorphous phase crystallizes on heating at about 250 K to a metastable structure (bcc). For ion doses of order 10^{20} m^{-2}, however, the amorphous phase is stabilized against crystallization to temperatures of 900 K or higher. As Kr is implanted into Cr+46 at % Ti coevapoated thin films which are amorphous as deposited at room temperature, a fine dispersion of crystalline Kr precipitates forms around which fine crystalline particles (1-2 nm; bcc) appear in the amorphous matrix at the particle-matrix interfaces. When the Kr crystals grow to some critical size, they melt, and the bcc matrix particles disappear from the neighborhood. These results illustrate two points; first, the chemical stabilization of the amorphous phase by the implantation of roughly 1 at % of Kr into $TiCr_2$, and second, in the alloy with larger Ti content, the destabilization of the amorphous phase in the presence of a crystalline seed, even one of the highly deformable van der Waals solid, fcc Kr.

3. Concluding remarks

The number of applications of the HVEM-Tandem Facility in in situ ion beam related materials research has steadily grown since dedication of the Facility in 1981, as the materials community has become increasingly aware of the truly unique potential of this user-oriented operation. As illustrated by the brief summary of some of the research performed in the Facility, the user emphasis is still on irradiation damage and the consequences thereof. However, with the 650 kV ion implanter on line, we can expect to see increasing interest from potential users in doing in situ experiments involving ion implantation of metals and semiconductors.

For additional information about the HVEM-Tandem Facility and the procedure for accessing its capabilities, prospective users are invited to contact the HVEM-Tandem Facility Operations Manager [(312) 972-5222]. There is no charge for use of the Facility for materials research of a non-proprietary nature.

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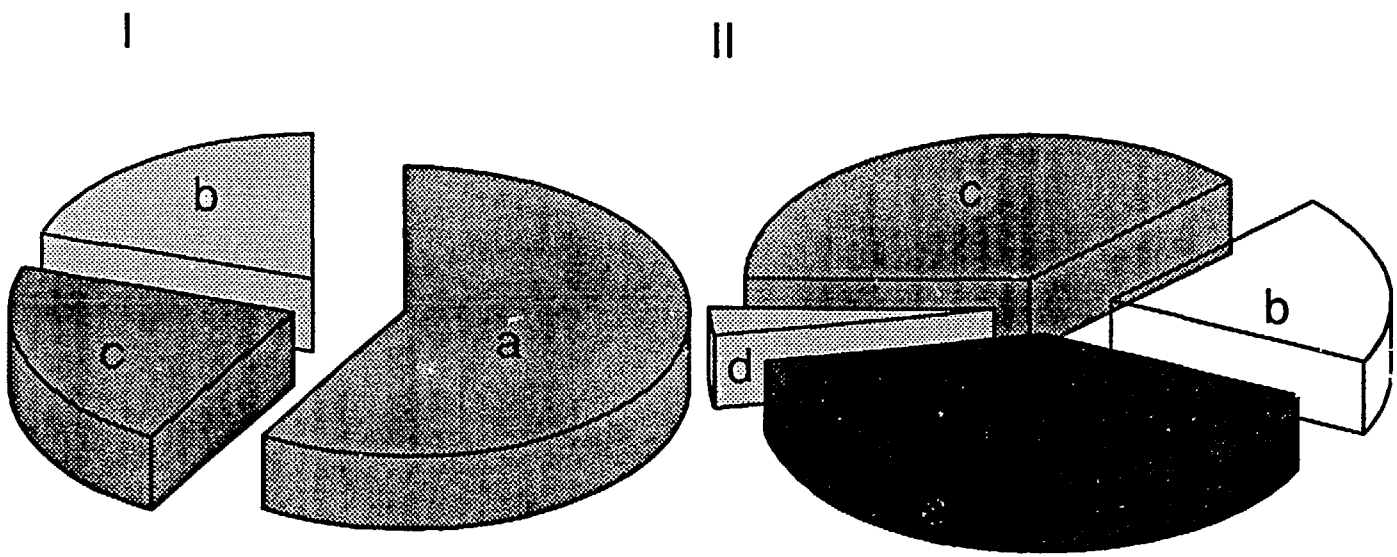
Table 1. Specimen stages for the Argonne HVEM [a)]

Stage Type	X-Tilt	Y-Tilt	Temperature (K)
Single-tilt	±45	--	300
Double-tilt (Cartridge-type)	±45	±30	300
Double-tilt , Heating	±30	±30	300–1100
Double-tilt , Nitrogen	±45	±30	78–300
Single-tilt, Helium, Electrical Resistivity [b)]	±45	--	10–300
Double-tilt, Helium, Electrical Resistivity	±45	±30	10–300
Double-tilt, Helium	±45	±30	10–300
Double-tilt, Straining [c)]	±30	±30	300
Single-tilt, Heating [d)]	±45	--	300–1100
Single-tilt, Heating (Ribbon-type) [d)]	±45	--	300–1300
Single-tilt, Straining, Heating	±45	--	300–950

a) All stages are compatible for ion irradiation
b) With two additional electrical leads
c) Birmingham stage
d) Compatible with environmental cell

Figure caption

Fig. 1. Distribution of usage of the HVEM-Tandem Facility during 1987. I: By experimental type. a: Irradiation effects. Universities: 40%; ANL: 35%; total: 75%. b: In situ deformation and environmental effects. Universities: 12%; ANL: 1%; total: 13%. c: Microstructural characterization. Universities: 8%; ANL: 4%; total: 12 %. II: By user type. a: ANL/university graduate research: 38%. b: Universities: 19%. c: ANL: 40%. d: Industrial and other national laboratories: 3%.


I
b
c
a
II
c
b
d