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**CURRENT RESEARCH USING THE ANL HIGH VOLTAGE ELECTRON MICROSCOPE-
TANDEM ACCELERATOR FACILITY***

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CURRENT RESEARCH USING THE ANL HIGH VOLTAGE ELECTRON MICROSCOPE-
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SUMMARY

Recent work at the Argonne National Laboratory, (ANL), HVEM - Tandem Accelerator user facility is summarized: Direct observation of cluster defects formed by in-situ ion irradiation at low temperature using the ion-beam interface has led to important fundamental results on defect production processes. Results on solute segregation at elevated temperatures induced by electron and ion irradiation are reported. Other published work is briefly summarized and/or referenced.

INTRODUCTION

A description of the ANL HVEM-Tandem Accelerator Facility together with system performance was presented at the Denton Conference in 1982 [1]. Operational experience with the system has shown that ion beam current densities of 10 microamp/cm^2 can be delivered to sample in the HVEM. This represents an order of magnitude increase over design aim. This improvement permits complete ion implantations at medium dose levels to be observed in-situ. The skim cup used to monitor the ion beam inside the HVEM that was described earlier has been upgraded, the new configuration is shown in Fig. 1. This is a combined skim cup and movable Faraday cup. The device permits continuous monitoring of the ion beam and thereby improves the

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accuracy of measurements of fluences delivered to the specimen. A double tilt He cooled specimen rod for the HVEM will be tested shortly. This will permit wider selection of imaging conditions than is presently available with the single tilt stage [2].

A summary of recent radiation damage related work is given in Table I. This work includes measurements of the energy to form single Frenkel defects at low temperature, cluster damage production by heavy ion irradiation, and defect-solute interaction at elevated temperature. The work which has resulted in important new findings has direct relevance to the understanding of radiation damage in reactors, ion implantation processes, and sputtering behavior. Table 2 contains a list of work mostly in progress and is included to illustrate the wide variety of studies being carried out at this unique Department of Energy, national user facility.

RADIATION DAMAGE STUDIES AT LOW TEMPERATURES

The work of King et al. [2] on damage production by electrons in fcc metals has led to a reevaluation of the Frenkel pair threshold energy damage surface in copper crystals and to the determination of more realistic damage functions for copper and nickel polycrystals [3] in the low energy knock-on regime. The new damage functions correct the commonly employed "Modified Kinchin and Pease" formula [4,5] which considerably overestimates the number of defects formed in the low energy regime.

Several groups of workers, see Table I, are now exploiting the capability to carry out in-situ ion irradiations at low temperatures where vacancies are immobile in order to measure the structure and formation efficiencies of heavy ion induced cascades and to determine under what circumstances their vacancy rich cores collapse into dislocation loops. A brief review of this

work was recently presented at the SM²IB Conference at Heidelberg, September 1984 and will be published soon [6]. In the case of pure metals, the dislocation loops at the collapsed cascade sites are visible and the defect formation efficiency must be obtained from measurements of the ion fluence incident upon the specimen foils. However, in the case of single crystal foils formed from ordered Cu₃Au alloy [7] extensive study of contrast and imaging conditions has shown that it is possible to choose imaging conditions that render visible the disordered zones formed at all cascade sites and to distinguish those sites that have collapsed into dislocation loops. The method is therefore absolute and can also be used to calibrate the dosimetry system. An example of a complementary pair of images is shown in Fig. 2. The photomicrograph (a) is a dark field image using the superlattice reflection $g = \{110\}$, and shows the displacement cascade through structure factor contrast, while micrograph (b) is a bright field kinetic image using $g = \{220\}$ showing the dislocation loop through strain contrast. Both images were taken at low temperatures following 100 keV Ar⁺ irradiation at 10 K and in this micrograph all disordered zones have collapsed. The number density, size distribution, and structure of the cluster defects have been examined as a function of particle mass, fluence, and energy, and important new findings have emerged. In general, it is found that in fcc Cu, Ag, Au, [8] and Cu₃Au, the defect formation efficiencies are high at low temperature where vacancy motion is frozen-in, are linear with ion dose, and independent of energy above 50 keV. Thermal annealing to room temperature does not increase the number of loops following low temperature irradiation, but in the case of Cu₃Au there is a systematic $\approx 20\%$ increase in defect efficiency when the irradiation temperature is raised to 300 K. The results show that the vacancy rich cores of the cascades collapse "athermally" during the thermal spike phase of cascade

formation i.e., during the initial 10^{-10} - 10^{-12} seconds. A detailed interpretation of the Cu_3Au data based on the thesis work of Black will be published soon.

In contrast with the above fcc metals, the cascade collapse behavior of iron [9] when irradiated at 40 K with self ions exhibits a dose dependence exponent of $3/2$, and a threshold dose of 8×10^{12} particles/cm². Threshold doses of similar magnitudes are also observed in nickel [10]. The results indicate among other things that cascade overlap is necessary to cause cascades to collapse into dislocation loops, confirming a postulate made by Dunlop et al.[11] to explain their resistivity data for neutron irradiated iron.

RADIATION DAMAGE STUDIES AT ELEVATED TEMPERATURES

In-situ electron irradiations of reactor materials in the HVEM is an important tool in the study of radiation damage in the void swelling regime of technological important Fe-Ni-Cr based alloys. However, because of differences between the displacement damage production in bulk samples irradiated in the reactors, and thin foils irradiated with focused electron beams in the HVEM, the correlation between data has proved difficult. Because of the need to understand the damage mechanisms in both nuclear and fusion reactors, further work is necessary. Groups working at the ANL facility using model alloys have recently demonstrated two important results impacting on such work. In the first study Régnier and co-workers [11,12] have shown that in alloys composed of elements with widely differing Z's, radiation damage can occur below the defect formation thresholds for the solvent elements. In the second study, Okamoto et al. [13,14] have quantified the redistribution of solute elements in electron and proton irradiations where there are steep gradients in the defect production profiles.

In the work of Régnier and co-workers, it is shown that atomic displacement of the heavier solvent element can occur through energy transferring collisions from the lighter solvent. In the case of electron irradiations made in-situ in the HVEM on Pt-C [11] and Ni-Be alloys [12], electron energies well below the energy to form Frenkel pairs in the Pt or Ni, result in visible effects. In Fig. 3, it is shown that an electron dose of $7.1 \times 10^{22}/\text{cm}^2$ at 350 kV delivered to a sample at 450 C results in both dislocation climb and segregation of Be to the internal sinks.

In alloys, irradiation can result in significant mass transport of solute elements through what is termed "radiation induced segregation". Solutes with significantly differing atom size to the solvent may be transported through coupling to the internal fluxes of point defects characteristic of the irradiation field. The flux of oversize solutes, J_s , moves in the opposite direction to the defect flux, $-J_d$, whereas undersize solutes may move in the same direction, i.e., $+J_d$. From Fick's law, it can be seen that the local enrichment or depletion of solute C_s is determined by the sense and magnitude in the curvature of the defect concentration profile, C_d viz

$$\frac{\partial C_s}{\partial t} = \mp \nabla J_s \propto \pm \nabla^2 C_d$$

Some examples where the distribution of solutes after irradiation have been successfully compared with computer calculations are given in Figs. 4, 5, and 6. The microstructure of a Ni-12.7 at/o Si alloy irradiated at 500 C with 250 keV protons and sectioned perpendicular to the beam direction is shown in Fig. 4 together with the calculated damage rate, defect concentration, and accumulation $\nabla^2 C_d$ versus depth. The undersized solute, Si flows out of the peak damage zone giving rise to a depletion of Ni_3Si precipitates in the

region of intense damage. Defects escaping to the free surface transport solute and cause the formation of a surface film. By contrast, in the cases of electron irradiations of foils in the HVEM, the axial dose rate is uniform whereas strong gradients exist in the radial or lateral directions, Fig. 5 lower left. The structures developed in a solid solution Ni-10 a/o Al alloy irradiate to 700 C with 1 MeV electrons at a dose rate of 10^{-3} dpa/sec is shown in the upper four micrographs. Coherent precipitates, visible because of strain contrast are seen initially, while incoherent Ni_3Al are formed later as in the micrograph for 48 minutes irradiation. The aluminum, an oversize solute, segregates both axially and radially, with the axial segregation dominating at short times. The calculated concentration at the center, shows that the Al concentration exceeds the solubility limit only when the slower radial diffusion processes begin to dominate. Blocky Ni_3Al precipitates can then be seen in the dark field image (lower right). Figure 6 is a comparison of the development of the microstructure as a function of dose for a 12.7 a/o Al alloy and a 10 a/o Ge alloy (an undersize solute). In the Ni-Al case solute flows from an annular region set by the inflection points of the damage rate profile of the electron beam, shown Figure 5 lower left. A dense precipitate core is formed at the center of the beam profile. Figure 6 right shows that for the undersized Ge, axial gradients produced by the flow of defects to the foil surfaces initially cause precipitation to occur at the upper and lower surfaces, but as the solute is gradually drained into the foil by radial diffusion these precipitates redissolve finally depleting the central region of the radiation damage zone of Ge solute.

OTHER RESEARCH

In Table II work on ion implantation, amorphization and radiation effects in oxides are tabulated. Okamoto, Averbach, and Stritzker [15] have demonstrated that when the thin bilayer films of Cu and Bi are mixed by ion bombardment at liquid He temperatures, a metastable amorphous film is formed with a super conducting transition temperature of 5°K. Donlon, McCune, and Mitchell [16] have found that in high dose nitrogen implants of aluminum, nitrogen bubbles but not nitrides are formed.

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FIGURES

Fig. 1. Column of HVEM showing details of the internal skim cup and movable Faraday cup.

Fig. 2. Electron micrographed Cu_3Au foil irradiated with 100 keV Ar^+ ions $\lesssim 40\text{K}$.

(a) Dark field image showing disordered zones at sites of all ion impacts.

(b) Corresponding bright field image showing dislocation loops formed by collapse of vacancy rich cores at the cascade sites.

Fig. 3. Beryllium segregation to dislocations at electron energy below displacement threshold of Ni. (a) Before irradiation,

(b) $1.24 \times 10^{22}\text{e}/\text{cm}^2$, (c) $7.1 \times 10^{22}\text{e}/\text{cm}^2$, $\vec{h} \sim [011]$, $\vec{g} = [200]$.

Fig. 4. Showing the correlation between the solute accumulation profile V^2Cd which was calculated from the damage rate productions curve for 250 keV protons incident on Ni-12.7 a/o Si (upper) and the distribution of Ni_3Si precipitates observed following irradiation at temperature 500 C (lower).

Fig. 5. Illustrating the influence of strong defect gradients produced by in-situ 1 MeV e^- irradiations on the axial and lateral flow of solutes in Ni-10 a/o Al alloys at 700 C.

Fig. 6. Micrographs showing contrasting solute redistribution effects during 1 MeV e^- irradiation at elevated temperatures. Left column, Ni-12.7% Al alloy in which solid solute and defect flows are opposed and for Ni-10 a/o Ge (right column) where flows are parallel.

Table 1. Fundamental Studies on Defect Production.

Study	System		Workers
	Ion	Target	
Damage Threshold Surface	e^-	$Cu.^{\dagger}$	King, Merkle, Meshii [2]
Damage Function Determination	e^-, p^+	$Cu.^{\dagger}$ $Ni.$	Baily, King, Merkle, Meshii [3]
Cascade Formation and Cascade Collapse	Kr^+	$Ag.^{\dagger}$ $Au.$	Haga, Baily, King, Merkle and Meshi [8]
	Ar^+	$Cu_3Au.^{\dagger}$	Black, Jenkins, Kirk [7]
	Kr^+, Cu^+	$Cu_3Au.^{\dagger}$	
	Fe^+ Ni^+	$Fe.^{\dagger}$ $Ni.^{\dagger}$ $Cu.^{\dagger}$	Robertson, Kirk, King [9]
Defect in Interstitial Alloys	e^-	$Pt-C$	Régnier, Lam, Westmacott [11]
	e^-	$Ni-Be$	Régnier, Lam [12]
Quantitative evaluations of Solute	e^-	$Ni-Al$	Okamoto, Lam
Segregation Profiles in proton and e^- irradiations	e^-	$Ni-Ge$	Okamoto, Allen [14]
	e^-	$Ni-Si$	
	p^+	$Ni-Si$	
Defect Production Efficiencies at Elevated Temperatures	e^-, p^+	$Ni-Si$	Muroga, Okamoto, Wiedersich [13]

† Single crystals

Table 2. Studies of Ion Induced Amorphization, Ion Implantation, and Radiation Damage of Oxides and Glasses.

Study	System Ion Target	Workers
Ion Beam Mixing in Evaporated Thin Films	$Kr^+ \rightarrow Cu, Bi$ $Kr^+ \rightarrow Al, Au$	Okamoto, Averbach, Stritzker [15] Okamoto, Averbach *
Amorphization Processes	$Kr^+ \rightarrow Si.^{\dagger}$	Seidman, Okamoto *
Ion Damage in Semiconductors	$Ar^+ \rightarrow GaP.^{\dagger}$ $Ar^+ \rightarrow GaAs.^{\dagger}$ $Xe^+ \rightarrow Si.^{\dagger}$	Jenkins, Robertson, Kirk *
Damage Nuclear Waste Materials	$He^+ \rightarrow SiO_2$	DeNatale, Howitt *
Radiation Damage of Spinel	$He^+, Ar^+ \rightarrow MgAl_2O_4$	Mitchell, Lee, Jenkins*
Nitride Formation by Implantation	$N_2^+ \rightarrow Al$	Donlon, McCune, Mitchell [16]

*New work, unpublished
 $\dagger$ Single crystals

Fig. 1. Column of HVEM showing details of the internal skim cup and movable Faraday cup.

In-situ irradiation at low temperature
100 keV $\text{Ar}^+ \rightarrow \text{Cu}_3\text{Au}$

Fig. 2. Electron micrographed Cu_3Au foil irradiated with 100 keV Ar^+ ions $\lesssim 40\text{K}$.

- (a) Dark field image showing disordered zones at sites of all ion impacts.
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