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A STUDY OF DEFECTS PRODUCED IN TUNGSTEN BY 800-MEV PROTONS USING FIELD-ION MICROSCOPY*

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Defects produced in tungsten by 800 MeV proton bombardment have been studied on the atomic scale with a Field Ion Microscope. The material was subjected to a fluence, as measured by radiochemistry, of 10^{22} pm^{-2} (-0.1 displacements per atom) at a temperature of 300K. A vacancy concentration of 10^{-3} was observed (calculated thermal equilibrium vacancy concentration is -10^{-56} at a calculated temperature of 300K). No vacancies were observed in the unirradiated samples. Since vacancies are essentially immobile (D_v - 10^{-57} cm^2/sec) at the irradiation temperature used in this study, it is believed that the observed concentrations are those of the radiation produced vacancies that did not spontaneously recombine. The observed interstitial concentration was lower than the vacancy concentration consistent with a higher diffusion rate for interstitials.

Additionally, a depleted zone was observed consisting of approximately 300 vacancies. This "void" volume lies along a [121] pole and has an elongated shape. It is postulated that this damaged region was caused by a recoiling W atom after it had undergone an "internuclear cascade" after collision with an incident proton. This type of defect may be the nucleus for subsequent void growth when the irradiation is carried out in the void-growth temperature regime.

The irradiation was conducted at the Clinton P. Anderson Los Alamos Meson Physics Facility (LAMPF). The Field Ion Microscopy was performed with a microscope at LAMPF that is dedicated to studies of irradiated material. The study forms a baseline for other planned experiments at LAMPF which aim to provide further understanding of the atomic-scale damage caused by medium-energy proton beams. The application of medium-energy proton damage as a simulation for 14 MeV neutron damage, as is encountered in advanced energy systems such as in magnetically and inertially confined thermonuclear reactors, has been addressed elsewhere.

1. INTRODUCTION

Materials subjected to irradiation environments have been studied to predict their physical and morphological changes. Irradiation of metals results in the production of point defects and transmutation products. Subsequent diffusion of the radiation-produced defects generally leads to microstructural changes in a material with attendant mechanical and physical property changes.

The life of the first wall will be dependent on the generation and transport of point defects, implantation of gas ions, and cyclic fatigue resistance of the first wall and/or coatings, caused by radiation and the attendant thermal cyclic stress. The kinetics of microstructural evolution under irradiation requires the determination of the rate of transport of these defects to sinks such as void nuclei, dislocations and grain boundaries. This rate is in turn dependent on the rate of production and concentration of point defects.

The most important consideration for estimating microstructural evolution kinetics is the concentration of point defects (vacancies and interstitials) and the temperature. In a metal under irradiation, the concentration of point defects can change with time depending on the rate of production, absorption at sinks, and

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***Los Alamos National Laboratory, Los Alamos, New Mexico temperature of irradiation. The number of point defects generated is dependent upon the flux of particles, the interaction cross-section and the energy transferred through collisions. The total number possible is described by the displacements per atom (dpa). This number includes the number which recombine in the short period[1] ($<10^{-12}$ seconds) following an initial displacement cascade and/or thermal spike. The number that recombine is important in determining the number of point defects which remain and are available for diffusion. It has been theorized that as few as one percent of the point defects predicted by simple cascade theory actually result in observable radiation effects.[2]

The vacancies which are observed in this experiment are most likely those which did not spontaneously recombine following a cascade event. The other sinks for vacancies are effectively eliminated from contributing to the reduction in the number of vacancies because of the low diffusion rate of vacancies in tungsten at the temperature of irradiation.

The thermal vacancy concentration can be calculated from:[3]

$$C_v \cong \exp(-E^F/kT)$$

where C_v is the thermal equilibrium vacancy concentration, E^F is the energy of formation, k is Boltzmann's constant and T is the temperature in K. For tungsten, the value of E^F is approximately 3.3 eV. These values yield a thermal vacancy concentration of 10^{-56} at 300K. Given this low calculated thermal vacancy concentration, the vacancies observed are most likely the result of irradiation. The vacancy diffusion coefficient may be calculated from:[4]

$$D_v = D_{vo}\exp(-E^m/kT)$$

where D is the diffusion coefficient, D_{vo} is the pre-exponent in cm^2/sec and E^m is the activation energy for vacancy mobility in eV. The values for tungsten are:[3,5]

$$D_o = 3.5 \times 10^{-2}\ cm^2 sec^{-1}$$
$$E^m = 3.3\ eV$$
$$k = 8.6 \times 10^{-5}\ eV/K$$
$$T \sim 300K$$

At 300K, D_v is found to be $1.33 \times 10^{-57}\ cm^2 sec^{-1}$ and from $x = (4D_v t)^{0.5}$, where x is the diffusion distance, it is estimated that the vacancies will move 1 lattice spacing (3.165 Å) in 6×10^{33} years. This extremely small value for the diffusion distance indicates that vacancies are immobile and will not diffuse for the conditions of this experiment.

The equation which describes the concentration of vacancies due to irradiation is:(6)

$$\begin{aligned} C_v = {} & (1 - \varepsilon)P - \alpha C_i C_v - C_v D_v\{4\pi r_v N_v + Z_v[\rho_d^o \\ & + 2\pi(r_{il}N_{il} + r_{vl}N_{vl}]\} \\ & + 4\pi r_v N_v D_v C_v^o \exp\{(2\gamma/r_v - P_g)b^3/kT)\} \\ & + Z_v C_v D_v^o\{\rho_d^o + 2\pi[r_{il}N_{il}\exp(-(\gamma_{sf} + F_{el})b^2/kT) \\ & + r_{vl}N_{vl}\exp\{(\gamma_{sf} + F_{el})b^2/kT)]\}. \end{aligned}$$

This equation describes the sources of vacancies from irradiation and their possible sinks. The symbols' meanings are defined in Table 1. This equation can be simplified for the conditions of this experiment since D_v is very small and the fluence was low. The equation reduces to:

$$C_v = (1 - \varepsilon)P - \alpha C_i C_v.$$

It is the purpose of this study to evaluate data which yield the number of defects which do not spontaneously recombine after a cascade event. This study, therefore, is aimed at the basics of vacancy generation resulting from irradiation. Tungsten was chosen as the material for examination because of its ease of preparation for the Field Ion Microscope (FIM), low thermal vacancy concentration at the temperature of irradiation and low diffusion rate at a convenient irradiation temperature.

II. EQUIPMENT

The equipment use his study was a Field Ion Microscope. The Alamos, Clinton P. Anderson Meson Physics ity, (LAMPF), was the source of protons used i he irradiation.

2.1 Field Ion Microscope

The Field Ion Microscope was built at the Los Alamos National Laboratory for the study of radiation effects to materials. It is a stainless steel system consisting of two cold traps which use either liquid nitrogen or liquid helium as the cryogenic liquid. The outer cold trap pre-cools the imaging gas while the inner cold trap cools the tip. A micro channel plate (MCP) is incorporated into the system to reduce photographic exposure time and allow for controlled field-evaporation of the tip. The system is diagrammed in Figure 1.

TABLE 1

r_v	void radius
N_v	number of voids
D	diffusion coefficient
C	concentration
γ	surface energy
P_g	internal gas pressure in a void
b	Burgers vector
P	Frenkel pair production rate
ϵ	cascade efficiency for producing vacancy loops(ϵ << 1)
T	temperature
Z	bias factor
α	direct recombination coefficient
γ_{sf}	stacking fault energy
F_{el}	elastic strain energy of a curved dislocation
ρ_d^o	network dislocation line length
O	superscript for thermal equilibrium values of D and C
x,i,vl,il	subscripts for vacancy, interstitial and their loops respectively

2.2 Irradiation

The irradiation was conducted with the LAMPF beam of protons in Line D. The average intensity of the beam in this area is 2 μA. The calculated temperature of the specimens at this current is approximately 300K.

III. PROCEDURE

The experimental sequence is as follows: 1) irradiation of the annealed tungsten and companion aluminum wires; 2) imaging the irradiated tungsten wire and the unirradiated control in the FIM; 3) evaluating any induced damage; and 4) observing the transmutation product ^{22}Na by its decay in the aluminum sample, measuring an accurate fluence for the tungsten.

3.1 Specimen Irradiation

Thin wires (7.6 x 10^{-5} m diameter) of tungsten were vacuum annealed (residual gas pressure < 10^{-6} torr) for one hour by resistive heating to approximately 2300K prior to

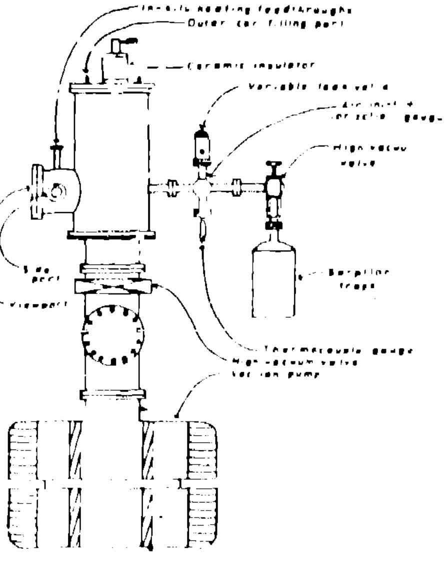

Figure 1
Schematic of FIM

irradiation. The wire was sectioned into 7.6 x 10^{-2} m lengths and mounted in a holder (Figure 2) for insertion into the proton beam for irradiation. Each tungsten sample was supplied with a companion aluminum sample to determine an accurately measured dose by determination of the amount of ^{22}Na produced in the aluminum.

3.2 Specimen Autoradiography and Radiochemistry

Following irradiation, the specimens were removed from the sample holder. They were placed on a photographic plate so that the radioactive transmutation products expose the film. The result is shown in Figure 3. This procedure determined the region of highest activity and therefore the highest fluence.

^{22}Na is a transmutation product which can occur by the spallation of a helium atom from aluminum. From the activity of this product and from the known cross-section for its production, the fluence can be determined. The displacements per atom (dpa) can then be calculated for the tungsten yielding an implied damage level in the tungsten. This process has been described elsewhere.(7)

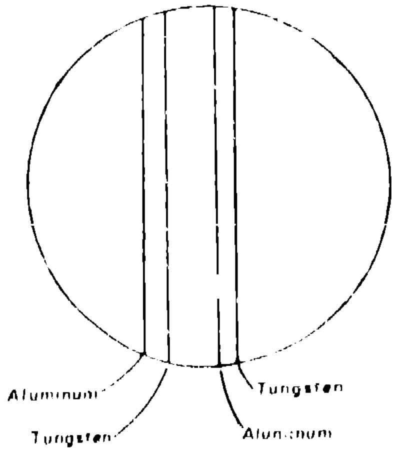

Figure 2
Schematic of irradiation arrangement

Figure 3
Autoradiography of W wire

3.3 Specimen Preparation

The tungsten wire was first sectioned at the center of the area of highest fluence as determined by autoradiography. A sample was cut from the center (0.5 cm length). This was attached with a silver paste onto a nichrome wire loop. The wire loop, in turn, was spot welded onto a 3.18 x 10^{-3} steel rod which could be inserted into the tip holder in the microscope. To achieve a suitable end form, the tungsten was electrochemically polished in a 1 N potassium hydroxide solution using 2 to 4 volts A.C. By repeated immersion of the sample into the solution, a sharp, conical end form could be achieved. The tip formed was visually checked at 160 X in an optical microscope to verify a suitably sharp end form.

3.4 Imaging

The tips were imaged in the FIM at liquid nitrogen (78K) temperature. Initial system vacuum was 10^{-8} torr. The imaging gas used, helium, was backfilled into the system to 10^{-5} torr.

The tip was conditioned by field evaporation in the FIM to achieve an atomically smooth, regular surface on the tip. This surface, and each subsequent {110} layer was photographed.

IV. RESULTS

4.1 Control

To verify that any defects observed were most likely the result of irradiation, several control samples of same tungsten were not irradiated. These were imaged in the FIM and their micrographs examined. In this manner, the initial, unirradiated vacancy concentration could be determined. The four most prominet {112} were examined for defects (vacancies and interstitials). Out of approximately 5×10^3 atomic sites examined, no point defects were observed in the unirradiated, control specimens.

4.2 Irradiated Specimens

Bombardment with 800 MeV protons caused damage to the tungsten and a low concentration of vacancies (approximately 10^{-3}). In addition, a void and interstitials were also observed.

The stable vacancy concentration in the irradiated material was found to be approximately 2.5×10^{-3}. The number of atomic sites examined was about 5×10^4. The displacements per atom (dpa) for the irradiated samples is calculated to be 1.13×10^{-1} as detailed below.

The void, Figure 4, consisted of approximately 300 vacant lattice sites. The void in three dimensions appeared to have an elongated pear shape.

V. DISCUSSION

Medium energy proton bombardment results in the production of vacancies and interstitials. Also, one void was observed.

This irradiation was conducted at a temperature (approximately 300K) at which the vacancies are essentially immobile. The calculated vacancy diffusion distance in tungsten at this temperature is approximately 10^{-57} cm^2/sec, as indicated in the introduction. It is therefore unlikely that a large number of the vacancies could diffuse to a sink. Only a few interstitials were observed indicating they are more mobile and diffuse at lower temperatures.

For a metal it is possible to provide an implied damage level in displacements per atom from a modified Kinchin and Pease equation:

$$dpa = 0.8\sigma_d/2E_d$$

where σ_d is a displacement energy cross section and E_d is the displacement energy of the metal atom. Multiplying this equation by the fluence gives the total dpa. Some constants are:[8]

$$\sigma_d = 1478 \text{ barns-keV}$$

$$E_{dw} = 47 \text{ eV}$$

which gives a dpa of 0.113 for the fluence in this experiment.

From the calculated dpa, approximately 2% to 3% of the displaced atoms as calculated from simple cascade theory contribute to the final vacancy concentration. These are the vacancies available for vacancy activated mechanisms, such as void growth and vacancy loop formation. The increase in vacancy concentration can therefore be approximated for this experiment by the rate dependent equation given below:

$$dC_v/dt = (1 - \varepsilon)P - \alpha C_i C_v$$

where C_i is the interstitial concentration, C_v is the vacancy concentration, α is the recombination coefficient and P is the production rate of vacancies. This implies that approximately 97% of the vacancies as predicted by the dpa calculations above recombine. The rate at which voids and loops grow under irradiation is proportional to the number of vacancies available to them, which corresponds to approximately 3% of the dpa.

The void observed is thought to be the result of either a primary proton or secondary particle interaction. Due to the small diffusion rate of the vacancies, it is likely that the void could

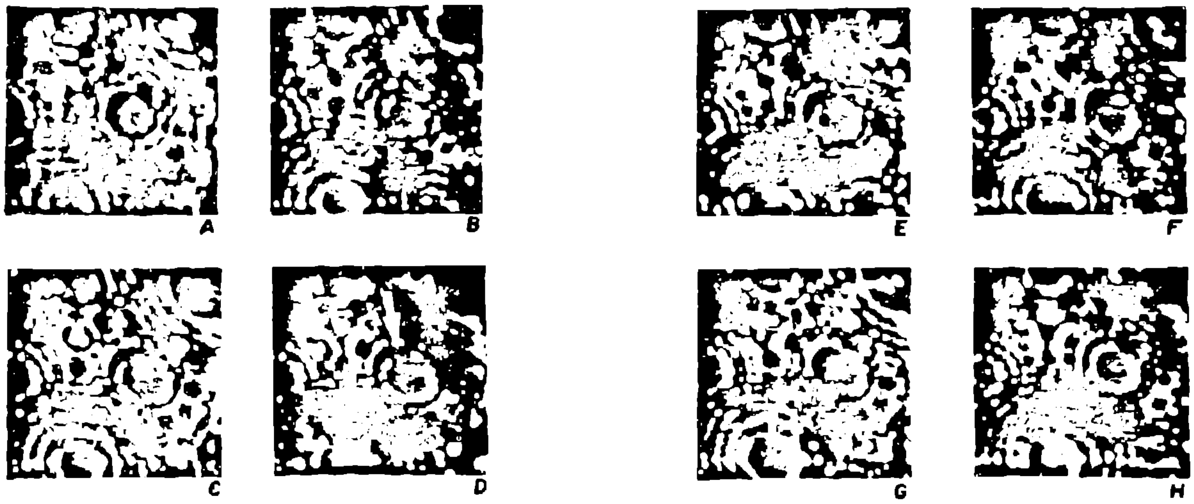

Figure 4

Void in tungsten, Figures A - H are each 1 - {110} layer apart

not have nucleated and grown to this size by vacancy condensation. The void is not spherical in shape but rather appears to have an elongated pear shape. It is postulated that such a void would provide an ideal nucleation site for void growth under conditions of temperature where vacancies are mobile, ($T > 0.3T_m$, T_m is the temperature at the melting point).

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