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Void Nucleation and Growth in Heavy
Ion and Electron Bombarded Pure Metals

PROGRESS REPORT

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The photo micrograph on the front cover is of a hardness indentation in aluminum. The picture was taken with a monochromatic light source with a wavelength of 5860 Å. The hole depth was determined to be 17600 ± 1500 Å by counting the number of interference fringes from the edge to the bottom of the indentation. The depth of the hole was monitored during the vibropolishing of an irradiated specimen to determine the amount of damaged material removed. Subsequent electrochemical thinning for transmission electron microscope allows us to observe the microstructure of the irradiated specimen at various depths below the bombarded surface. Knowledge of the precise distance from the surface allows us to calculate the amount of damage present at the point of observation. This particular sample is being prepared for 2 MeV Al^+ bombardment in which the damage zone is only ~25,000 Å thick.

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Preface

This document is not intended to be a typical quarterly report which has a wide external distribution and which only summarizes positive results. The main purpose of this report is for internal communication with the Nuclear Engineering, Materials Science and Physics Departments of the University of Wisconsin as well as to keep the sponsor of this work, namely the Metallurgy and Material Branch of the Division of Research, USAEC, informed of our progress. The reporting in this and subsequent reports may be quite detailed and may even be quite frank about some errors in our theoretical or experimental approach. However, it is felt that such a report is valuable from the standpoint of both the students and staff members because it will provide an accurate description of how we arrived at certain conclusions, provide historical perspective for new members of this study and lay the groundwork for more polished topical reports and open literature papers. It is therefore requested that the contents of this and other reports not be referenced without the consent of the authors.

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Summary

Experimental modification of the University of Wisconsin 2MV Van de Graaff Accelerator has been made to permit high current Al ion bombardment. A Physicon heavy ion source has been purchased and modified to produce Al^+ ions. The source is presently being tested outside the accelerator. Three irradiation chambers have been built to operate in the temperature ranges 50-110°C, 100-700°C, and 100-1100°C.

Modification to the University of Wisconsin 2 MV Electron Accelerator has been made to allow long term, high current electron bombardment. A sample chamber has been designed and constructed which will handle large heating loads, up to several hundred watts per cm^2 . The University of Wisconsin 1 MV transmission electron microscope, (an AEI, EM-7 model) has been delivered and is 90% assembled as of 1 June 1972. Present schedules call for acceptance testing to be completed by 1 September 1972. Sample preparation techniques for precise thinning of charged particles bombarded with Al have been developed.

Objective

The objective of this study is to clarify the important parameters for nucleation and growth of voids in pure aluminum and vanadium. The factors governing the production of voids during neutron irradiation of these metals are difficult to separate because both transmutation reactions and displacement spike cascades are superimposed on the pure displacement damage during irradiation. This study will use a high energy beam of Al^+ or V^+ ions to produce the necessary damage while eliminating the transmutation effects. A high energy beam of electrons will be used to eliminate both the transmutations and displacement spike effects. Selected doping of the samples with helium, hydrogen, Si, Cu, and Zn will help to understand the role of these impurities in nucleating voids. This study will concentrate on high purity V and samples of 99.9999% Al already extensively analyzed by ORNL scientists after neutron irradiation.

A. Introduction

1. Background for Present Study

The production of voids in pure metals and alloys during high temperature neutron irradiation has been one of the most serious problems facing designers of Liquid Metal Fast Breeder Reactors (LMFBR's)⁽¹⁻³⁾ and will be a formidable problem for the designers of Controlled Thermonuclear Reactors (CTR's).⁽⁴⁻⁷⁾ The swelling induced by these voids, and the stresses produced by swelling gradients, have required considerable redesign of reactor components and will have a large influence on the reactor configurations of the future. It is therefore imperative that satisfactory theoretical models be developed to correctly identify the factors that control the nucleation and growth of voids.

Several authors^(2,8-11) have formulated models for these phenomena and while most theories agree on the general features of void formation, they all have certain shortcomings. A very complete summary of the work in this area up to mid-1971 can be found in the proceedings of the International Conference on Voids in Metals which was held in Albany, New York, June 1971, and in the British Nuclear Energy Society Conference on "Voids Formed by Irradiation of Reactor Materials", Reading University, March 1971.

The first point of general agreement between workers in the field is that there must be a supersaturation of

vacancies in the metal before voids can be nucleated and subsequently grow. Secondly, the supersaturation of vacancies can only produce voids if the vacancies can move to nucleation sites. Hence, there is a minimum temperature required during irradiation to form voids. This minimum temperature is approximately 30% of the absolute melting point and is assumed to be associated with the vacancy migration temperature. There is also a maximum temperature above which the thermal production of vacancies reduces the supersaturation level below that required for void formation and growth. This upper temperature limit is approximately 50% of the melting point in most metals.

The third point of agreement is that since vacancies and interstitials are produced in equal numbers during irradiation, there must be some sinks which preferentially remove interstitial atoms during irradiation leaving an excess of vacancies in the lattice. The biased sinks are commonly assumed to be dislocations which are either present before irradiation or formed by the precipitation of interstitial atoms into dislocation loops.

The major disagreement in this field now centers around whether the voids are homogeneously or heterogeneously nucleated. The homogeneous theories are based on the assumption that voids are formed by the statistical agglomeration of vacancies or in the center of randomly distributed displacement spikes. The heterogeneous nucleation theories

assume that impurity atoms (either elements produced by transmutation reactions or residual gases such as oxygen, nitrogen, or hydrogen) are required to stabilize the void embryos regardless of how the latter are formed.

Verification of either of these concepts in the past has been hindered by the complex nature of the damage produced by neutrons. Transmutation reactions such as (n,p) or (n,α) reactions are inherent with high energy neutron bombardment as are the displacement spikes. Fortunately, there is now a way to isolate the transmutation and spike effects from pure displacement damage.

The use of high energy charged particle bombardment has proven to be an unexpected and rewarding tool for this type of study. (12-14) Ion bombardment experiments can be specifically tailored to give some insight into the void nucleation mechanisms. For example, unlike neutron irradiation studies, the presence or absence of inert gases in the metal is strictly up to the experimenter. Scientists also have much more control over the impurity level of the samples during ion bombardment because these experiments are conducted in a vacuum; such a test eliminates a large amount of the contamination inherent with current neutron irradiation programs. The high defect production rate and the possibility of varying it over several orders of magnitude can prove or disprove theories that are inherently defect production rate sensitive. Finally, by choosing the proper incident particle,

such as an electron, one can eliminate displacement spike effects and thereby determine their importance in void nucleation.

The objective of the present study is to use three types of charged particles, namely 2 MeV Al and V ions and 1-2 MeV electrons to study the nucleation and growth of voids in pure Al and V. The aluminum system will be studied initially and the work will be extended to V at a later date.

2. Previous Work

The use of high energy-heavy ion bombardment to stimulate high fluence, high temperature neutron damage in metals has been recently accepted and explored by the scientific community.⁽¹²⁻¹⁴⁾ This relatively new approach to the study of radiation damage has generated considerable theoretical interest in the effects of extremely high defect production rates. The most notable achievement of this technique is the "preview" of some microstructural defects and dimensional problems⁽¹⁵⁻¹⁷⁾ we may expect to find in metals subjected to 20 years or more of intense radiation damage in future LMFBR's or CTR's.

The potential advantages of using an external beam of energetic ions to accelerate the radiation damage rate by several orders of magnitude were first recognized in 1969 by Nelson and Mazey at Harwell.⁽¹⁸⁾ They found that they could produce in a matter of hours, a damage state in 316 stainless

steel which normally required more than a year of fast nuclear reactor irradiation to produce. The initial work was performed with low energy (100 KeV) heavy ions on stainless steel, but these same scientists later found that 20 MeV carbon ions could produce similar results in a much larger volume of sample. (12)

Two other approaches were immediately taken by scientists at Battelle-Northwest (BNW) and Atomic International (AI) Laboratories in the U.S. The BNW group concentrated on using higher energy (5-10 MeV) heavy ions, such as Cu, (13) Ni, (19-21) Se, (15,21) or Ta (17,20,22) to bombard stainless steel, Ni, Mo, Nb, Ta, and V. These studies confirmed the work of Nelson and Mazey and revealed some rather intriguing three dimensional defect structures in Ni, (15,17) Nb, (17,20) and Mo. (17) The AI group chose to pursue the use of 1.4 MeV protons to produce damage in stainless steel and nickel and found equally interesting results. (14) Other groups have now investigated the use of protons, (23) deuterons, (24) nitrogen ions (16) and high intensity beams of electrons (25,26) to simulate the high temperature damage produced by neutrons. The results of these latter experiments are currently appearing in the open literature and any extensive review of that work would be premature at this time.

It is now clear from the above experiments that 1) displacement spikes are not absolutely necessary for the formation of voids because high energy electron bombardment can produce voids in metals^(25,26) and 2) helium atoms are not absolutely essential for the nucleation of voids in metals or alloys.^(13, 15, 17,19-22) These observations will be studied in more detail with respect to damage rate, irradiation temperature, and total damage levels in order to better define the nucleation conditions.

3. Choice of Aluminum

There are five main reasons for choosing Al for this study apart from the fact that a large body of information exists on the physical and nuclear properties of this metal. First of all, Al can be obtained in a relatively pure form. Samples which contain less than 1 ppm of metallic impurities are readily available. Second, voids form in Al at relatively low temperatures, i.e. 55 C.⁽²⁾ This means that high purity starting materials will not be further contaminated by the irradiation environment (above any room temperature contamination already present) as occurs in most high temperature studies of voids in pure metals. Third, there is a considerable amount of data already available on the effects of neutron irradiation of Al.⁽²⁷⁻²⁹⁾ This information will provide an excellent background on which to compare the present results. Samples of the high

purity Al used by the ORNL workers have already been obtained and were used in an internally funded UW program concerned with 1 MeV proton bombardment.⁽³⁰⁾ Fourth, the nucleation of voids in Al has been shown to be extremely sensitive to impurity levels of H, He, Si, Mg, or Ni atoms.⁽²⁷⁻²⁹⁾ Unfortunately, neutron irradiation studies are not able to properly isolate these effects. Finally the displacement threshold for formation in Al has been established at the low level of ~ 0.01 dpa (displacements per atom).⁽²⁷⁾ This level of damage can easily be obtained in less than a minute of irradiation with the 2 MeV Al beam expected from the University of Wisconsin 2 MV Van de Graaff Accelerator (see Figure 1 for the damage distance curve for approximately one hour of 2 MeV Al ion bombardment). This damage level can also be reached in a few days of irradiation with the currently available beam (300 microamps per cm^2) of 2 MeV electrons and in less than a minute in the highly focussed beam (3 amps per cm^2) of 1 MeV electrons in the high voltage microscope.

In summary, it is clear that low temperature (50-100°C) bombardment of high purity Al (99.9999%) in a vacuum ($\sim 10^{-6}$ Torr) with 2 MeV Al ions and 1-2 MeV electrons will provide an extremely "clean" system for analysis. The data from this study should significantly contribute to the definition of critical parameters for void formation in this metal.

Figure 1

4. Choice of Vanadium

The high solubility of interstitial impurities in V during high temperature neutron irradiation is a further complicating feature to the theory of void formation in that metal. Current nuclear reactor environments in the 400-800°C temperature range required for void formation in V can induce a high amount of oxygen, nitrogen, or even carbon contamination. The effects of the residual gases on microscopic properties such as void formation are relatively unknown. Therefore, the main thrust of these studies will be to eliminate the contamination effect as well as to separate the spike and transmutation effects. Irradiation with 2 MeV vanadium ions in a vacuum of 10^{-6} to 10^{-7} Torr at 400°C will result in a minimal amount of interstitial impurity pickup.

Another reason for the choice of V for these studies is its technological importance in the nuclear field. Vanadium and V-Ti alloys have been considered for the LMFBR program as well as for future CTR service. Some data already exist which indicate the V-Ti Alloys may exhibit a special resistance to void formation⁽³¹⁻³⁴⁾ although the theoretical reasons for this resistance are not clearly understood. It is anticipated that as future reactor systems progress to higher power densities, the need to understand this phenomenon will increase.

Finally, V reacts in a chemical fashion similar to Nb, another primary CTR material candidate. It is quite possible that the factors which govern the nucleation and growth of voids in V subjected to realistic reactor environments will also apply to Nb under similar irradiations. In this way the interpretation of the data from this study may be applied in a broader sense to other bcc metals.

B. Progress in Experimental Programs

1. Aluminum Ion Bombardment

a. Irradiation System

The irradiation system presently consists of a 2 MeV electrostatic generator, a 15-degree electrostatic analyzer, a bending magnet, an ultra-high vacuum target chamber, and the necessary beam handling and vacuum equipment as shown in Figure 2. The combination of the electrostatic analyzer and bending magnet filters all but the desired bombarding ion, and allows precise selection of its energy and mass. The magnetic quadrupoles are necessary to defocus the beam in order to avoid regions of above-average density and consequent non-uniform fluence over the irradiated zone.

Several modifications have been made in order to carry out the aluminum-on-aluminum experiments: conditioning of the generator to allow operation at 2 MeV, replacement of the present Ortec RF ion source with a Physicon Model 910 heavy-ion source, and modification of the target assembly and the vacuum system.

b. Generator Conditioning

The generator has been run with and without beam for lengthy periods at successively higher voltages, in order to condition the accelerating tube to allow stable operation at 2 MV. This operation was accomplished after a thorough cleaning and overhaul of the generator's mechanical components, voltage

IRRADIATION FACILITY FOR 2 MeV ALUMINUM ION
BOMBARDMENT STUDIES

Figure 2

dividing and energy control systems. The reconditioned accelerator has been run at 2 MV for about two weeks during this reporting period.

c. Ion Source Testing and Installation

Conversion from the RF ion source to the heavy-ion source has required extensive rebuilding of the interior of the high-voltage terminal. The modifications include removal of the RF source and its power supplies, installation of a 2 KVa, 400 Hz, 3 phase alternator, additional telemetry and control rods, substantial power supplies for the source, as well as the source and a new lens to match it to the accelerating tube.

The source is presently being run on a low voltage test stand to work out the optimum conditions for the production of aluminum ions. When these conditions have been found, the source, power supplies and alternator will be installed in the accelerator, hopefully during the next quarter.

The source is an oscillating electron source and consists of a central filament in a graphite-quartz plasma chamber all contained within a solenoidal field. A schematic of the source is shown in Figure 3. A more detailed description will be given when the optimum operating parameters have been worked out. The material to be ionized may be introduced to the plasma chamber via three modes: 1) gaseous materials directly through an inlet tube, 2) high vapor pressure materials from a crucible adjacent to the plasma chamber, and 3) materials

SCHEMATIC OF
ALUMINUM ION
SOURCE

Figure 3

which form stable oxides or have low vapor pressures by means of a CCl_4 reaction. In the third method, which must be used for aluminum, a stream of CCl_4 vapor is passed over a sample of aluminum adjacent to and heated by the plasma chamber. According to Sidenius and Skilbreid,⁽³⁵⁾ the following reactions will take place:

with the volatile AlCl_3 and the COCl_2 and CCl_4 entering the plasma chamber. The source will then ionize these gases, producing a beam made up of Cl, O, C, and Al. All four components will reach the slits past the electrostatic analyzer, giving a strong energy control feedback signal, but the magnet will allow selection of only the aluminum ions for irradiation of the target.

The source has been operated with H_2 and argon gases modifications have been made to the power supplies and the lens system. A change from an orbitron pump to a diffusion pump on the test bench was made because the gas load from the source was higher than anticipated. The adsorbed gases on the graphite were particularly difficult to remove. The diffusion pump will be installed on the accelerator to be used in the range of pressures from 10^{-3} to 10^{-5} Torr, i.e. where the orbitrons have relatively low pumping speeds.

d. Target Chambers

There are three target chambers, which have been designed and built for this project. They allow the temperature of the sample to be precisely controlled over three temperature ranges: 50°C to 110°C with an aluminum target holder (Figure 4), 100°C to over 700°C with a stainless steel target holder, (Figure 5), and 100°C to over 1100°C with a molybdenum target holder which is of the same general design as that in Figure 5. All three sample chambers maintain electrical isolation of the sample itself thus allowing integration of beam current. Each holder precisely masks the irradiated area which permits an accurate measure of the total current density to be made.

The low-temperature sample holder is limited in its temperature range by the Cambion thermoelectric module which is used to heat or cool the sample from 50°C to 110°C. It should be possible to operate at temperatures as low as 50°C, as long as the total beam heating is less than a few watts.

Both the stainless steel and molybdenum target holders are heated by electron bombardment from a filament biased at a potential up to minus 8500 volts. The isolated sample is heated by radiation from the target holder and sample temperatures are measured by thermocouples attached to the sample masking plate. These target chambers provide up to two inches of motion to be transmitted through a bellows to the target holders, allowing four to six samples to be run with one loading. The target vacuum system is isolated from the accelerator

SCHEMATIC OF LOW TEMP (50°-100) SAMPLE CHAMBER
FOR HEAVY ION BOMBARDMENT STUDIES

Figure 4

SCHEMATIC OF MEDIUM (100-700°C) AND HIGH (100-1100°C) TEMPERATURE
SAMPLE CHAMBER FOR HEAVY ION BOMBORDMENT STUDIES

Figure 5

system by a 1/2 inch Granville-Phillips straight-through valve, allowing it to operate at pressures as low as 10^{-8} Torr when the accelerator pressure is at approximately 10^{-5} Torr. The use of an orbitron pump and all metal gaskets minimizes organic sample contamination.

The low temperature holder has been used extensively in preliminary proton-on-aluminum⁽³⁰⁾ void studies and the stainless-steel target chamber was operated up to 700°C for blistering studies on Nb. Both chambers operated quite satisfactorily. The higher temperature Mo chamber will be used mainly in the vanadium ion work to be pursued later in this program and the aluminum ion work will be conducted with the thermoelectric and stainless-steel systems.

2. Two Million Volt Electron Bombardment Studies

a. Brief Description of Accelerator

The belt-charged electrostatic generator has been so successful as a voltage source for positive particle acceleration that it has displaced all other static machines and most other types of direct-voltage generators. The use of high-pressure gas insulation has freed the generator from external disturbances and has resulted in a relatively compact design. Details such as the principles of operation, considerations of the electrical breakdown in compressed gases, the insulating column, the charge carrying belt, the accelerating tube, and problems such as the electric loading of the positive ion electrostatic generator have been extensively and thoroughly

described elsewhere. (36-39) There have been extensive debates between several schools of thought for many years over the advantages and disadvantages of various vacuum pumping schemes, horizontal versus vertical column mounting, and about which type of particle source is the best for a particular application. The problem of voltage calibration has been a continuing concern for very accurate nuclear work and beam diagnostics continue to be a challenge.

The electron accelerator differs from the positive-ion accelerator only in that the Electron gun terminal is charged to negative potential and a thermionic cathode replaces the ion source. However, since the cathode does not evolve gas, as do most positive-ion sources, the vacuum pumping problem is somewhat simpler and the accelerator tube can be of somewhat smaller diameter. These low voltage electrostatic electron accelerators have been widely used as X-ray sources for both radiography and therapy and to obtain an emergent electron beam through a thin metal vacuum window. This beam has been applied to such uses as the ionization of target materials and the electron beam sterilization and processing of a wide variety of packaged goods in addition to the study of photonuclear disintegration processes.

The electron accelerator being used in these studies was manufactured by the National Electrostatics Corp. of Middleton, Wisconsin. A general diagram of the irradiation facility is shown in Figure 6. It is capable of providing

ELECTRON ACCELERATOR AND TARGET SCHEMATIC

Figure 6

electron beam currents up to 300 μ A at energies up to 2 MeV. The present design will permit an increase in operating energy up to 3 MeV with the addition of another accelerator tube module.

This accelerator has been designed for ease of operation and long service with little attention. The main design improvements over previously available instruments include the modular construction in which two standard length 1 MV modules make up the support column and the insulating support posts which are identical and interchangeable. These posts are thermally matched to prevent damage from temperature shock, they are well protected from spark damage, and their 1 MV rating per module is thought to be very conservative.

The most unique feature of this machine is the design of the charge-carrying belt (Figure 7). In place of the traditional endless belt of insulating material such as insulating paper or rubberized fabric, the NEC accelerator uses a pellet chain which consists of metal charge carriers joined by insulating links. The charge carriers are charged and discharged by induction. Because the charge flow is more easily and dependably controlled, the voltage is inherently steadier than that provided by an insulating belt. The lint and dust problem has been minimized and the pellet chain should operate for long periods of time without attention. The maximum current per chain depends on the linear chain speed and for this installation it is about 150 μ A.

NEC PELLETRON — Charging Chain
Consists of Metal Cylinders joined
by links of insulating material.

Figure 7

Three tube sections are utilized for each 1 MV column module. They are all metal and ceramic, are bonded with a metal, and are bolted together utilizing metal gasket seals. The tube sections for all these standard NEC modules are identical and interchangeable. The normal pressure range in the column is from about 5×10^{-5} Torr to slightly below 1×10^{-8} Torr. Getter-ion pumping eliminates the danger of oil contamination of the vacuum system by accidents and the terminal power is furnished by an insulating shaft driving a 400 cycle generator in the terminal.

b. Design of Irradiation Chamber

The stainless steel target chamber for the electron irradiation of the thin metal foils is described in this section (Figure 8). The target chamber is connected to the end of the accelerator column at Flange A. The high vacuum side of the column is separated from the target chamber by a thin window of titanium that is approximately 0.001" in nominal thickness. This window will permit targets to be easily changed and irradiated at a variety of pressures from atmospheric down to reasonably good vacuums and there is also the possibility of introducing trace amounts of gases to study their effect on the formation of voids. This type of electron accelerator window has found wide acceptance lately and seems to provide a rugged long-lived vacuum barrier between the accelerator column and the target chamber. The window is water-cooled with a peripheral collant channel.

Figure 8

A water-cooled copper plate aperture with variable openings will aid in defining the beam on the target foil. This type of aperture is commonly used in electron accelerators for this purpose and has been found to be quite satisfactory.

The target holder is essentially a copper block with a small clamping arrangement to firmly attach the metal foil to the front face of the block. The target coolant will be water or freon depending on the cooling load and the critical part of the holder is the thickness of the copper just under the target foil. It must be as thin as possible in order for the heat transfer rate to be as great as possible and yet it must be thick enough for structural integrity and it must exceed the range of the most energetic electrons that will be used. Calculations show that a copper backing plate thickness of about 0.050" is a reasonable compromise in this case. The clamping arrangement insures that the target foil will at least be peripherally cooled and since only about 5% of the beam energy is deposited in the target foil and about 95% in the copper backing plate the critical cooling load is the prevention of the overheating of the backing plate.

Two approaches for maintaining the foil temperature at a certain limit have been considered. In the first case, several thermal wetting agents were considered in order to enhance the conductivity between the target foil and the backing plate. Mercury was eliminated because of its alloying tendencies with both copper and aluminum. Indium was unsatisfactory because its higher melting point meant that in order to

separate the target foil from the backing plate it would be necessary to heat the unit to an unacceptably high temperature. Gallium seemed to be the ideal metal for this purpose but extensive calculations showed that there could be a substantial diffusion of this metal into the aluminum target foil even at the low irradiation temperatures (about 100°C) that are initially planned. There is great uncertainty about the diffusion coefficient of Ga in Al especially since it may be substantially altered by the growth of voids in the aluminum. These considerations have led us to consider attempting the experiments, at least initially, without a thermally conducting film between the target foil and the copper backing plate. Should it become necessary to make this modification at a later time, gallium seems to be the most likely material to use. The second approach is to attempt the irradiation with no conducting film between the target foil and the backing plate. This method leads to uncertainties in the cooling calculations but the elimination of a possible gallium effect on the void formation process is a definite advantage.

Flange B in Figure 8 is the primary supporting structure for the target holder, aperture, thermocouple leads, and any additional signal carrier that may be necessary. The copper coolant tubes will be brought through the cap inside ceramic insulators and the seal made with a high performance silicon vacuum sealant that is used on high temperature vacuum furnaces. The interconnections are made from the copper coolant

channels to nylon tubing thus permitting an integration of the current that is being deposited in both the aperture and the target assembly. The total integrated current on target is important for it determines the theoretical dpa that has been introduced in the foil. The cap on Flange B is completely removable and it will permit rapid target and aperture changing without the necessity of altering the vacuum in the accelerator column.

Flange C is the connection to the target chamber vacuum system. This system is designed to permit a wide range of vacuums to be developed in the target chamber and, just as with the target chamber itself, becomes a semipermanent part of the business end of the electron accelerator. Only the cap on Flange B and its attached components are designed for easy and frequent removal. Experience in other laboratories seems to indicate that the titanium window should have a long life and would need to be changed only rarely. This window will be visually examined each time the target is removed and a safety cap will be placed on the end of the target chamber to prevent inadvertent damage to the window during those times when the target holder is out of the chamber.

Standard thermocouples (i.e. copper-constantan) will be used for temperature monitoring at all critical points and enough redundancy will be included to insure reliability over the fairly long irradiation times that are anticipated. Certain structural details such as centering and support

adjusters for the target holder are not shown. The target chamber has been designed with enough flexibility to permit long term irradiations of a variety of targets under a wide range of experimental conditions.

3. Status of EM-7, 1 MV Transmission Electron Microscope

The microscope has been purchased, delivered and ~90% assembled as of June 1, 1972. Minor construction and assembly problems have been encountered but it is expected that final acceptance testing will be completed by September 1. It is possible that the first samples will be irradiated in the microscope by October 1972.

The operating characteristics of the UW microscope are expected to be similar to those described by Makin⁽⁴⁰⁾ for existing EM-7 facilities. With proper modifications, the typical maximum current of 1 MV electrons that can be attained is ~30 Amperes per cm² (1.8×10^{20} electrons per cm² per second). This beam would produce a maximum displacement rate of $\sim 1 \times 10^{-2}$ dpa per second in Al. However, no such modifications are anticipated for the UW scope and under normal operation the maximum beam current is expected to be ~3 Amperes per cm⁻². This means that much of the work will be conducted at damage rates of $\sim 10^{-3}$ dpa sec⁻¹. The temperature rise, in the case of Al, is expected to be less than 1°C above the ambient foil temperature at that damage rate and the vacuum around the samples is anticipated to range from 5×10^{-6}

to 10^{-5} Torr. Although the maximum thickness Al sample that could be used for this work could be as much as ~ 5 microns⁽⁴⁰⁾, samples as thin as two microns or less will probably be used to obtain optimum imaging characteristics.

4. Sample Preparation Techniques for Transmission Electron Microscopy

The long range of electrons in a light element such as Al considerably simplifies the sample preparation procedures for TEM. For example, the range of a 1 MeV electron is ~ 1500 microns while that for a 2 MeV electron is ~ 3500 microns. Since our sample thickness is ~ 75 microns (0.003 inches), it is obvious that the damage will be uniform throughout the sample and it will not be critical to determine the exact position in the foil to calculate the dpa value. Such will not be the case in Al ion bombarded Al as we shall see in the next section.

The general procedure for thinning the samples after electron bombardment is as follows:

1. The irradiated foil (~ 0.5 to 1 cm^2 in area) mounted for vibropolishing in a Vibro Met Polisher.
2. Two other dummy samples are mounted symmetrically on the same specimen holder. This tripod arrangement has yielded the most uniform results because it tends to eliminate any rocking during the vibropolishing.

3. Spherical or pyramidal indents are made with a Leitz Wetzlar Durimet micro hardness tester at periodic intervals across the surface.
4. The depth of the indents are measured with a Hacker Interference Microscope, similar to the method used at ANL.⁽³⁴⁾
5. Vibropolishing with 0.3 micron Al_2O_3 in distilled water is continued until a prescribed amount of material is removed (1-10 microns). Typical vibropolishing rates are 1 micron per hour. The exact amount of material removed is calculated by measuring the depth of the hardness indents and subtracting it from the prepolished values.
6. A 3mm diameter sample is removed from the foil by electrolytic machining.
- *7. The front surface of the samples is masked with lacquer and the back of the sample is dimpled with a 20% perchloric acid and 80% ethyl alcohol solution at 20°C.
- *8. The final perforation is made in a 7% perchloric acid, 70% ethyl alcohol, 10% butyl cellulose and 13% distilled water solution at 0°C.
9. TEM observations will be made at 100 kV with Hitachi or AEI microscope. If necessary, TEM observations can be made with the AEI EM-7 microscope operation at reduced voltages.

* Alternative solutions are also being considered.

b. Aluminum Bombarded Samples

The samples from these irradiations will be prepared in much the same way as previously described with a few exceptions. The reduced range of 2 MeV Al ions, approximately $2\frac{1}{2}$ microns, requires close monitoring of bombarded surface removed. Therefore frequent monitoring of the vibropolish removal rate is performed to insure that an excessive amount of material is not removed. It may also be necessary to use a finer grade of polishing material and experiments with 0.05 micron diamond dust are underway.

C. Plans for Future Work

Modifications to the Al ion source should be completed in the next quarter and the source installed into the accelerator. Beam profile characteristics and stability will be examined and optimized. It is hoped that acceptable beam currents will be obtained by the end of August such that the irradiation program can begin.

The final modification to the 2 MeV electron accelerator and sample chamber will be completed in the next quarter and the beam characteristics defined and optimized. Methods of precise temperature control of the Al sample during bombardment will be established. The irradiation of the first Al sample at 100°C to ~0.01 dpa should be completed by the end of August.

The final acceptance of the EM-7 microscope should be completed by 1 August and it is expected that it will take 1-2 months of beam analysis and characterization before a complete irradiation program can be undertaken.

The doping of Al samples with He and Zn atoms will start in the next quarter. These samples will allow us to study the effects of impurities on the nucleation of voids in the September-February period.

References

1. E.R. Astley and L.M. Polentz, Power Engr., p. 49, July, 1970.
2. T.T. Claudson, J.L. Straalsund and H.R. Brager, "Radiation Damage in Reactor Materials," IAEA, Vienna, 1969, p. 165, Vol. 2.
3. P.R. Huebotter, T.R. Bump, W.T. Sha, D.T. Eggen and P.J. Fulford, ANL-7786, April 1971.
4. M.T. Robinson, "Nuclear Fusion Reactors, "UKAEA Culham Laboratory, Sept., 1969.
5. D.G. Martin, Ibid.
6. See Radiation Damage Section of the Proceedings of the International Working Sessions on Fusion Reactor Technology, June 28 - July 2, 1971, p. 137-222, Conf-7106024.
7. Fusion Reactor First Wall Materials, edited by L.C. Ianniello, WASH-1206, April, 1972.
8. R. Bullough and R.C. Perrin, "Irradiation Effects on Structural Alloys for Nuclear Reactor Applications," ASTM-STP-484, 1970, p. 317.
9. S.D. Harkness and C.Y. Li in "Radiation Damage in Reactor Materials," IAEA, Vienna, 1969.
10. K.R. Russell to be published.
11. H.W. Wiedersich, in Second International Conference on the Strength of Metals and Alloys, Pacific Grove, California, ASM, 1970, p. 784.
12. R.S. Nelson, D.J. Mazey and J.A. Hudson, J. Nuc. Mat., 37, 1, 1970.
13. G.L. Kulcinski, D.G. Doran and J.J. Laidler, Radiation Effects, 7, 195, 1970.
14. D.W. Keefer, H.H. Neely, J.C. Robinson, A.G. Pard, and D. Kramer, "Irradiation Effects on Structural Alloys for Nuclear Reactor Applications," ASTM-STP, 484, 1970, p. 332.
15. G.L. Kulcinski, J.L. Brimhall, and H.E. Kissinger, J. Nuc. Mat., 40, 166, 1971.
16. J.H. Evans, Nature, 229, 403, 1971.

17. G.L. Kulcinski, and J.L. Brimhall, "Ordered Defect Structures in Irradiated Metals" to be published in an ASTM Special Technical Publication From the June 1972 meeting at Los Angeles, California.
18. R.S. Nelson and D.J. Mazey, "Radiation Damage in Reactor Materials, IAEA Vienna, 1969, p. 157.
19. G.L. Kulcinski, J.J. Laidler and H.R. Brager, "Irradiation Effects on Structural Alloys for Nuclear Reactor Applications," ASTM-STP-484, 1970, p. 405.
20. G.L. Kulcinski, J.L. Brimhall, and H.E. Kissinger, p. 449, Proceedings of International Conference on Voids in Metals, Albany, N.Y., June 1971. AEC Symp. Series 26, April 1972.
21. G.L. Kulcinski, A.B. Wittkower, and G. Ryding, Nuclear Instruments and Methods, Vol. 94, 1971, p. 365.
22. G.L. Kulcinski and J.L. Brimhall, International Conference on Defects in Refractory Metal, M $\ddot{o}$ l, Belgium, September, 1971.
23. J. Donhowe, unpublished data, University of Wisconsin.
24. S.D. Harkness and F.L. Yaggee, ANL-766, 1970, p. 44.
25. D.I.R. Norris, Phil. Mag., 23, 135, 1971.
26. K. Urban, Phys. Stat. Sol., (a) 3, K167, 1970.
27. N.H. Packen, J. Nuc. Mat., 40, 1, 1971.
28. A. Jostsons, E.L. Long, J.O. Stiegler, K. Farrell, and D.N. Braski, ORNL-TM-3494, 1971.
29. K. Farrell, J.T. Houston, A. Wolfenden, R.T. King, and A. Jostsons, ORNL-TM-3493, 1971.
30. J.M. Donhowe, unpublished data.
31. F.W. Wiffen and J.O. Stiegler, J. Metals, Vol. 20, P. 117A, 1968, also see ORNL-440, 1969.
32. F.W. Wiffen, To be Published in the Proceedings of the International Conference on Voids in Metals held in Albany, N.Y., June 1971. AEC Symp. Series 26, June, 1972.
33. J.D. Elen, 7th Int. Congress on Electron Microscopy, 1970, Grenoble, Vo. 2, P. 351.
34. A. Taylor, A.T. Santhanam and B. Loomis, "Fast Neutron and Ion-Induced Swelling of Niobium and Vanadium Alloys", presented at the 4th annual Spring Meeting of AIME, Boston, May 8-11, 1972.

35. G. Sidenium and O. Skilbreed, E.M. Separation of Radioactive Isotopes, Vienna, 1961.
36. R.G. Herb, C.M. Turner, C.M. Hudson, and R.E. Warren, Phys. Rev., 58, (1940) 579.
37. R.J. Van de Graaff, J.G. Trump, and W.W. Buechner, Repts. Progr. in Phys., 11 (1948) 1.
38. R.J. Van de Graaff, Nuclear Instr. and Methods, 8 (1960) 195.
39. M.S. Livingston and J.P. Blewett, Particle Accelerators, McGraw-Hill (New York, 1962).
40. J. Makin, AERE-R6957, November 1971.